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PROCEEDINGS OF THE 13TH ESARDA ANNUAL SYMPONIUM ON SAFEGUARDS AND NUCLEAR MATERIAL MANAGEMENT

L. STANCHI, EDITOR

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FOREWORD

The 13th ESARDA Symposium, held in the magnificent setting of the Palace of the Popes in Avignon, France is characterized by two main features.

One is the very high number of papers in total and more specifically in the field of measurements. This means that there is great interest in this field again, both in Nondestructive Assay and in Destructive Analysis. The total number of papers accepted in the Symposium was: 60 included in oral sessions and 66 presented as posters.

The second important fact is the considerable participation of Eastern European Countries. There were several papers and 6 participants from the USSR, with appreciated contributions. We also had papers and participants from Czechoslovakia, Hungary and Poland, countries which are candidates to join an important European initiative i.e. COST (Scientific and Technical Co-operation) which has the aim of promoting research in Europe on a transnational basis.

I wish to thank all the authors who, in the era of word processors, were obliged to use scissors and glue to arrange their papers in a two column format. Nevertheless several authors will discover that their papers are a little modified in their making-up.

Finally I am very pleased to note that some authors presented results obtained the week before the conference. The ESARDA Symposium is in this respect very alive and the proceedings, which will appear promptly, are therefore well updated. The presentation of the manuscripts only at the conference time meant that photocopies could not be made beforehand, which would have been useful, especially for the interpreters, but I am convinced that the game is worth the candle.

L. Stanchi

ESARDA

ESARDA is an association of European organizations formed to advance and harmonize research and development of safeguards. It also provides a forum for the exchange of information and ideas between nuclear facility operators and safeguarding authorities.

Its partners as of 1st January 1991 were:

- The European Atomic Energy Community
- The Kernforschungszentrum Karlsruhe (KfK) - F.R. Germany
- The Centre d'Etude de l'Energie Nucléaire - Studiecentrum voor Kernenergie (CEN/SCK) - Belgium
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Secretary: ***C. Foggi*** *(CEC, JRC-Ispra)*

Closing Summing up of the Chairman of the Symposium 733

ADDENDUM 3.1 Gamma and X-Ray Measurement of Nuclear Material Concentration and Quantities
T. Dragnev, F. Grinevich, V. Rukhlo, D. Rundquist, J. Safar, E. Szabo . 739

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This list needs to be updated but we judged unreasonable to delay the circulation of the proceedings for such secondary matter.
The updated list of participants will appear in the ESARDA Bulletin No. 20.
I wish to apologize for this inconvenience both with the readers and (especially) with the participants omitted or ill treated because of incomplete or erroneous address.

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ESARDA 13th Annual Symposium

Opening address by the Chairman of ESARDA

B H Patrick (UKAEA, Harwell)

Ladies and gentlemen

It is my privilege and pleasure, on behalf of the ESARDA organisation, to welcome you to the 13th Annual Symposium on Safeguards and Nuclear Materials Management. In recent years, the pattern of the annual ESARDA meetings has been alternate internal meetings with a specific theme, restricted to ESARDA members and Working Group observers, and open symposia, such as the present one. The latter type provides an opportunity for many representatives from countries around the world to interact with ESARDA participants and thereby increase the international dimension which is such an important part of safeguards. I shall say more on this point later.

Our hosts this year are The Comissariat à l'Energie Atomique of France. We are very much indebted to our French colleagues for arranging for the meeting to be held in this superb location, Le Palais des Papes, so full of historical significance. I hope you all have an opportunity during the week to visit other parts of the Palace and to absorb the atmosphere. I am sure we shall all let our minds drift back several centuries at times during the next few days, imagining what it was like when this Palace was a powerful influence in the land. Princes, ambassadors, cardinals arriving with their entourages to have an audience with the Pope; luxurious furnishings, extravagant decorations, beautiful paintings must have contributed to the magnificence of it all. I shall resist trying to make any comparison with our meeting this week, in order not to get into deep water. But I hope that, in letting your minds wander, you will not do it during the sessions as there is much of interest in the programme!

It is very encouraging to see such a good turnout. ESARDA itself is nothing; it is the people who contribute to its activities who give it its strength. Without them there would be no harmonisation of R&D in safeguards in Europe and no useful forum for discussion between developers and plant operators. Your input is a vital ingredient in ensuring the success of the Association. This year, as in the past, we shall have presentations from all the ESARDA Partners and from further afield. But it is particularly gratifying to see the interest from a number of countries in Eastern Europe. I should like to extend a particular welcome to the representatives from those areas and say that we look forward greatly to their contributions and to interacting with them.

The Symposium is organised along the usual lines, with sessions devoted to the topics which we all recognise as categories of prime importance in safeguards. Presentations take two forms, oral and poster. In the latter case, specific times are set aside when authors should be available alongside their boards and I commend these times to you as I know from experience that a great deal can be gained from one-to-one discussions.

Posters, once set up, will be on display for a whole day to give maximum viewing time and I am sure authors will be delighted if you seek them out with specific questions, if they are not there when you look at their presentations.

There is one new feature in this year's Symposium - a review talk. For some time, I have been concerned at the lack of reviews of important aspects of safeguards. In most scientific disciplines that I am aware of, reviews appear in journals and at major conferences but this does not seem to be the case for safeguards. It is true that some review-type articles appear in the ESARDA Bulletin, the Journal of the INMM and occasionally in the IAEA Bulletin, but there has been a noticeable lack of review presentations at the ESARDA Symposia. This makes it very difficult for a newcomer or someone wanting to know the state of the art and practice in a particular area without having to plough through individual contributions to a topic contained in the proceedings of past conferences. In an attempt to begin to bridge this gap, the UK commissioned a review of passive neutron coincidence counting by Dr B W Hooton, a former Chairman of ESARDA. He will give a talk on this subject in Session 5B. As time will not allow him to cover the topic in depth, we intend to publish a detailed report in a few months' time. We hope this will be a useful contribution to the safeguards community and that this initiative will stimulate others to follow suit by tackling other important safeguards areas.

I look forward to an interesting meeting. The programme looks to be an exciting and interesting one, and I am sure that, in addition to the knowledge gained from the presentations, many useful contacts will be made outside the formal sessions. Safeguards, although now a fairly mature subject, is nevertheless facing many challenges and problems. The ESARDA analysis in 1988 of the nuclear fuel cycle up to the year 2000 highlighted the increasing use of MOX in thermal reactor systems and the problems which that would bring. The financial pressures on the industry focus attention on the need to increase the efficiency of inspection activities to reduce the unit cost of safeguards. Several large scale reprocessing plants are expected to be commissioned in this decade and it is important to show that credible and effective safeguards can be applied. During this meeting, we can expect to find the issues arising from these and other current topics being addressed and I believe we shall find that practical solutions can be found which will enable the nuclear industry to advance in the confident knowledge that safeguards measures will be found to give the necessary assurance.

I hope you all have a very enjoyable stay in Avignon.

LE CONTROLE DES MATIERES NUCLEAIRES
EN FRANCE

IMPLEMENTATION OF DOMESTIC AND INTERNATIONAL SAFEGUARDS EN FRANCE

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ABSTRACT

As one of the most important industrial electronuclear states in the world, with a comprehensive national fuel cycle (from mines to reprocessing plants) on the one hand, and as a nuclear supplier as well as a nuclear weapon state on the other hand, France deemed it necessary to establish its own domestic safeguards system, in order to struggle against hazards of malevolence at the national level and to contribute efficiently to nuclear non-proloferation at the international level. The french legal regulations are based on the main following principes : 1) Licensing of activities related to import-export, to the possession or to the transportation of nuclear material ; 2) Control performed by the operator, including NMCA, C/S an physical protection ; 3) Inspection by the domestic Safeguards Authority ; 4) Penalties. Specifities of domestic and international Safeguards (Euratom, IAEA) concerning objectives and scopes, information required, accounting, inspection types, R & D are illustrated and possible interactions are considered.

1 - INTRODUCTION

Second pays après les Etats-Unis, avec un parc électronucléaire de 56 tranches installées représentant 56 GWe nets, la France se classe au premier rang mondial en ce qui concerne la part du nucléaire (75 %) dans le bilan électrique national. Elle a été ainsi amenée à se doter d'un cycle du combustible complet allant de la mine au retraitement des combustibles irradiés, utilisé à la fois pour ses besoins nationaux et pour la fourniture de services aux autres pays. Au niveau gouvernemental, la sécurité nucléaire relève de chacun des ministres compétents.

Pour assurer la cohérence des actions entreprise, il a été mis en place, sous l'autorité du Premier Ministre, le Comité Interministériel de la Sécurité Nucléaire, (CISN) présidé par le Premier Ministre. Ce comité se réunit de façon périodique ; le suivi permanent des affaires est assuré par le Secrétariat général du Comité de la Sécurité Nucléaire (SGCISN). La France dispose par ailleurs d'installations nucléaires d'une part pour ses programmes de recherche et de développement et d'autre part en tant qu'Etat doté de l'arme nucléaire pour les programmes relatifs aux applications militaires. Elle est enfin l'un des principaux pays industrialisés fournisseurs de matières, d'équipements, de technologie et de services nucléaires.

Compte tenu de cette situation et conscient de ses responsabilités particulières, tant au plan national pour la sécurité du public qu'au plan international dans le domaine de la non-prolifération nucléaire, le gouvernement français a mis en place un dispositif de contrôle national placé sous l'autorité du Haut Fonctionnaire de Défense du ministère chargé de l'industrie pour :

- assurer la protection et le contrôle des matières nucléaires (vol ou détournement).

- assurer la protection des installations contre des actes de malveillance.

Si d'autres pays au monde (Etats-Unis, Japon, Suède par exemple) ont un système national de contrôle, il convient de mentionner la situation originale de la France au sein de la Communauté européenne, puisque aucun autre Etat membre n'a instauré un système national de comptabilité et de contrôle des matières nucléaires indépendant, de telle sorte qu'une partie des prérogatives est déléguée à EURATOM. Ces Etats membres conservent par contre, comme la France, leurs compétences nationales exclusives pour toutes les questions relatives à la protection physique et au transport des matières nucléaires.

Seul est abordé dans cet exposé le contrôle des matières nucléaires. C'est en effet l'unique domaine dans lequel d'éventuelles interactions peuvent être envisagées entre le contrôle national et l'application des contrôles de la Direction du Contrôle de Sécurité d'EURATOM (DCS) et de l'Agence Internationale de l'Energie Atomique (AIEA) en France, pour la vérification du respect des engagements pris par le gouvernement français en matière de non-prolifération des armes nucléaires et autres engagements d'utilisation à des fins pacifiques.
Cependant, comme précisé plus loin, le domaine de la protection physique des matières ne sera pas totalement exclu de cette communication.

2 - LE CONTROLE FRANCAIS DES MATIERES NUCLEAIRES

La loi du 25 juillet 1980, relative à la protection et au contrôle des matières nucléaires, et les textes d'application définissent les principes et les dispositions générales devant être mises en oeuvre pour prévenir et le cas échéant détecter sans délai toutes disparitions, pertes, vols ou détournements de matières nucléaires. Ces principes et dispositions générales s'appliquent à l'ensemble des matières nucléaires, qu'elles soient utilisées dans des activités entièrement civiles ou destinées aux besoins de la défense nationale.

Selon la nature et la quantité de matières, les détenteurs sont soumis à un régime d'autorisation, de déclaration annuelle ou d'exemption (voir tableau annexé).

La volonté du législateur apparaît dans les maîtres mots "autorisation", "contrôle", "sanctions".

a) Autorisation :

Une autorisation préalable, délivrée par le ministre chargé de l'industrie, est requise pour quiconque veut exercer des activités d'importation, d'exportation, de détention, de transfert, d'utilisation ou de transport de matières nucléaires. Les matières concernées sont le plutonium, l'uranium, le thorium, le deutérium, le tritium et le lithium enrichi en isotope 6, ou les composants contenant ces éléments, quelle que soit leur forme physique ou chimique, à l'exception des minerais.

A l'appui de sa demande, l'exploitant ou le transporteur fournit un dossier décrivant les mesures qu'il compte mettre en oeuvre pour assurer la protection et le contrôle des matières. Ce dossier peut éventuellement faire référence à des documents internes (procédures, consignes, etc...)

Les pouvoirs publics, avec l'aide de leur appui technique, procèdent à l'analyse de ce dossier afin de déterminer si les mesures présentées correspondent bien aux exigences réglementaires, en particulier pour ce qui concerne les niveaux de protection.

b) Contrôle :

Il s'exerce à deux niveaux :

- En premier lieu par l'exploitant qui doit assurer le contrôle des matières nucléaires par des mesures appropriées et complémentaires de suivi et de comptabilité, de confinement, de surveillance et de protection physique.

Les dispositions de suivi et de comptabilité ont pour objectif de connaître en permanence les quantités, la localisation et les mouvements et de détecter, puis d'infirmer ou de confirmer, toute anomalie concernant les quantité, qualité, localisation ou emploi des matières nucléaires, en utilisant toutes les mesures disponibles, y compris les données d'exploitation.

Les mesures de confinement doivent prévenir les mouvements de matières non autorisés ou non justifiés. Les mesures de surveillance doivent garantir l'intégrité du confinement, vérifier l'absence de sortie de matière par des voies anormales, ainsi que l'absence de fraude sur les appareils ou équipements utilisés pour le suivi, la comptabilité, le confinement et la surveillance, y compris pour la transmission et le traitement des données.

Enfin, les mesures de protection physique des matières, des locaux et des installations ont pour but de les protéger contre les actes de malveillance à l'aide de systèmes de protection en profondeur, un contrôle des personnes et une intervention éventuelle des forces de l'ordre. Les matières nucléaires en cours de transport font l'objet de mesures de protection destinées à prévenir tout vol ou détournement. Les principes de la protection physique, bien que plus rigoureux en ce qui concerne les seuils et certaines matières (uranium naturel et appauvri, tritium, lithium 6), sont conformes à ceux énoncés par les directives de Londres auxquelles la France est partie (INCIRC/254) et par la convention internationale sur la protection physique des matières nucléaires (INFCIRC/274) que la France a signé en 1980 et dont le parlement français a autorisé l'approbation en juin 1989. S'agissant des mesures spécifiques de chaque installation ou des transports, la confidentialité étant la condition essentielle de leur efficacité, elles ne doivent être connues que des personnes autorisées par l'exploitant ou le transporteur, en accord avec les pouvoirs publics.

En second lieu, le contrôle est exercé par le Service de Protection et de Contrôle des Matières Nucléaires (SPCMN) placé sous l'autorité du Haut Fonctionnaire de Défense du ministère chargé de l'industrie. L'autorité nationale fait appel en appui technique, aux moyens appartenant à l'Institut de Protection et de Sûreté Nucléaire du CEA (Département de Sécurité des Matières Radioactives). Ce contrôle de l'application de la réglementation et de la doctrine comporte en particulier :

- l'analyse de la protection et du contrôle des matières nucléaires (dossier d'autorisation, modifications des installations, respect des engagements pris par l'exploitant et des prescriptions de l'administration),

- l'instruction approfondie des études de sécurité demandées à certains exploitants pour évaluer leur système de protection des matières nucléaires et déterminer les actions possibles sur la base d'analyses à caractère technique,

- les inspections effectuées par des agents habilités par les autorités de l'Etat et assermentés. Ces inspections tiennent compte notamment de la sensibilité des matières nucléaires et des conclusions des inspections précédentes. Elles sont en principe notifiées aux exploitants ; toutefois des contrôles inopinés peuvent être effectués pour vérifier la mise en oeuvre courante des dispositions particulières demandées par les pouvoirs publics à certains exploitants, ou décidées sur le champ à la suite d'incidents ou d'événements exigeant que des mesures, même transitoires, soient mises en oeuvre et que des actions correctives soient engagées sans délai.

Aussi bien pour le contrôle effectué par l'exploitant que pour le contrôle par les pouvoirs publics, un accent particulier est mis sur l'importance de la mise en place des principes de l'assurance et de l'organisation de la qualité, ou de principes équivalents.

c) Sanctions

Certains agissements portant sur les matières nucléaires constituent des délits assortis de sanctions pénales parfois très lourdes (pouvant atteindre 10 ans d'emprisonnement). Parmi les infractions figurent en particulier l'appropriation indue de matières nucléaires, l'exercice sans autorisation des activités visées par la loi, l'obstacle à l'exercice du contrôle par les pouvoirs publics, le défaut de déclaration de perte, vol, disparition ou détournement, la détention, le transfert, l'utilisation ou le transport hors du territoire de la République Française de matières visées par la convention internationale sur la protection physique des matières nucléaires, sans autorisation des autorités étrangères compétentes.

3 - COMPARAISON DU CONTROLE NATIONAL FRANCAIS ET DES CONTROLES INTERNATIONAUX ET INTERACTIONS EVENTUELLES

3.1. - Aspects juridiques :

Sur le plan juridique, le principe selon lequel les accords internationaux priment les règlements nationaux ; aussi le contrôle national tient compte des engagements internationaux du gouvernement français en matière d'utilisation pacifique et non explosive des matières nucléaires. Ainsi, si la loi de 1980 et les textes d'application tenaient compte en particulier de notre adhésion au Traité d'EURATOM et aux directives de Londres, ainsi que de notre offre volontaire de soumission aux garanties de l'AIEA (INFCIRC/290), la ratification par le Parlement de la convention internationale sur la protection physique des matières nucléaires a conduit à la modification de cette loi et du code de procédure pénale.

3.2. - Objectifs et champ d'application:

Une différence politique fondamentale existe sur le plan des objectifs. Alors que le contrôle national vise le terrorisme national ou international mettant principalement l'accent sur le vol de matières nucléaires par une personne ou un groupe de personnes habilitées à travailler normalement dans une installation ou un groupe d'agresseurs externes, les contrôles internationaux ont pour objectif d'assurer la communauté internationale que les Etats honorent les engagements qu'ils ont pris en matière de non-prolifération et d'utilisation pacifique. Il en résulte une différence essentielle sur le plan de la responsabilité. Pour le contrôle national, celle-ci repose en premier lieu sur l'exploitant considéré comme un acteur important de la sécurité nationale ; elle est complémentaire du contrôle exercé par les pouvoirs publics qui vise à vérifier les conditions dans lesquelles il applique la réglementation. Au contraire pour les contrôles internationaux la responsabilité est assumée par l'Etat qui doit pouvoir rendre compte lui-même du respect de ses engagements.

Par ailleurs le champ d'application du contrôle national est plus large que celui des contrôles internationaux :

- pour les matières nucléaires, puisqu'il concerne en plus des matières brutes et des matières fissiles spéciales, le deutérium sous toutes ses formes (alors que les accords conclus par la Communauté avec le Canada et les Etats-Unis sont restreints à l'eau lourde), le tritium et le lithium enrichi en lithium 6, principalement utilisés dans les programmes de production d'armes nucléaires,

- pour les installations concernées, puisque le contrôle national vise non seulement les matières nucléaires civiles, mais aussi les matières nucléaires utilisées ou susceptibles de l'être pour les besoins de la défense nationale,

- pour ce qui concerne l'étape du cycle du combustible, à partir de laquelle commencent ou cessent les contrôles : les minerais sont exclus du champ d'application de la loi, mais sont soumis aux contrôles d'EURATOM, alors que les contrôles de l'AIEA commencent seulement au stade de la matière nucléaire propre à la fabrication du combustible ou à l'enrichissement, (métal, oxyde ou fluorure) .

- pour ce qui concerne le contrôle des matières nucléaires contenues dans les déchets, le contrôle national ne nécessite plus de dispositions spécifiques à l'égard des actes de malveillance à partir du moment où les déchets ont été conditionnés et stockés sur un site de stockage de l'ANDRA ; les règles de sûreté sont alors considérées comme satisfaisantes pour les objectifs de la sécurité. Pour l'AIEA, il est prévu que les contrôles cessent lorsque l'Agence a constaté que les matières nucléaires contenues dans les déchets ne sont plus utilisables pour aucune activité nucléaire ou sont pratiquement irrécupérables. Les contrôles internationaux cessent en principe de s'appliquer après que les vérifications aient eu lieu préalablement au conditionnement. Il paraîtrait déraisonnable en tout état de cause que pour des motifs de non-prolifération de tels contrôles s'appliquent aux sites de stockage définitif de déchets non utilisables ou irrécupérables.

3.3. - Dossier à fournir par l'exploitant et dispositions appliquées :

Sur le plan national, l'exploitant adresse au ministère chargé de l'industrie un dossier de demande d'autorisation et de contrôle de l'installation, avec en particulier un chapitre suivi et comptabilité rédigé selon un plan type. Pour EURATOM, les "déclarations des caractéristiques techniques fondamentales" et pour l'AIEA le "questionnaire sur les renseignements descriptifs", bien que différents dans la forme et le détail du contenu, présentent certains points communs pour le chapitre décrivant les dispositions et mesures de suivi et de comptabilité mises en oeuvre par l'exploitant.
Pour ce qui concerne les dispositions nationales, une autorisation de détention, de transport, d'utilisation, d'élaboration ou d'import-export, est délivrée par le ministère de l'industrie sous forme d'un arrêté fixant les conditions et les limites d'exploitation. A cet arrêté est joint un état récapitulatif dressé par le SPCMN, qui recense l'ensemble des correspondances et des dossiers qui lui sont adressés par le titulaire de l'autorisation. Ainsi ce document témoigne-t-il des engagements de ce titulaire envers les pouvoirs publics et acceptés par eux pour assurer la protection et le contrôle des matières nucléaires. Cette autorisation ne dispense évidemment pas des obligations du chapitre VII du Traité d'EURATOM.

3.4. - Comptabilité :

Des différences entre le système national et les systèmes internationaux existent pour :

- les matières nucléaires, plus nombreuses dans le système national, comme déjà indiqué,

- les découpages en ensembles techniques au plan national et les zones de bilan matières pour EURATOM et l'AIEA qui, répondant à des objectifs différents, ne sont pas nécessairement identiques, bien qu'il y ait toujours une cohérence parfaite entre un ou plusieurs ensembles et une zone de bilan matières,

- les délais de transmission des données comptables, le jour même pour le contrôle national, le mois pour les contrôles internationaux,

- les renseignements à transmettre, les descriptions détaillées des matières, tels que ceux fournis au contrôle national, ne concernent pas les contrôles internationaux, alors qu'au contraire les codes d'engagement particulier demandés par EURATOM ne sont pas de la responsabilité directe du contrôle national.

Dans le domaine de la comptabilité, l'impératif est de ne pas obliger l'exploitant à faire des déclarations distinctes, en élaborant un modèle de rapport comptable satisfaisant aux obligations des deux systèmes. Le système national de comptabilité centralisée permet d'adresser à EURATOM des rapports comptables dans le format voulu (1) (2).

A côté de cet impératif il y a le voeu constant de rapprocher autant que possible les deux systèmes.

3.5. - Confinement, surveillance et protection physique :

Bien que la protection physique soit de la compétence exclusive des Etats, des interférences paraissent possibles entre les deux systèmes de contrôle. En effet dans les opérations qu'ils conçoivent, les contrôleurs internationaux doivent se conformer aux restrictions d'accès motivées pour des raisons de sécurité nationale. Ceci milite en faveur d'un système de confinement et de surveillance de l'organisme international, indépendant de celui de l'exploitant et de son système de protection physique, et sur lequel ce dernier n'aura aucun retour d'information à moins de détection d'anomalie. La présence d'un tel système indépendant renforce malgré tout les dispositions mises en place dans le cadre du contrôle national.
Un autre exemple d'interférence possible entre les deux systèmes concerne la protection physique des transports qui repose en particulier sur des règles strictes de confidentialité quant aux renseignements relatifs aux matières nucléaires et à la date du transport. Or une certaine information doit être notifiée à la Direction du Contrôle de Sécurité d'EURATOM et éventuellement à l'AIEA pour que les inspecteurs aient la possibilité de vérifier sur site certaines réceptions ou certaines expéditions et poser ou ôter des scellés. Afin d'éviter tout risque de fuite d'informations sensibles du point de vue de la protection contre la malveillance en cours de transport, l'emploi d'un système codé de transmission et la plus grande discrétion de la part des inspecteurs internationaux sont nécessaires. La même remarque est d'ailleurs applicable en ce qui concerne les données sur le suivi et la comptabilité (localisation et quantités de matières sensibles) ou sur la surveillance des matières nucléaires dans les installations. Ceci devra d'autant plus être pris en considération à l'avenir que des moyens modernes de transmission (en particulier par satellites) seront utilisés entre les installations et les sièges des organismes de contrôle.

3.6. - Inspections :

a) C'est dans ce domaine qu'apparaissent les distinctions les plus importantes au niveau des procédures et des moyens mis en oeuvre. Ceci est la conséquence des différences entre le contrôle national et les contrôles internationaux qui conduisent à des objectifs d'inspections spécifiques. Contrairement au contrôle national, les contrôles internationaux se fixent pour objectif de détecter des détournements de matières nucléaires effectués par un Etat en une seule fois (abrupt diversion) ou en plusieurs fois sur une année (protracted diversion) d'une quantité significative de matières nucléaires.

La notion de détournement en une seule fois conduit à la prise en compte du facteur temps lié au délai de détection. Il en résulte pour la fréquence, la nature et la portée, deux types d'inspection : celles qui visent le contrôle de l'inventaire annuel et celles dites "intérimaires" relatives aux flux et à l'objectif d'inspection lié au facteur temps.

Dans le cas du contrôle national, les inspections peuvent être de portée générale (examen de la protection d'un site par exemple), ou viser un thème précis (suivi et comptabilité ou confinement, surveillance et protection physique d'une installation ou d'une partie d'installation) ou porter sur l'analyse des procédures établies par l'exploitant, des résultats obtenus et des moyens utilisés pour les obtenir.

A l'occasion des contrôles d'inventaire ou des mouvements de matière, les inspecteurs français effectuent, comme les inspecteurs internationaux, des vérifications indépendantes par analyse non destructive ou prélèvement d'échantillons pour analyse destructive. Mais ils vérifient surtout l'efficacité des procédures et la rigueur de leur application par l'exploitant, en particulier pour l'inventaire, les mouvements de matières (réception, expédition, transferts), les écarts expéditeur-destinataire, les différences d'inventaire ou les écarts de bilan de campagne.

Au contraire, les vérifications effectuées par les contrôles internationaux pour le suivi et la comptabilité portent systématiquement sur des mesures indépendantes utilisant des analyses non destructives ou des échantillonnages pour analyse destructive par un laboratoire extérieur. Ces mesures basées sur des plans d'échantillonnage statistiques ont pour objectif de détecter le détournement d'une quantité significative de matière nucléaire avec une probabilité déterminée (souvent de 95 %).

On constate d'ailleurs que l'utilisation de critères différents aboutit en France à une activité d'inspection de la part d'EURATOM supérieure à celle du contrôle national, avec en particulier une inspection continue des installations de retraitement de la COGEMA à La Hague et d'enrichissement d'EURODIF et une inspection trimestrielle des réacteurs à eau sous pression d'EDF. Le nombre d'inspecteurs internationaux habilités à opérer en France est beaucoup plus élevé que celui du contrôle national.

b) En ce qui concerne le droit d'accès, l'autorité nationale compétente peut faire effectuer à tout moment et en tout lieu toute inspection qu'elle juge nécessaire. Dans le cas des contrôles internationaux, un droit d'accès, à tout moment et en tout lieu, est reconnu par le Traité d'EURATOM et le Statut de l'AIEA. Cependant en pratique les dispositions particulières de contrôle pour EURATOM et les arrangements subsidiaires pour l'AIEA précisent les modalités d'exercice de ce droit d'accès et le limitent dans les faits à des points stratégiques, en particulier les points de mesure principaux.

4 - LE PROGRAMME FRANCAIS DE R & D DANS LE DOMAINE DE LA SECURITE DES MATIERES NUCLEAIRES

Depuis plusieurs années la FRANCE a un programme important de recherche et développement dans le domaine de la sécurité des matières nucléaires. L'effort le plus important est celui consacré à la protection physique qui n'est pas traitée ici d'une part parce que ce sujet n'entre pas dans le cadre ESARDA, d'autre part parce que nous tenons à lui conserver une certaine confidentialité pour mieux assurer l'efficacité des mesures adoptées. Nous nous limiterons donc à ce qui concerne le suivi, la comptabilité et le confinement surveillance.

4.1. - Motivations :

La priorité de ce programme est évidemment de faciliter l'exercice du "contrôle national". Un autre but vise à l'amélioration des contrôles internationaux, en contribuant à la mise au point de méthodes et d'appareils de contrôle dont la mise en oeuvre gêne le moins possible les exploitants tout en assurant l'efficacité des contrôles.

Si certaines recherches sont au départ effectuées pour résoudre un problème spécifique du contrôle national ou des contrôles internationaux, d'autres ont des objectifs communs.

4.2. - R & D en confinement surveillance :

Les développements les plus importants actuellement en cours dans le domaine du confinement surveillance sont les suivants :

4.2.1. - Scellés :

La Direction des Applications Militaires du CEA participe au développement des scellés à ultra-sons VALSEUR et TITUS en collaboration avec le CCR d'ISPRA (4).

Le Département de Sécurité des Matières Radioactives de l'IPSN améliore un scellé à fibre optique "SAFO" associé à un système portable de lecture et de vérification automatique. Le logiciel d'analyse et comparaison des images développé à cette occasion donne déjà des résultats très intéressants. Les efforts actuels portent sur la miniaturisation du dispositif de lecture et d'enregistrement des images, ainsi que sur l'augmentation de la résistance à la fraude des scellés.

4.2.2. - Surveillance d'un stockage de Plutonium en temps réel :

Le plutonium et le tritium dégageant en permanence une énergie calorifique appréciable , cette propriété est mise à profit pour vérifier la continuité de la présence de ces matières dans leur stokage, par simple lecture continue des températures données par des thermistances judicieusement placées.

Les efforts actuels portent sur la transmission par radio de ces températures et sur leur tri informatique, de manière à bien distinguer les ouvertures des conteneurs, des mises en route des ventilations (5).

4.2.3. - Dénombrement, identification et transfert :

Dans le cadre du programme français de soutien des garanties de l'AIEA, le CEA et la COGEMA ont développé un système intégré de dénombrement et d'identification des transfert des assemblages combustibles irradiés entrant dans les piscines de stockage de l'usine de LA HAGUE. Ce système de surveillance appelé CONSULHA est basé sur une détection gamma et neutron, ainsi que sur des prises de vues par caméra-vidéo. Ce dispositif fonctionne maintenant depuis environ un an, et ses premiers résultats font l'objet d'une communication pendant ce congrès (6).

4.3. - R & D concernant des appareils de mesure :

4.3.1. - Mesure de densité des liquides :

Les développements les plus importants actuellement en cours concernent la mesure in situ des densités et des niveaux de liquides dans des cuves par bullage lent de gaz en condition quasi-statique et mesures de pression du gaz. Ce dispositif qui n'utilise aucune source électrique pour la prise d'information est bien adapté aux liquides agressifs, radioactifs et ionisants créant un milieu hostile soit pour le matériel, soit pour l'opérateur. Il a déjà été utilisé avec succès dans diverses usines françaises du cycle du combustible (MALVESI, LA HAGUE), notamment pour des mesures de densité du nitrate d'uranyle. Les efforts actuels portent sur l'automatisation du cycle de mesure et sur la diminution du temps de mesure à moins de 4 minutes (7).

4.3.2. Contrôleur isotopique non-destructif (CIND) :

L'appareil en cours de développement est une amélioration du dispositif utilisé par COGEMA pour mesurer l'enrichissement de l'uranium dans les conteneurs d'UF_6 de type 30B ou 48 Y. Ce dispositif utilise une méthode d'interrogation neutronique active à l'aide d'une source d'Am/Li qui présente l'avantage d'être peu sensible aux variations d'épaisseur des parois des conteneurs résultant de la corrosion, ainsi qu'à l'épaisseur des dépôts internes d'UF_6 souvent collés aux parois.

Les efforts actuels portent sur l'allègement du dispositif initial, l'amélioration du rapport signal/bruit et de la stabilité des signaux émis (nouveaux détecteurs), ainsi que sur la facilité de mise en oeuvre (mesure en bout des conteneurs avec un positionnement plus reproductible de l'appareil par rapport au conteneur).

4.3.3. - Amélioration du traitement du signal dans les mesures neutroniques passives :

Ce dispositif en cours de développement est une amélioration du traitement classique des mesures neutroniques passives par les systèmes de coïncidence à registre à décalage, qui est particulièrement intéressant pour les échantillons de masse importante dans lesquels les effets de multiplication neutronique ne sont pas négligeables (8).

5 - CONCLUSION

La comparaison entre le système français de contrôle et les contrôles d'EURATOM et de l'AIEA peut conduire à se poser la question des rapports entre ces deux systèmes.
Après dix ans d'expérience la comparaison effectuée montre que, malgré des objectifs différents, ils ne peuvent pas s'ignorer, et qu'ils sont complémentaires.

L'existence d'un système national de comptabilité et de contrôle des matières nucléaires, fiable et efficace, est considérée par l'AIEA comme un élément fondamental pour l'efficacité de ses propres contrôles. De même une partie de la R & D effectuée pour les besoins du contrôle national peut, au prix d'une adaptation tenant compte des objectifs spécifiques des contrôles, apporter une aide dans l'application des contrôles internationaux en France, mais aussi dans d'autres pays.

L'application des contrôles internationaux en France devrait permettre de renforcer les dispositions mises en place par l'exploitant dans le cadre du contrôle national dans la mesure où les autorités nationales pourraient bénéficier d'un retour d'expérience de la part des organismes internationaux. D'une manière générale, les anomalies constatées par les contrôles internationaux, fournissent des indicateurs susceptibles de conduire l'autorité nationale compétente à infléchir ses prescriptions. Une certaine concertation pourrait être utile en certain cas pour éviter une duplication ou certaines difficultés.

En conclusion, il paraît intéressant de rechercher les avantages qui peuvent résulter d'un échange mutuel d'informations et d'une concertation bien organisée.

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- REGIME D'AUTORISATION, DE DECLARATION OU D'EXEMPTION
- SEUILS APPLICABLES

	Pu U_{233}	U > 20 % U^5	U < 20 % U^5	U nat U app Th	D	H_3	Li_6
AUTORISATION	> 3 g	> 15 g U^5	> 250 g U^5	> 500 kg	> 200 kg	> 2 g	> 1 kg Li_6
DECLARATION	< 3 g > 1 g	< 15 g U^5 > 1 g U^5	< 250 g U^5 > 1 g U^5	< 500 Kg > 1 kg	< 200 kg > 1 kg	< 2 g > 0,01 g	< 1 kg Li_6 > 1 g Li_6
EXEMPTION	< 1 g	< 1 g U^5	< 1 g U^5	< 1 kg	< 1 kg	< 0,01 g	< 1 g Li_6

THE ROLE OF NUCLEAR ENERGY IN THE EUROPEAN COMMUNITY

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Abstract

The overall objective of the energy policy of the European Community is to achieve a secure supply of energy at reasonable cost and low environmental impact.

This overall objective is embedded in the steps taken by the European Community towards the Single European Market. This subject will be addressed briefly, as well as the developments in eastern Europe including the European Energy Charter.

With regard to nuclear energy in the Community, facts and issues related to electricity production, the front end and back end of the nuclear fuel cycle and current environmental and safety issues will be presented. A common industrial strategy is required in the Community including safety regulations, technical specifications and products for international markets.

Concerning Safeguards, the Commission is fully committed to the obligations from chapter VII of the Treaty and to any relevant international agreements concluded by the Community. This is reflected in the close cooperation of the Commission with the IAEA and in the increase of resources, both in staff and budget, which the Commission has allocated to the Safeguards Directorate to cope with the increasing requirements, for example for bulk handling facilities.

Based on a request from the European Parliament, the Commission has issued in 1989 a first Report on the Operation of Euratom Safeguards. The Commission services are currently preparing the second report of this type.

Finally, some remarks with regard to the future of nuclear energy and challenges of safeguards in the Community will be made.

1. Introduction

It is reported that Albert Einstein surmised that the fate of nuclear power would be decided in the market squares. For European democracies, both new and old, the ability to use nuclear energy depends more on social and political factors than on economic and technical ones.

In the area of nuclear energy, as in some other high technology industry there exists a large gap between what is known and understood by the experts and what the public perceives to be true. This public perception, even if it is wrong, has enormous consequences, because public opinion can have a significant effect on how funds are spent.

The public associates immediately "Three Mile Island" and "Chernobyl" with nuclear energy but who associates "Chelyabinsk" with the use of natural gas ?

It is important, therefore, that adequate information on the risks of nuclear energy be made accessible and understandable to the public. The public has the right to know that all areas in the nuclear fuel cycle are fully developed and are well mastered by scientists and engineers.

One important step amongst others in this regard, was the publication of the International Nuclear Event Scale for prompt communication of the safety significance of incidents and accidents. This scale was designed by an international group of experts convened by the IAEA and the OECD Nuclear Energy Agency and will permit a more objective judgment and classification of the significance of events both to the experts and to the public.

2. Current Energy Issues in the European Community

The overall objective of the energy policy of the European Community is to achieve a secure supply to satisfy demand at reasonable cost and low environmental impact.

2.1. The Internal Energy Market

This overall objective is embedded in the steps taken by the European Community towards the Single Energy Market.

In the past, the Community approached energy matters largely in playing the role of a coordinator of national energy policies of the individual Member States. The European Community has developed a joint energy policy only in particular sectors, namely the European Coal and Steel Community, Euratom and a specific nuclear fusion programme. Since the first oil crisis, the Commission and the Council have agreed on recommendations and guidelines, e.g. for using oil and natural gas, for saving energy, for energy import policy and for energy supply reliability. Euratom has essentially concentrated on research and development activities, health and safety, supply and external relations issues. Consequently the introduction of commercial nuclear power was, and still is, a national decision within each Member State.

Since 1985 (Commission white paper for a single market) and more specifically since 1988 (publication of white paper for an internal energy market) the Commission has initiated a strategy for a common energy policy which could eventually be embodied in the EEC Treaty.

It is important here to emphasize that the Community's role is to set the conditions for a Single Energy Market, i.e. to create a framework for a liberal, barrier-free, secure and competitive trading environment in which industry can operate effectively.

1992 is approaching fast, but good progress can be reported in some areas which are fundamental for the establishment of an internal energy market:

- The Council adopted a directive on transparency of prices, a basic pre-requisite for the operation of an open energy market. Twice per year, prices paid by consumers for energy from the various suppliers will be published.
- The Council also adopted a directive on Community trade in electricity, dealing with the use of the different electricity distribution networks in the Member States across national frontiers.
- This year the Council will adopt a directive on natural gas, which should lead, through better implementation of transit across national frontiers, to increasing trade between existing suppliers and more competition.
- Further proposals in other energy sectors are in preparation, the main focus being the introduction of greater competition by the removal of restrictive national barriers. As a direct consequence of this, the large single market should lead to reduced energy costs to the benefit of all consumers.
- Some open questions still exist in the area of access of third parties to electricity and gas networks of the Community. The Commission undertook during the last 12 months extensive consultations on this topic with the industry as well as with the Member States. The Commission will formulate its proposals in this area later this year.
- Another area of Commission initiative will be the establishment of environmental standards in the energy sector and a certain harmonisation with regard to indirect taxes.
- The Community also adopted a directive to deal with public procurement: certain works or supplies in the energy sector must therefore go out to tender Community - wide and not be subject to pressure to award contracts to local or national suppliers.

Supply is only one side of the coin. The Community has not forgotten the demand side of the equation. Substantial amounts of money and effort have been spent on developments to save energy and to increase energy efficiency. In the area of new energy technology, the Community has spent since 1974 about 1.5 billion ECU supporting some 2.500 projects. The new Community technology programme THERMIE will continue this line of development.

2.2. The international dimension

The Community is not an island nor an isolated "fortress" as some believe. The Community still depends for nearly 50% of its energy supply on resources from outside the Community.

Long before the recent Gulf crisis, the Community has recognised the necessity of strengthening the energy relations with the neighbouring countries of EFTA, the near east and far east, and, of course, the countries of eastern Europe.

The sincere desire to express our solidarity with the populations of the USSR and the countries of eastern Europe, the need to support their energy infrastructure development and liberalization and our concern with the serious deterioration of the energy situation has lead to a number of initiatives and increased cooperation with those states:

- The EC-USSR agreement of 1990 includes provisions for co-operation on nuclear energy, nuclear safety, environmental protection and indeed the energy sector as a whole.
- The Commission coordinates the work of the 24 group, which decided in October 1990 that energy was a priority for cooperation with eastern Europe.
- The Community programme PHARE, originally considered for Poland and Hungary, has been extended to Bulgaria, Romania, Czechoslovakia and Yugoslavia. An amount of 6 Mio ECUS per country could be used during 1991 in the energy field.

2.3. The European Energy Charter

As a follow up of Prime Minister Lubbers initiative for a European Energy Community at the summit in Dublin in June 1990, the Commission proposed a European Energy Charter at the Paris summit in November 1990.

In its substance, the European Energy Charter envisages a scheme for ensuring security and diversification of supply on a pan-European basis, linking the massive energy resources (supplies) of the Soviet Union with the markets (demands) in Western Europe.

Amongst others, two major political issues will have to be considered:

- The geographical scope of the Charter. Presently it is considered that the Community countries, the countries of eastern Europe, the USSR and the EFTA countries should at least be signatories.
- The management and monitoring of compliance with the Charter and its sectoral agreements.

The definition of principles and objectives for such a cooperation and the measures necessary for its implementation will be the subject of an international conference later this year. The Charter will be a two-tier system: the Charter itself, as the code of conduct on broad economic, energy and environmental principles, and then the specific agreements associated with it.

3. Nuclear Energy in the Community, Facts and Issues

Nuclear energy, especially in the context with the European Energy Charter, will be a priority issue for transeuropean cooperation.

This source of energy, which is at present a key source of electricity production not only in the Community, but also in eastern Europe and in the USSR, should be evaluated on its own merits and compared objectively with other energy sources for electricity production in terms of safety, respect of the environment and direct and indirect costs.

3.1. Electricity production from nuclear energy

The net total electricity production in the European Community was about 1700 TWh during 1990 (EUROSTAT/1991). About 56% of this total production was generated by conventional thermal power stations, 35% by nuclear power stations and 9% by hydro-electric and geothermal plants.

The five largest countries of the Community, namely (in order of production) Germany, France, United Kingdom, Italy and Spain account for 86% of the total electricity production.

In the six of the twelve countries of the Community, which produce electricity from nuclear energy the proportions in 1990 were as follows:

- the 55 reactors in operation in France have accounted for 75% of the electricity production. 13% of the French electricity production was exported to other Community Member States;
- in Belgium, the 7 reactors in operation delivered 60% of the electricity production;
- the 9 Spanish reactors accounted for about 36% of the electricity production;
- the 22 (west) German reactors accounted for 33% of the country's total electricity production;
- the 34 reactors in the United Kingdom accounted for about 20% of the electricity production and
- the 2 reactors in the Netherlands accounted for 5% of the electricity production.

What are the future perspectives?

In its "Illustrative Nuclear Programme for the Community" of 1985 the Commission set out as one objective that the electricity production in the Community based on nuclear energy should reach a share of 35% by 1990.

While this objective has been met it is inconceivable that the target of 40% envisaged for 1995 will be reached.

The nuclear reactor programmes in the Member States have changed in the aftermath of Chernobyl. After the accession of the GDR to the Federal Republic of Germany, the construction of 6 reactors there was stopped. France and the United Kingdom have 5 plants under construction and projects for 4 further reactors exist. Germany is considering the construction of new reactors to cope with the expected electricity demands in the eastern part of Germany and to reduce the pollution caused by the extensive use of lignite for electricity production in old technology plants.

It is projected that the electricity demand in the European Community will increase by about 20% over the next decade from 1700 TWh to about 2000 TWh.

The decision on which individual energy source shall be used to meet the increasing demand will not only depend on costs but also on environmental considerations.

Energy costs have a substantial impact on the competitiveness of the economy within and outside the European Community.

The governments of France, the United Kingdom, Belgium and Germany have issued in April 1991 a "Common declaration about the use of nuclear energy for peaceful purposes" in which they agree, inter alia, to stabilise the CO_2 emissions in the Community until the year 2000 on the level of 1990.

This agreement, which is in line with other conclusions/recommendations made previously, for example at the World Energy Conference in Montreal (1989), could imply an increased use of natural gas and nuclear energy to replace existing plants burning coal and oil and to meet additional future energy demands.

3.2. Nuclear Fuel Cycle Developments

While the number of power reactors in the Community may not increase substantially, there are and will be major developments and investments in the so-called back end of the fuel cycle in the Community. For the front end of the fuel cycle, there still exists surplus mining and industrial capacity and large stockpiles. Rationalisation of this sector will take several years owing to the presence of a secondary market for sales of surplus uranium and enrichment services.

With the respect to the back end of the fuel cycle, France and the United Kingdom are constructing and commissioning three large reprocessing plants at Cap La Hague and Sellafield to process the irradiated fuel from inside and outside the Community.

The increased recycling of Plutonium in MOX fuel for light water reactors has lead to 3 projects for new MOX fabrication facilities in Germany (Siemens MOX plant), France (MELOX plant) and the United Kingdom (MDF plant).

Enrichment technology is being improved and new enrichment processes using laser technologies are being developed.

Further construction and commissioning of interim fuel storage installations, waste conditioning plants and final waste storage facilities is planned in France, Germany and in the United Kingdom.

Experience in the decomissioning of nuclear facilities will increase considerably. In addition to the 29 completed decommissioning projects in the Community, there are at present a further 22 projects in progress and 14 projects planned.

With regard to power reactors, it can be expected that industry will stay with light water reactors for about a further 20 years. Improvements will focus on safety features and to a more efficient use of fuel, by improved fuel design, core management, higher burn up and the extended use of recycled plutonium and uranium.

European research centres and the manufacturing industry have agreed to work together in the design of a European Fast Breeder Reactor (EFR), which would be licensable in all European countries involved.

3.3. Nuclear energy and environment

While the choice of the energy options is with the Member States, it is the duty of the Commission to ensure at its highest levels the respect of the Euratom Treaty, for the well being of all Europeans and for a better assimilation of this energy source into society as a whole.

Related to safety and environment, the Commission is supporting directly or indirectly a number of projects.

Authorities in the Soviet Union recognised that the accident of Chernobyl was a tragedy which reached well beyond the boundaries of the USSR. For the first time they broke a tradition of silence.

The European Community took immediate steps to reinforce its activities to improve nuclear safety. Likewise, international organisations specialising in nuclear safety, both public (IAEA and OECD) and private (WANO) started programmes to perfect or update older or less sophisticated reactors.

The Commission has promoted a twinning operation amongst nuclear power stations in western and eastern Europe. The exchange of plant experience accumulated by more than 200 nuclear plants will represent a benefit to all involved to optimise plant operations and as a consequence, enhance safety.

With the financial support of the Community, programmes to evaluate the safety of the the Soviet VVER reactors, to increase their safety levels and to intensify the training of staff are being carried out.

The concept of an open single European energy market requires that, power be generated on an economically competitive basis subject to comparable environmental protection regulations and the necessity of a joint energy policy. Today there are substantial differences between the Member States on environmental standards and safety regulations which influence investment, operating and maintenance costs, and even production capacity.

Against this background, nuclear energy is, in fact, beginning a new phase in its evolution. Nuclear power should become the subject of a common industrial strategy, developed by the interested parties working together. The declaration of the four Member States on the use of nuclear energy referred to previously, is a step in the right direction.

This common industrial strategy should combine the following elements to achieve its objective:

- the European states, within their own sovereignity, should adopt the same regulations in relation to the safety of nuclear power stations;
- the producers of electricity by nuclear plants should reach an understanding and define together the technical specifications of future reactors;
- the main nuclear contractors should get together to outline any proposal for a common product to offer to the international market. French and German companies have already started down this road.

Summing up, it would mean conceiving and bulding a common European reference reactor. Drawing from plans already in use in France and Germany, this European project could be a common enterprise with the Community producers of nuclear electricity, and should they wish, the Member States of EFTA, eastern European countries as well as the USSR, could take part.

In this way, all interested European countries would have taken part in the conception of the reactor with the possibility of adopting identical regulations of nuclear safety and observing the same codes of construction.

Europe would then have standards assuring irreprochable levels of safety and of quality control. The European economic area, then being homogeneous, will increase free competition amongst all the suppliers of nuclear equipment or systems services.

In this way, the objectives of safety and economic viability will be attained in a free market way rather than by Community regulation. For the rest of the century, the nuclear option will remain one option impossible to do without. The indispensible condition is that this energy source receives public acceptance and credibility.

4. Safeguards in the European Community

One of the means to achieve credibility and public acceptance of the nuclear energy is to continue to perform credible safeguards in Europe. To this end and to fulfill the mandate of Article 77 of the Euratom Treaty, the Commission has, since 1958, deployed a corps of Euratom inspectors.

The Commission has since that time not only supported the increasing use of nuclear energy in the Community, but has assured in a complementary way that additional resources were made available for safeguards of the civil nuclear fuel cycle as would be the case for safety considerations in other industries involving high technology.

The Commission is fully committed to the obligations from chapter VII of the Treaty and to any relevant international agreements concluded by the Community. This is reflected in the close cooperation of the Commission with the IAEA and in the increase of resources, both in staff and budget, which the Commission has allocated to the Safeguards

Directorate following approval by the Community's budgetary authorities to cope with the increasing requirements at bulk handling facilities.

At the end of 1990, Euratom safeguards applied to 795 installations of the European Community. The total stock of civil nuclear material under Euratom safeguards at the end of 1990 was about 83.000t depleted uranium, 44.000t natural uranium, 32.000t low enriched uranium, 13t high enriched uranium, 203t plutonium and 2.600t of thorium.

The Euratom Safeguards Directorate had at the end of 1990 227 staff members of which 165 were operational inspectors. During 1990 more than 7.500 mandays of inspections were carried out. The operational costs (excluding staff costs and main frame computer) were about 6 Mio Ecu.

The Commission provided in early 1990 for the first time, a comprehensive survey on the operation of Euratom safeguards in the civil nuclear fuel cycle to the Council and the European Parliament.

The Commission continues to support the worldwide role and responsibilities of the IAEA safeguards department. This is in line with the commitments undertaken by Member States in the area of non proliferation policy.

The close cooperation with the IAEA is reflected in a high effectiveness of safeguards in the Community Member States and a high level of verification technology applied in the nuclear installations of the Community.

The Commission has noted with interest the statement made in February 1991 by Dr. Blix about possibilities to strengthen the non proliferation regime.

The Commission is continuing to reflect on the contents of this statement, which will clearly have to be examined closely by all concerned both inside and outside the European Community.

These examinations will include, but will not be limited to the issues set out below.

Should the IAEA be provided with more comprehensive safeguards information notibly in the area of reporting of international transfers and of yellow cake.

Advanced access of the IAEA to installations not yet having nuclear material and further improvements of IAEA inspectors designation should also be addressed.

With regards to early provision of details of nuclear facilities operators and Member States of the Community have already been cooperative.

The idea to extend the scope of non proliferation safeguards in Nuclear Weapon States has been noted. Yet another possibility could be to consider a demonstration approach, for example, the selection of a plant of each type in the fuel cycle could be a more appropriate and efficient way rather than a full IAEA safeguards coverage of the civil installations in Nuclear Weapon States.

The effectiveness of Euratom safeguards depends on the manner in which the inspection service is organised and motivated, on the promptness and the extent to which operators and state authorities fulfill their responsibilities and on the · resources available to safeguards.

The Commission has established a medium-term planning which, if adopted and implemented, will make a decisive contribution to discharge effectively its responsibilities under the Euratom Treaty and to enable the implementation of non proliferation safeguards in the Community to remain at its present high level.

Member State Support Programmes:
Invaluable Assets to IAEA Safeguards

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Abstract

In order to improve the efficiency and the effectiveness of its safeguards programme as well as to meet the new challenges which constantly arise, the IAEA is required to develop new safeguards instruments, procedures and techniques. The majority of the Agency's development needs is addressed by the development activities conducted through Member State Support Programmes (MSSPs). In order to maximize the use of these extra-budgetary resources, the Agency undertook an initiative in 1988 to review the modus operandi of the MSSPs which resulted in a number of changes in its organizational structure and administrative procedures. A thorough review and restructuring of the existing programmes was completed during 1989/90 via Agency/MSSP bilateral meetings. Some of the results of this review as well as some examples of invaluable contributions made by MSSPs will be presented.

I. Introduction

Since the very beginning when the scheme of international safeguards was conceived and materialized through conclusion of relevant international legal instruments, the importance of safeguards R&D efforts was recognized in order to establish a credible safeguards system. For example, INFCIRC/153 refers to the technological developments in the field of safeguards, stipulating that "the Agency shall take full account of technological developments in the field of safeguards and shall make every efforts to ensure optimum cost-effectiveness..." (paragraph 6). "Technical developments" is also referred to in INFCIRC/153 as one of "the criteria to be used for determining the actual number, intensity, duration, timing and mode of routine inspections of any facility" (paragraph 81). Safeguards could not be efficiently and effectively implemented without extensive use of various types of equipment and instrumentation both for nuclear material accountancy, a safeguards measure of fundamental importance, and for its important complementary measures - containment and surveillance.

The main responsibility for developing, acquiring and testing the necessary equipment and instrumentation lies with the Division of Development and Technical Support (SGDE) within the Department of Safeguards. However, the International Atomic Energy Agency (IAEA) does not possess enough human and financial resources to operate a special research and development laboratory for conducting required safeguards R&D activities and is therefore very much dependent on the support of its Member States. The Agency's role is expected to be more on the identification of R&D needs and their priorities, communicating these to the Member States and coordinating the resultant R&D activities carried out by the Member States.

II. Member State Support Programmes

Evolution of MSSPs

In the late 1970s and early 1980s, the Agency's safeguards implementation programme expanded rapidly due to substantial increases not only in the total amounts of nuclear materials under safeguards but also in the diversification of their chemical and physical forms as used in a wide variety of facility types. This necessitated a substantial increase in the scope and scale of corresponding safeguards R&D work. Fortunately, this increase was accompanied by the growth of a scheme that greatly assisted the Agency in developing the tools it needed to meet its challenges - the establishment of Member State Support Programmes (MSSPs) for Agency Safeguards.

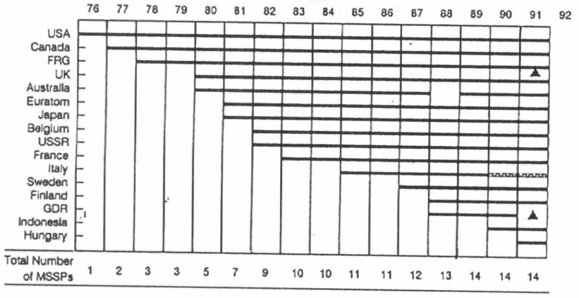
The scheme of MSSPs was initiated by the USA in 1976 and followed later by other countries such as Canada (1977), the Federal Republic of Germany (1978), United Kingdom (1980), Australia (1980), Euratom (1981), Japan (1981), Belgium (1982), USSR (1982), France (1983), Italy (1985), Sweden (1987), Finland (1988), the German Democratic Republic (1988) and Indonesia (1990).

Table 1 summarizes the historical trend of how MSSPs have expanded. Due to financial problems, the Australian Support Programme was temporarily terminated in 1988 but established again in 1989. The change of its national nuclear policy forced Italy to announce the termination of its formal support programme in 1989. However, arrangements have been made to allow some carried over tasks to be conducted while its funds left with the Agency last. The unification of Germany in 1990 made the GDR Support Programme to discontinue, while some of its tasks being incorporated into the FRG Support Programme. Talks are currently under way to identify areas of support by Hungary and to define tasks under its support programme.

Table 1

Historical Trend of

Member States Support Programmes (MSSPs)



Thus, since the start of the scheme, there has been considerable expansion. Currently there are 14 MSSPs with approximately 150 active tasks and a total budget estimate of not less than US $ 22 million. It is clear from these figures that the original concept of a joint co-operative effort has proved to be successful. The MSSP is mutually beneficial, both to the Agency and the Member States.

For the IAEA, especially under the current zero growth budget situation, there are many benefits to be expected from the MSSPs. These include: the existence of a flexible source of funding and project management; access to the varied resources of some of the world's leading nuclear facilities, the availability of realistic environments for testing and evaluating safeguards equipment and training Agency personnel; and access to valuable sources of information on trends in the nuclear industry and facility construction schedules.

For the Member States involved in the MSSPs, on the other hand, their benefits include: assistance in preparing their facilities to deal with the actual safeguards measures to be deployed; assurance that the equipment and techniques used will meet their safety requirements; provision of a forum through which they can ensure that the IAEA is made aware of practical constraints imposed by particular facility environments and operating procedures; and information on the latest techniques and instrumentation.

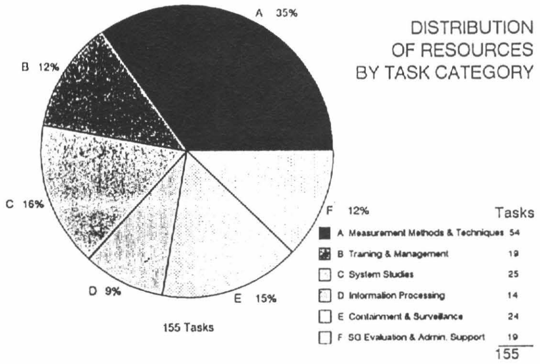
The main requirement in early years when the idea of MSSPs was conceived was for the development of safeguards instrumentation, since the equipment needed for inspection purposes was often not available commercially and the IAEA did not have the resources to develop, test and produce its own. Another major need was for training of safeguards inspectors. The Member States not only generously provided training materials, equipment, facilities and lecturers but also organized and funded entire courses on a continual basis. A certain number of other activities, such as the development of new safeguards approaches, information treatment, data evaluation, quality assurance and management and administrative support were also included in the MSSPs.

Current Status of MSSPs

Table 2 shows the distribution of currently active MSSP tasks by task category as follows:

A. Measurement Methods and Techniques
B. Training and Management
C. System Studies
D. Information Processing
E. Containment and Surveillance
F. Safeguards Evaluation and Administrative Support

Table 2


DISTRIBUTION OF RESOURCES BY TASK CATEGORY

As can be seen from the table, nearly half of the active tasks are related to safeguards equipment for nuclear material accountancy measurements (category A) and containment and surveillance (C/S, category E). Two thirds of the category A tasks are devoted in the area of NDA techniques - development of both portable equipment, such as CVD (Cerenkov Viewing Device) and SFAT (Spent Fuel Attribute Tester), and installed equipment, such as upgraded CANDU bundle counters and CDM (Core Discharge Monitors), including preparation of measurement procedures and development of measurement software. The rest of the category A tasks are related to DA (destructive assay) technique, such as provision of reference materials, quality control at SAL (Safeguards Analytical Laboratory) and development of various measurement techniques including hybrid K-edge densitometery, coulometry and robotized sample preparation.

Two thirds of C/S (category E) tasks cover surveillance devices, mostly in the area of video surveillance such as COSMOS (Compact Surveillance & Monitoring System), MOS (Multi-camera Optical Surveillance System) and MUX-II (Multiplex Surveillance System), including video-record reviewing techniques. The rest is in the area of containment such as development of ultrasonic seals (In-situ Readable Ultrasonic Seal System - IRUSS project) and electronic seals (Variable Code Sealing System - VACOSS).

In the area of training (category B), nearly half of the active tasks is related to organizing and funding various basic inspector courses, training courses on specific types of facilities, such as CANDU reactors and enrichment and fuel fabrication facilities, and on specific types of safeguards technique such as CVD and GBUV (Gamma Burn-up Verifier). Quite a few tasks are related to provision of CFE's (cost-free experts) assigned to the Training Section for managing various courses.

Through system studies tasks (category C), facility-specific safeguards approaches are being developed for new types of facilities that are foreseen to come under the Agency's safeguards in the near future, such as a large scale commercial reprocessing plant, a MOX (mixed oxide) fuel fabrication plant, a spent fuel disposal facility in geological repository and a pilot spent fuel conditioning facility. Substantial efforts are also devoted to formulation of "new safeguards approaches", such as a fuel-cycle oriented approach, a zone approach and a randomized inspection approach, and a safeguards effectiveness evaluation methodology.

Examples of category D tasks in the area of information processing are those related to ISIS (Integrated Safeguards Information System), PC applications such as IFSS (Inspection Field Support System) and LAN (Local Area Network).

Category F tasks are mostly devoted to the provision of CFE's or consultants services in the area of safeguards evaluation and administrative support as well as funding of travels to participate in meetings related to active tasks.

Speaking of CFE's, nearly 20% of the total number of currently active tasks is devoted to this particular form of extra-budgetary support to the Agency by the Member States, covering almost equally each of the above-mentioned task categories - e.g. experts in the area of design, development, operation or maintenance of safeguards equipment and instrumentation for NDA, DA or C/S as well as specialists in training, system studies, computer science, programming, data evaluation, statistical analysis, quality assurance, occupational health and safety, and so on. The total number of CFE's currently available in the Department of Safeguards is 31, that amounts to 15% of the total number of regular professional staff in the support divisions. This demonstrates how substantially the MSSPs extend the Agency's human resource capabilities. CFE's are now found in any Division other than Operations Divisions that carry out actual safeguards inspections.

Another example of significant contributions made by the MSSPs is the fact that virtually all the safeguards equipment presently used by the Agency has been developed, tested, improved or documented through MSSPs. More specifically, 46 out of 57 devices that have been authorized for routine inspection use are either direct products of MSSPs or made available in parallel with them while the others, being purchased commercially, have also been improved or documented as a result of various MSSP activities.

Table 3 shows the principal programme areas where current MSSP efforts are concentrated. It is expected that this trend will remain also in the near future.

Table 3

Principle Programme Areas

	Number of Tasks
1. Cost Free Experts	31
2. CANDU Reactors	16 (+9)*
3. Reprocessing and MOX Facilities	24
4. New Facility Types and Approaches	16

* The number in the parenthesis indicates those in the "stand-by" status, pending finalization of workplans or "end-user" endorsement.

III. The Support Programme Initiative[1]

As stated above, MSSPs have grown substantially over the last decade or so, adding invaluable assistance to Agency Safeguards in achieving the levels of effectiveness and efficiency that currently characterize its activities. By 1988, it reached the stage where more than 300 tasks were covered by 13 MSSPs, at a cost estimated in excess of 14 million US dollars per year. Inevitably, the administrative, liaison and coordination functions performed by the IAEA in cooperating with the MSSPs had increased significantly.

In view of these developments, a project called "Support Programme Initiative" (SPI) was initiated in April 1988 for reviewing the modus operandi of the MSSPs with the objective to identify ways to improve the Department of Safeguards overall efficiency while maintaining or increasing the already high level of effectiveness. As a result of this review, a system has been designed which emphasizes a product-oriented, client-developer or client-supplier relationship between the Department of Safeguards and MSSPs with well defined task objectives, implementation schedules, and lines of responsibility.

Some guiding principles for the approach taken are:

- Increase Agency control in establishing the IAEA R&D Programme and prioritization of MSSP tasks;
- Focus on the product - what the Department <u>needs</u> and when and what the MSSPs are expected to provide and how it will be utilized;
- Take full account of the special capabilities, facilities and interests of different MSSPs in the allocation of tasks;
- Separate MSSP administrative activities from technical aspects;
- Increase the technical involvement of the Department end users, while streamlining the administration through centralization and increased use of modern communication methods;
- Utilize task results in the Department of Safeguards and feed back experience to Member States; and
- Increase interaction between MSSPs.

The SPI resulted in the adoption of the Department's structure for managing and administering the MSSP tasks, establishment of the procedures to assure the effectiveness of these tasks and the new information and communications system (SPRICS) to increase the Department's efficiency.

The Department's Support Programme Organization

The Department's organizational structure was adopted to make full use of line functions and to assure continuing input from end-users through the complete task cycle. In order to separate MSSP administrative activities from technical aspects, the Support Programme Administration (SPA) was established under the Director of SGDE, supported by a small group responsible for centralized administration of the Agency's interaction with MSSPs. The responsibility for monitoring and controlling specific tasks from technical points of view lies with the corresponding task officers assigned by a Division Director who is responsible for the technical management of particular tasks.

Support Programme Procedures

A set of procedures has been established defining the life-cycle of a task and the corresponding responsibilities for its successful completion.

In the phase of task initiation, task proposals are made to one or more of the MSSPs using new task request formats, called SP-1's. R&D task proposals are derived from the Department's R&D programme*, while non-R&D tasks proposals from needs identified in Divisional work plans.

An SP-1 form is prepared by an end user and a Support Division jointly, with the respective Directors' signatures, indicating that each new task is expected to result in a needed product and is to be considered an investment for which the requestor (end user) will later be held accountable in implementing. An SP-1 clearly defines the Agency's needs and identifies the safeguards impact and consequences when the task is successful.

For those task proposals which a MSSP has accepted as a part of their programme, task officer assignments are made by both the Agency and the MSSP and corresponding task statements are formulated with clear definition of work plans indicating expected milestones for various "deliverables" or end-products.

Task progress is monitored primarily by a task officer and a respective work plan is revised as necessary in the course of its implementation.

A bilateral (Agency/MSSP) review meeting with each MSSP is held annually to review the progress of each task, determine its future direction, evaluate the utilization of completed tasks and present a package of new task proposals in draft SP-1 forms. Prior to each bilateral meeting, a task status report is prepared by Agency task officers and reviewed at an in-house review meeting. These reports are also made available to the MSSP concerned as well as to other MSSPs through the SPRICS.

Support Programme Information and Communication System (SPRICS)

In order to improve access to MSSP related information and enhance communication between MSSPs as well as between MSSP(s) and the Agency, the Support Programme Information and Communication System (SPRICS) was developed as a MSSP related data base and became operational in mid-summer 1989 with the full support of the major MSSPs. It is available not only on all Wang word processing terminals in the Department but also available at remote stations such as MSSP offices through public telephone lines. The US Support Programme

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* In parallel to the SPI, the Department, in consultation with the SAGSI (Standing Advisory Group for Safeguards Implementation), has adopted a new systematic approach for the establishment of the Department's biennial (e.g. 1991-92) R&D programme. The new method adopted provides the management with a tool for helping to make objective decisions on the basis of a set of prioritized needs[2].

Office is regularly using this facility, while tests are under way to extend this service to MSSP offices in Australia, Canada, France and Japan. The system has proven to be useful, improving the overall ease of communication among all parties involved, particularly by increasing the overall transparency of MSSP activities and assisting management decisions on questions of programme balance and priorities.

Experience Gained in SPI Implementation

The last one year and a half since the fall of 1989 can be seen as the transition period for the implementation of the SPI, during which a number of new procedures and organizational structures as mentioned above have been introduced and fully implemented in order to improve the overall management of MSSPs and enhance Agency/MSSP interactions. Presently there are 10 joint tasks where several MSSPs are working together on one task by integrating available MSSP resources. All the existing MSSP tasks have been carefully reviewed to identify their "end-users" and define workplans, resulting in a reduction of approximately 50% in the number of active tasks.

The general reaction of the MSSPs to the Initiative has been extremely positive. Especially they favour the new SP-1 form which has proved useful in clearly defining the Agency's needs and communicating them to MSSPs. In dealing with their funding authority, some MSSPs have even started to use these SP-1's for justifying their requests for funding.

Based on the actual experience gained, some minor modifications to the SP-1 have been made by incorporating comments received from the MSSPs. A standard list of "deliverables" has been prepared as an annex to the SP-1 for incorporation into workplans once task proposals are accepted by a MSSP.

The SPRICS has proved an invaluable resource for internal organization and managing of the programme. Upgrades, based on the initial years of experience are underway. It is anticipated that a number of Member States will connect to the system as its remote users in the coming year.

IV. MSSP Coordinators Meeting

In order to have a feedback from the MSSPs based on their experience gained through the implementation of the SPI, a meeting of MSSP coordinators was convened in Vienna in April 1991[3]. The main objective of the meeting was to review the IAEA/MSSP management and administrative framework and associated procedures, with a view to further improving programmatic efficiency and effectiveness. The meeting agenda was arranged in such a way that it also provided excellent opportunities for informal consultations among MSSP coordinators in order to increase interactions between MSSPs and identify areas for mutual cooperations.

The meeting was attended by some 40 representatives from 14 MSSPs and two observer countries, Italy and the Netherlands, and was successful to produce its final report with specific recommendations, inter alia, in the following areas:

1. Programme Definition;
    - the mechanism for the MSSPs to provide input to problem definition in the process of formulating the Agency's R&D programme,
    - the importance of "special tasks" in addition to normal tasks,
    - the means to provide the Agency with a profile of the MSSP resources, or a list of resources and capabilities that are readily available within the MSSPs both for R&D and non-R&D activities,
    - the potential areas of joint tasks,
    - the scheduling of a bilateral meeting in conjunction with a MSSP's budgetary cycle.
2. Programme Management;
    - the allocation of task activities between the Agency and the MSSPs,
    - the procedures for milestone monitoring and status reporting,
    - the incorporation of task implementation reports,
    - the detailed arrangements for standardization of administrative procedures for Agency/ MSSP interactions,
    - the need for further promoting interactions among MSSPs, including holding a technical meeting.

Several actions have been identified both for MSSPs and the Agency for implementing these recommendations.

V. Concluding Remarks

As can be seen from the above description, the assistance provided by the MSSPs has contributed significantly to the Agency's ability to meet its safeguards responsibilities in an effective and efficient manner.

The future directions of MSSPs are expected to be as follows:

- Increased focussing of MSSP tasks on problems defined in the Agency's biennial R&D programme;

- Achievement of increased efficiency through documentation and optimal standardization of administrative procedures for enhanced Agency/MSSP interactions;
- Further promotion of joint tasks that would result in products greater than those produced by a single MSSP by means of combining available financial, technical or experienced human resources to address a common problem;
- Increased focussing on MSSP resource profile for best utilization of available resoureces in the existing MSSPs;
- Addition of new MSSPs to expand available resources.

In view of the future growth in both the size and sophistication of the nuclear industry and the prevailing Agency's extremely tight budget constrains, the MSSP assistance will continue to be vital to the Agency in successfully executing its obligations.

References

[1] "Support Programme Initiative - Summary and Action Plan", updated June, 1989.

[2] "The IAEA Safeguards 1991-1992 R&D Programme", September 1990.

[3] "The Report of the 5th IAEA/MSSP Coordinators Meeting", 15 - 19 April 1991 (in press).

INTERNATIONAL SAFEGUARDS AND INTERNATIONAL NUCLEAR TRADE

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ABSTRACT

A particular feature of nuclear trade is the need to ensure a good equilibrium between technology transfer and the reguirements linked to non proliferation of nuclear weapons. The recent dramatic changes that occurred on the international scene have particularly underlined the difficulty to reach this equilibrium, reminding us the fragility of the existing order and the responsibility of the nuclear community in the field of non-proliferation. Consequently, there is a need for an international thorough review of the actual non proliferation tools developed until now. This review could lead to reconsider some of the "dogmas" upon wich the present safeguard system is based.

Les transferts et échanges d'information, d'équipement et de matières nucléaires ont été au début du développement des technologies nucléaires considérés comme tabous y compris dans leurs applications pacifiques.

Il a fallu la politique dite d'Atomes pour la Paix ("Atoms for Peace") lancée par le Président EISENHOWER et la première conférence de Genève pour que les informations à l'exception de celles concernant certaines technologies considérées comme particulièrement sensibles, commencent à être élaborée.

Dès cette époque, cependant, il avait été clairement reconnu qu'une telle ouverture ne pourrait se faire sans contre-partie. Cette contre-partie était l'acceptation par le pays récipiendaire de contrôles internationaux.

On peut donc dire que, dès l'origine, les contrôles internationaux ont été la condition sine qua non des échanges de technologies et matières nucléaires.

A cet égard, il est intéressant de s'interroger sur la situation telle qu'elle nous apparait aujourd'hui.

Dans un contexte national marqué par une forte progression démographique par l'augmentation des besoins énergétiques et par des préoccupations croissantes de protection de l'environnement, le marché nucléaire aujourd'hui déprimé, pourrait reprendre un nouvel essor pour participer à la satisfaction des besoins énergétiques de la planète.

Les pays qui désirent s'équiper sur le plan électronucléaire devront pouvoir le faire, mais il faudra que soient apportées des garanties sérieuses contre le risque de dissémination des armes. C'est l'intérêt de tous d'aider à cette difficile conciliation entre la coopération dans le domaine nucléaire et les impératifs de non prolifération des armes nucléaires. Le conflit que nous avons connu durant les premiers mois de cette année a mis en évidence la fragilité de cet équilibre et a provoqué une accélération de la volonté de la Communauté Internationale de renforcer le dispositif actuel de la non prolifération en utilisant les instruments existants.

I - UNE CONCILIATION DIFFICILE

Le commerce nucléaire international a toujours été confronté à une conciliation difficile entre l'émergence de nouveaux utilisateurs et le souci de non prolifération des armes nucléaires.

L'exportation d'équipements nucléaires recèle en elle-même un dilemme : alors que tous les Etats, qu'ils soient dotés d'armes nucléaires ou pas, sont d'accord pour éviter toute prolifération, comment faire pour que la vente d'une centrale nucléaire destinée à produire de l'électricité par exemple, dont la production permettrait de résoudre un handicap énergétique, ne puisse pas être utilisée par la suite à des fins nucléaires militaires. Ce dilemme de l'utilisation pacifique de l'énergie nucléaire apparait plus encore aujourd'hui comme l'enjeu majeur d'une politique mondiale de l'énergie nucléaire et ne peut être résolu que grâce aux contrôles internationaux.

Nous pouvons toujours en 1991 nous référer aux constats simples établis pour les travaux des experts sur l'évaluation Internationale du Cycle du Combustible (ou INFCE) :

- Le recours à l'énergie nucléaire reste un nécessité pour faire face aux besoins énergétiques mondiaux;

- Il n'y a pas de cycle nucléaire qui ne soit intrinsèquement non proliférant et aucun "verrou technologique" ne permet de développer l'électronucléaire en excluant tout risque de prolifération.

- Les moyens les plus efficaces d'une politique de non prolifération sont les engagements de non prolifération et de contrôle.

Les contrôles internationaux dont le but est de déceler le détournement de quantités significatives de matières nucléaires vers des activités militaires et de dissuader les Etats de tenter de tels détournements sont de fait l'outil essentiel de lutte contre la prolifération.L' AIEA est l'instrument des contrôles internationaux à l'échelle mondiale.

Ainsi, l'acceptation des contrôles est devenue une condition reconnue pour qu'un pays puisse être considéré comme un partenaire sérieux en matière de commerce nucléaire international.

Dans bon nombre d'échange nucléaires, notamment de matières, les garanties de l'AIEA sont aujourd'hui devenues une donnée essentielle.

Le régime de non prolifération a efficacement rempli son rôle et en particulier, les contrôles internationaux ont répondu à nos attentes. Cela n'empêche que, comme pour toute autre institution, il faut régulièrement remettre en cause, émettre des doutes, voire des critiques pour améliorer l'ensemble de ces dispositifs.

II - LES ELEMENTS D'UN RENFORCEMENT NECESSAIRE DU DISPOSITIF DE NON PROLIFÉRATION.

Les bouleversements que nous avons connu depuis deux ans et les événements récents ont rappelé à l'ensemble de la Communauté Internationale que les questions de non prolifération devaient être au premier plan de ses préoccupations.

Nous en avons eu en effet la confirmation :

Tout d'abord, le conflit du Golfe a provoqué une prise de conscience plus aiguë du fait que les armes non conventionnelles pouvaient être conçues dans certaines régions du monde comme des armes d'emploi et non de dissuasion.

Ensuite, il a été confirmé que la solution la plus aisée pour acquérir des matières permettant de fabriquer des armes n'est pas le détournement délibéré d'installations civiles à des fins militaires.

L'actualité a au contraire mis en évidence que la réalisation discrète d'installations spécifiques est la voie la plus probable pour acquérir l'arme nucléaire.

Ceci nous donne d'ailleurs une nouvelle confirmation que les contrôles ont joué pleinement leur rôle de surveillance et de dissuasion.

Jusqu'à présent certains avaient à l'esprit uniquement le risque constitué par le pays n'ayant pas signé le TNP et n'ayant pas soumis l'ensemble de leurs activités nucléaires aux garanties de l'AIEA.

Pour l'avenir, nous avons la confirmation qu'un risque réel est aussi constitué par l'Etat qui souscrit tous les engagements conformes au TNP et achète une sorte de certificat de bonne conduite en étant très coopératif en matière de contrôles internationaux, mais se livre à des activités clandestines en utilisant tous les points fragiles ou lacunes du système des garanties.

Ce constat inquiétant a provoqué une accélération de la réflexion de l'ensemble de la Communauté Internationale pour un renforcement du régime actuel de non prolifération.

III - LES REFLEXIONS ET PROPOSITIONS ACTUELLES POUR UN RENFORCEMENT DU REGIME DE NON PROLIFERATION

Il est utile de rappeler que les éléments qui constituent le régime actuel de non prolifération sont :

- Le TNP et ses accords d'application des garanties de l'AIEA.

- Les Directives de Londres qui sont un engagement unilatéral pris par 14 pays lors de leur signature et dont le nombre est aujourd'hui de 26.

- Les engagements pris à l'occasion d'accords intergouvernementaux de coopération.

- Des engagements d'exportation pris depuis 1974 et régulièrement révisés au cours des travaux du Comité dit ZANGGER chargé d'expliciter l'article III 2 du TNP et qui sont publiés par l'AIEA sous la référence INFCIRC.209.

Sous tous ces éléments du dispositif actuel de non prolifération une réflexion est largement engagée aujourd'hui et des propositions se font jour.

- Les Directives de Londres

A la suite de l'explosion indienne, les principaux pays industrialisés nucléaires se réunirent à l'instigation américaine au sein du "Club de Londres" pour envisager de renforcer le dispositif du TNP. Certains envisageaient, à l'époque, de frapper d'embargo nucléaire les pays n'ayant pas toutes leurs activités nucléaires sous contrôle de l'AIEA (Full Scope Safeguards) et d'interdire l'exportation de certaines matières ou équipements particulièrement sensibles. Ces idées ne furent pas retenues pour les Directives de Londres publiées par l'AIEA EN 1978 sous la référence INFCIRC 254.

Ces Directives prévoient que les fournisseurs ne doivent autoriser le transfert d'articles énumérés dans la liste dite de base, que contre l'assurance gouvernementale formelle des destinataires par laquelle ils excluent expressément des utilisations qui aboutiraient à l'obtention d'un dispositif explosif quelconque.

Pour les Etats non dotés d'armes nucléaires parties au TNP, cet engagement résulte de leur adhésion à ce traité pour toutes leurs activités nucléaires.

Longtemps repoussée, mais considérée aujourd'hui par tous comme inévitable, une révision de la liste de base s'annonce déjà puisque nous sommes sur la voie d'une inclusion des listes du comité ZANGGER dans les Directives de Londres.

Mais la liste de base des Directives de Londres, comme celle du Comité ZANGGER, concerne uniquement les produits fissile spéciaux, les matières nucléaires ou non, et les équipements, les installations et les technologies du cycle du combustible nécessaire à leur traitement, leur utilisation, et leur production.

La liste publiée dans le document INFCIRC.254 n'ayant fait l'objet d'aucune consultation depuis sa parution en 1978, devrait faire l'objet de précisions afin de tenir compte des évolutions techniques qui ont vu le jour depuis cette date.

Cela concerne notamment les installations d'enrichissement utilisant les procédés de séparation isotopique aérodynamique ou par échange chimique ou ionique ou encore de vapeur atomique ou moléculaire par laser ou à plasma, les équipements et installations spécialement conçus ou préparés pour la production d'UF6. Des précisions pourraient porter aussi sur les équipements et installations spécialement conçus ou préparés pour la fabrication de combustibles métalliques ou de combustible à base de plutonium.

Par ailleurs, en ce moment même, les 26 adhérents aux Directives de Londres ont entamé une réflexion afin d'instituer un nouveau régime international de contrôle à l'exportation qui s'appliquerait à des équipements et matières à double usage. Des liste d'articles ont été élaborées dont certaines correspondent à des articles à ajouter aux Directives de Londres et seraient contrôlables par l'AIEA, et d'autres comprennent des articles à double usage qui devront faire l'objet de contrôle à l'exportation pour lesquels un régime spécifique devrait être trouvé.

Cette étude est d'ailleurs à l'ordre du jour de réunions du Groupe de non prolifération de la coopération Politique Européenne.

- Les garanties de l'AIEA

Comme l'ont rappelé les Douze dans leur déclaration à l'occasion de la Conférence Générale de l'AIEA de septembre 1989, les garanties de l'Agence constituent la clé de voûte d'une politique d'utilisation pacifique de l'énergie nucléaire. Il est dès lors important de maintenir , voire de renforcer, sa crédibilité et son efficacité.

J'ai évoqué au début de ma présentation des lacunes du système des garanties, il est plus juste de parler de limitations imposées par les textes et même d'auto-limitations de l'Agence.

C'est en se référant à ce constat qu'un certain nombre de suggestions sont avancées actuellement.

Il est à constater en effet que les textes imposent des limites de compétence à l'AIEA.

On peut ainsi indiquer que seules sont contrôlables les matières définies à l'Article XX du Statut. Les équipements et les installations ne sont contrôlés que s'ils détiennent l'une de ces matières.

C'est dans ce sens que beaucoup pensent qu'une clarification devrait être faite quant au rôle de l'Agence dans le cas de la fourniture de certains équipements visés par les Directives de Londres qui requièrent des garanties et qui ne sont pas contrôlés en pratique.

Une autre limitation concerne le point de départ des garanties qui établit que les seules matières nucléaires ayant atteint un stade dans le cycle du combustible propre à la fabrication du combustible ou à l'enrichissement sont soumises aux garanties.

Pour l'uranium, cela signifie aujourd'hui le dioxyde (UO2), le métal ou l' hexafluorure (UF6). Des suggestions sont faites aujourd'hui pour une application des contrôles dès le stade du "yellow cake"

Il faut cependant inciter l'Agence à faire des études approfondie pour vérifier si une telle extension contribuerait de façon significative au régime de non prolifération, ce qui n'est pas à priori évident.

L'AIEA s'impose par ailleurs des auto-limitations dans l'exercice de ses contrôles.

Force est de constater que l'Agence a le droit d'effectuer une partie de ses inspections régulières sans notification tel qu'il est dit au paragraphe 84 de l'INFCIRC 153. L'exercice de ce droit aurait très certainement un important effet de dissuasion qui est reconnu comme faisant partie des garanties aux termes du paragraphe 28 de l'INFCIRC 153.

Il faut s'interroger aussi sur le fait que l'Agence n'a pratiquement jamais eu recours aux inspections spéciales.

L'Agence peut appliquer ce type d'inspection notamment lorsque les renseignements obtenus au moyen des inspections régulières ne lui suffisent pas pour s'acquitter de ses responsabilités.

Il est indéniable qu'une réactivation de ces dispositions serait un facteur de crédibilité supplémentaire pour l'Agence.

Il faut rappeler aussi que le Secrétariat de l'AIEA a le souci constant d'améliorer le système en recherchant de nouvelles approches de contrôles. Le Standing Advisory Group on Safeguard Implementation (SAGSI) mène des travaux qui s'éloignent de l'approche actuelle, installation par installation que l'on peut qualifier de philosophie "déterministe" des contrôles où les activités d'inspection découlent directement des stocks de matières présents dans l'installation. Les travaux s'orientent vers une approche davantage "probabiliste" prenant en compte tout ou partie du cycle du combustible. Cette approche devra pour être convaincante garantir au moins le même niveau de qualité que la précédente et aboutir au même résultat de dissuasion. L'Agence espère par ailleurs dans l'application de cette nouvelle approche obtenir une baisse sensible de l'effort d'inspection.

Un certain nombre de programmes de soutien aux garanties, dont celui de la France, contribuent à l'analyse en cours.

Nous avons de manière générale à soutenir l'Agence dans ses efforts de rationalisation des méthodes d'inspection et nous pensons que des suggestions pourraient être faites pour l'élaboration de nouveaux systèmes de programmation et d'évaluation des inspections.

Une détermination des objectifs tenant compte de la sensibilité des matières nucléaires, de la complexité des installations, du cycle du combustible, en est un exemple.

L'Agence devra peut-être aussi ne pas s'attacher de manière rigide aux valeurs quantifiées qu'elle s'est fixée pour interpréter les termes "temps de détection" et "quantités significatives" mais les pondérer en plus ou en moins en fonction d'une appréciation sur les installations replacées dans un cycle du combustible.

Au terme de ces quelques réflexions dont la mise en oeuvre permettrait de renforcer encore la crédibilité des contrôles internationaux, j'en suis à ma conclusion.

CONCLUSION

La question de non prolifération des armes nucléaires dans le monde va acquérir dans les années à venir une importance plus grande encore que par le passé.
La satisfaction des besoins énergétiques pour laquelle le nucléaire devrait apporter une réponse majeure ira de pair avec des garanties rigoureuses contre le risque de dissémination des armes.
Les réflexions engagées pour renforcer le régime actuel de non prolifération vont dans ce sens mais il ne faut pas sous estimer les difficultés. Il faut éviter :
- une déviation des contrôles à l'exportation telle que l'on aboutisse à une politique d'embargo nucléaire de facto, mais il faut des contrôles suffisamment précis pour éviter toute prolifération.

Les solutions à ce dilemme sont partiellement politiques mais la techniques aussi a son rôle à jouer et je crois que notre rôle ici est d'une importance extrême à cet égard. Vos travaux cette semaine y contribueront sans aucun doute.

CONTRIBUTING TO GOALS AND CONCEPTS FOR SAFEGUARDS IMPLEMENTATION

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Abstract

Safeguards in the European Community in the 1990s can be characterized both through the way it will cope with the increased availability and use of Plutonium in the European commercial fuel cycle and through the aims and related efforts to continue to improve the effectiveness and efficiency of the safeguards operation in general.

Based on the current operation of the industry and the construction schedules for fuel cycle facilities it is expected that the routine use of plutonium in LWR (MOX) will continue to increase significantly, that three to four MOX fabrication facilities with throughputs of up to 5 t Pu/a and three reprocessing facilities with throughputs of up to 7 t Pu/a will continue to operate or commence operation. In addition the safeguards of plants to condition irradiated fuel for direct final storage will have to be developed and implemented as such plants will come on stream.

To accomplish the requirements of European law under the constraints of limited if not less than sufficient resources the issue of safeguards goals needs to be re-opened and this in view of the well-known expectations in the public domain.

At the same time the related question of safeguards concepts or approaches needs to be addressed the resolution of which largely determines the effectiveness and efficiency of the safeguards operation.

The contribution presents the main lines of the present thinking of Euratom safeguards relating to technical goals, i.e. the 'Tryptic' of characteristic amounts, times and probabilities, and considerations leading to criteria to attain such goals.

Finally, concepts, efforts and implementations are outlined to continue to improve efficiency at large fuel cycle installations in line with the necessity to gradually replace manpower by machinepower in situations of difficult access or repetitive work.

1. Introduction

Safeguards in the European Community is performed pursuant to the Euratom Treaty which sets out the well-known objectives and requirements whereby the tasks of the Commission are, inter alia, to make certain, by appropriate supervision, that nuclear materials are not diverted to purpose other than those for which they are intended but also to establish with other countries and international organisations such relations as will foster progress in the peaceful use of nuclear energy.

The implementing Chapter VII of the Treaty, notably article 77, specifies these objectives by giving the Commission clear responsibilities and wide powers in its safeguards implementation.

As part of its basic tasks the Commission is required to fulfill the safeguarding obligations stemming from supply agreements with third countries, the most important of which are those concluded with the United States, Canada and Australia. The fulfillment of these safeguarding obligations require considerable resources from Euratom safeguards and put a considerable burden on the economic operators due to the need to track the relevant nuclear materials within the Community, to fullfil the various but not always congruent retransfer conditions and to provide, as appropriate and relevant, the inventories and balances to supplier States. Euratom has noted, when implementing such requirements, that they are differing, sometimes inconsistent but show, in any case, a marked absence of international consensus. Requirements leading to multi-labelling of the same portions of nuclear material are a case in point. The progress achieved in safeguards, in the wake of the NPT, by strongly coupling safeguards with verification should, in the view of Euratom, not be jeopardized by the pursuance of concepts such as "flag control" postulating the verification of indistinguishable atoms or by the re-introduction into safeguards of objects whose uses are unverifiable.

The Treaty also requires the Commission to satisfy itself that the safeguarding obligations under the agreements concluded with the IAEA are complied with. It may be recalled that the Community has concluded three safeguard agreements with the IAEA, i.e. the Verification Agreement INFCIRC/193 between the Community, its Non Nuclear Weapon States and the IAEA, the Voluntary Offer Agreement INFCIRC/263 between the Community, the United Kingdom and the IAEA and the Voluntary Offer Agreement INFCIRC/290 between the Community, France and the IAEA. These agreements comprise protocols, subsidiary arrangements and installation attachments but also special understandings such as on Observation, Participation and Joint Teams. These regulate the day to day interaction between the IAEA and the Euratom inspectorates. Against this background of agreements, arrangements and understandings any developments in IAEA safeguards continue to be evaluated, in particular the implementation of the recently introduced internal IAEA safeguards criteria for 1991-1995.

As the peaceful use of nuclear energy in the Community is given high and critical attention in the public domain this attention has also been focused on the control activities of nuclear including safety, environment, physical protection and last, but not least, on safeguards. While this attention should be considered, in principle, as a good thing since it will contribute to - at last - removing the shroud of mystery and secrecy in nuclear, more needs to be done to explain to a wider public the safeguards goals and concepts as well as their interaction with the safeguards resources. This appears necessary in order, inter alia, to promote the understanding that the control of complex technologies requires complex control concepts and implementations whose cost and resource requirements will increase as a function of the growth of the use of these complex technologies and of the expectations put on the effectiveness, completeness and coverage of these controls. Safeguards is no exception.

This contribution to the 13th Annual ESARDA symposium of 1991 aims at providing some notes on the goals in safeguards and related concepts for the implementation of safeguards in the Community during the 1990s.

2. Scope of Safeguards in the Community

The challenge to safeguards in the European Community during the 1990s is characterized by the need to deal with the increased use of Plutonium in the nuclear fuel cycle.

Starting from the total stocks of the civil nuclear material under Euratom safeguards by the end of 1990 at some 790 installations, i.e. 83,000 t of depleted Uranium, 44,000 t of natural Uranium, 32,000 t of LEU, 13 t of HEU, 2,600 t of Thorium and 203 t of Plutonium (still mostly contained in irradiated fuel) it is expected that these stocks of Plutonium will continue to increase by some 15-20 t annually.

While the use of recycled Plutonium in LWR has now become routine and is expected to increase, three large reprocessing plants will be under safeguards by 1995 at La Hague and Sellafield to process the irradiated fuel thereby creating a shift in strategic value of the material which necessitate a corresponding strengthening of safeguards. Three projects for new MOX fabrication facilities are in the design and construction phase. It is, again, expected that these plants will be operational by 1995.

The further construction and commissioning of interim fuel storages and waste conditioning plants which are presently planned in France, Germany and the United Kingdom will also increase the safeguards tasks and resource needs once these installations are operational. Major problems of concepts or approaches are not expected.

Pilot projects are in the planning stage for the compacting and final storage of irradiated fuel. While it is not expected that major facilities of this type will become operational prior to the year 2000 it should be noted that, concepts for credible safeguards for such final storages need still to be developed.

The resource requirements to effectively safeguard the above mentioned significant increase of the Euratom safeguards tasks have been estimated and, referring to the contribution of Mr. Maniatopoulos, the Commission has established a medium-term plan which needs to be adopted and implemented.

3. Safeguards Goals, Criteria and Guidelines

Relating to safeguards goals there appears to be a consensus that these goals comprise a so-called "tryptic", i.e. characteristic quantities to be detected, characteristic times describing the maximum response times of a safeguards system and characteristic probabilities describing both the risk of a false alarm and the risk of non-detection of the amounts within the specified time.

Whereas both the Agency and Euratom safeguards implementation are based on a methodology using such safeguards goals, the numerical values of which are well-known, the thinking on the further development of these goals and their translation into safeguards criteria give rise to observations.

No doubt, the establishment of quantified safeguards goals are required to allow the development of sampling plans, inspections schedules and resource planning or more generally, of safeguards logistics. But it continues to be doubtful that these goals would provide sufficient parameters to allow decisions in the safeguards reality.

Euratom has found over and over again that these goals and related mathematical/statistical models, e.g. for the evaluation of material balances define and describe the majority of the aspects of the complex structure of the information declared to and obtained by the safeguards inspectors at best in a very macroscopic mode (e.g. at less complicated installations).

Moreover, the contribution of un-quantifiable safeguards methods (e.g. C & S) to the attainment of these quantified goals could, by definition, not be quantified. On the other hand, quantified goals allow the establishment of quantified attainment criteria.

One of the particular difficulties in safeguards is constituted by the limitation of resources or, more precisely, by the time delays between the justification for, the recognition of and the actual disponibility of the necessary resources for the safeguards operation. The issue in need of further clarification is whether safeguards when faced with such (inevitable) time lags should adapt its goals, attainment criteria or inspection guidelines and, if yes, which of these. The answer is not obvious but it may be noted that the least adequate solution seems to be to create a vicious cycle between lack of resources and relaxation of goals, criteria or inspection guidelines since this would result in the deterioration of safeguards credibility with related consequences to nuclear.

The IAEA has issued safeguards criteria for 1991-1995 and Euratom services are in the process of analyzing these with respect to methodology, compliance with the agreements and special understandings and compliance with facility attachments as well as with basic Community policy such as the unity of the European nuclear market. Once this analysis is completed it is intended to hold consultations with the Agency prior to the decision to which extent Euratom can support the implementation of these internal IAEA criteria.

As far as, however, the methodology of safeguards is concerned, Euratom believes that for its safeguards under the Treaty the establishment of safeguards criteria is less than useful or adequate. Apart from the points related to quantification referred to above, Euratom considers that safeguards criteria of the kind issued might be suitable for a formalized and centralized evaluation-oriented safeguards system whereas (safeguards) inspection guidelines of the type under development appear suitable for decentralized and flexible decision-oriented systems, making, inter alia, full use of the experience of long-term safeguards inspectors.

4. Concepts and Approaches

The increased availability and use of Plutonium in the European commercial fuel cycle in the 1990s combined with the need to maintain the present high standards achieved in the safeguards development and operation necessitate the further improvement of safeguards efficiency and the related refinement of safeguards concepts and approaches.

Improvements of the efficiency in safeguarding Uranium are expected to be achieved gradually through the introduction of improved instruments and equipment and through rationalization measures adapted specifically to the intermittent inspection regime which is exclusively applied at the installations in this part of the fuel cycle.

Developments to be mentioned are the progressive introduction of new C&S equipment, e.g. using front end motion detection, the improved methods used for item verification, the shifting from DA to NDA and, last but not least, improved on-site data ,handling and evaluation. The implementation of "Random" inspections - while mathematically quite elegant - is being tested by Euratom in an operational production plant but first results are less than encouraging.

Thus, in the Uranium part of the fuel cycle including concentration, conversion, fabrication and enrichment facilities - the latter require slightly different approaches due to the inherent commercial and non-proliferation sensitivity of the technology used - as well as for LWR-LEU no significant changes are envisaged but gradual increases of efficiency.

For LWR-MOX the safeguards concepts continue to be based on item verification techniques, i.e. based on the verification of the integrity and identity of distinguishable fuel elements and, notably, on the use of C&S and video surveillance techniques during all phases of reactor operations. These concepts have been implemented requiring, however, still considerable inspection effort. It is expected that this inspection effort can be reduced when either the time interval between fresh MOX arrival and core loading can be reduced to not more than a few weeks or, alternatively, the equipment allowing conclusive under water measurement will become fully operational.

Safeguards of MOX fabrication plants and reprocessing plants will continue to require continual inspectors presence.

Based on consultations with operators in the Community and, to some extend, with the Agency, modern safeguards systems were developed and are progressively being implemented.

Under the aspect of safeguards concepts no dramatic new developments can be reported since the well-known concepts such as:

- verification of BTC during the construction phases and later re-verification
- verification of all input and output streams;
- verification of, at least monthly,of the hold-ups using various techniques or FBOM
- maintenance of the continuity of knowledge in the product stores

are being used or envisaged for the new plants.

As far as, however, the approaches are concerned the main features of the safeguards systems for the new plutonium plants are a significant shift from inspector attended to unattended measurement or surveillance operation.

This is caused, inter alia, by

a) the need to minimize radiation exposure of plant personnel and inspectors

b) the need to minimize stoppage for routine safeguards purposes of automated production and by

c) the requirement to use identical or similar components in all plants so as to minimize development costs and to maximize standardization.

The effect is a transition from operations costs to investment costs which, albeit expensive initially, are expected to "pay off" within a limited break even time. These systems are also expected to minimize repetitive inspectors work thereby further contributing to enhanced safeguards effectiveness.

5. CONCLUSIONS

Safeguards in the European Community in the 1990s can be characterized both through the way it will cope with the increased availability of and use of plutonium and through the approaches aimed at improving the efficiency of the safeguards operation.

The discussion on safeguards goals, criteria or inspection guidelines needs to be re-opened and their interaction with the availability of resources needs to be clarified.

For the new large plutonium plants, modern safeguards systems have been developed and are under impelementation which, while using known concepts, provide for a shift from operation costs to investment costs due to the introduction of unattended measurement and surveillance stations.

Given these developments and their progressive implementation it is expected that the required rationalisation of Euratom safeguards will take place while maintaining its effectiveness.

A SCHEME FOR RANDOMIZED INSPECTIONS*

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Abstract

We describe a general randomized inspection scheme that satisfies initially stipulated quantitative requirements on detection probabilities and timeliness for nuclear material safeguards verifications, and at the same time makes more efficient use of inspection resources. The scheme reduces intrusiveness into the operations of the inspected plant and strengthens the effectiveness of the inspection due to the unpredictability and the increased intensity of the inspections.

The same formalism is applicable to sequential inspections, inspections by strata, and inspections of facilities over a large complex, such as a country. The methodology is applicable to a much wider class of inspections including arms control verification.

Introduction

The IAEA has been investigating the randomization of inspections as a means of optimizing their increasingly limited resources. Markin 1988 /1/ and 1989 /2/ discussed the random allocation of inspection resources among facilities and the various strata of material. Fishbone and Nagele 1990 /3/ considered several practical aspects of randomized inspections, including confidentiality, detection probability, and the problems of associated logistics. The implementation of short-notice, random inspections (SNRI) is being tested at the General Electric (GE) Low-Enriched Uranium (LEU) fuel fabrication facility (Eberhard and Kessler 1990 /4/). Canty, Stein and Avenhaus 1988 /5/ examined the relation between measurement error, false alarm rate and randomized inspections. Previously, Gordon and Sanborn 1984 /6/ examined some aspects of randomization of flow verifications for enrichment plants. In particular, they emphasized the importance of pre-declaration of flow and inventory in randomized inspection schemes.

In this paper, the Draft IAEA Safeguards Criteria for the Period 1991-1995 (the Criteria), which will govern the implementation and evaluation of the attainment of inspection goals by the IAEA, are used as the baseline to consider randomized inspections. We propose and examine a randomized scheme which can, at a minimum, satisfy the Criteria and reduce the required IAEA inspection resources. We note that the Criteria can be satisfied, even though flow verification is incomplete because of short material-residence time. Incomplete flow verification reduces the effectiveness of the IAEA safeguards, since some of the material is not safeguarded. The proposed randomized inspection scheme can improve the completeness and timeliness of flow verifications.

Our approach differs from others in that in our scheme the concept of randomization has been integrated into the overall inspection design to achieve a certain set of inspection goals in terms of timeliness and detection probability. In most other approaches a randomization is considered in ad-hoc manner: each inspection procedure is designed without first considering its application in a regime of randomized inspections. Thus, it is necessary to assess the impact of randomization on inspection goal attainment, safeguards conclusions and evaluation criteria after the fact. In our scheme, on the other hand, the concept and practice of randomization is included in the inspection design a priori.

The central point of our paper is that the factors to be considered in the design of inspections include: times, facilities, material balance areas, material strata and items, and that all of these elements can be addressed on an equal footing. Our randomization approach can be applied to the entire complex of these elements, and the (interrelated) procedures applied to the individual elements are so chosen that the overall, initially stipulated (a priori) detection probability and timeliness criteria for the inspections as a whole are satisfied at each inspection opportunity. In contrast the current regular (non-randomized) IAEA inspection randomizes only over the items, but not over the other elements cited above. In this way, our approach can be viewed as an extension of the current IAEA regular inspection regime.

In the proposed scheme, an inspection has two stages:

(i) a randomized scheme for deciding whether an actual inspection is to be carried out, followed by

(ii) an enhanced sampling program for the inspections which are conducted.

This combination will achieve the same overall probability of detection as the conventional scheme. When the inspectors go to a facility, they will make more measurements than in the current scheme. On other occasions, they will remain at headquarters; nevertheless, the scheme will attain the same overall detection probability. The potential savings in resources come from the omitted inspections and their associated overhead, including travel, opening and closing meetings, health and safety procedures, and instrument preparations. On the other hand, there will be increased inspection when inspectors do go to the facilities. We believe that the decrease in effort from omitted trips will exceed the increase in effort during the inspections that are performed. (For example, about 30% of the trips to an LEU fabrication and conversion facility may be saved as against about a 20% increase in sample size to be measured when inspections are performed.)

It is important to emphasize that in this scheme the onset of an actual inspection is determined randomly, before any requirement for timeliness or other interim scheduling. The overall inspection goal is attained because the onset of an inspection is randomly determined *a priori*, and the relevant sampling plan for that inspection is designed to satisfy the

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Criteria in a timely manner. We also point out that the current IAEA verification scheme is a special case of the new, more general scheme proposed; it is one particular way of satisfying the Criteria, but it is not the only way.

In addition, we will show that the proposed scheme not only achieves the detection probability goal at each inspection, but is also able to detect the diversion of a goal quantity of material distributed randomly throughout the year.

Draft Safeguards Criteria

In this section, we review the Draft IAEA Safeguards Criteria relevant to randomized inspections. Since the Criteria will be used to govern the implementation of safeguards activities in the field, and the evaluation of inspection goal attainment by the IAEA, the Criteria serve as necessary and sufficient conditions for satisfactory safeguards. Our discussion will concentrate on the Criteria for LEU conversion and fabrication facilities. Such LEU facilities are good candidates for considering randomized inspections because of the large material flows, the short residence time of the materials, and the relatively low goal for detection probability in the Criteria. However, the validity of the principles discussed in this note is not limited to LEU facilities. For example, power reactor facilities with fresh mixed oxide (MOX) or spent fuel also will be good candidates for such randomized inspections. Because of the short timeliness requirement when fresh MOX or spent fuel is present, many interim inspections are required (11 per year when fresh MOX fuel is present, 4 per year with only spent fuel). Although the decrease in inspection effort per reactor may be small, the total potential savings for the IAEA may be appreciable, since there are many power reactors operating under IAEA safeguards.

The Criteria stipulate that for LEU conversion and fabrication facilities there is one physical inventory verification (PIV) per year and 5 interim inspections on a bimonthly basis to verify material transfers to and from the facility. (For some other types of facilities, interim inspections are performed to satisfy the timeliness requirements.)

During an inspection, LEU is verified with medium detection probability (about 50%) for gross, partial, or bias defects (depending on the type of material) for the diversion of one significant quarterly (SQ) of the material. This inspection serves the IAEA safeguards objective for "...the timely detection of diversion of significant quantities of nuclear material...and deterrence of such diversion by the risk of early detection."

Operationally, the detection probability requirement is translated mathematically into a sample size requirement via:

$$n = N(1-\beta^{1/m}) \qquad (1)$$

where N is the total number of items of interest at the inspection, m is the number of defective items making up an SQ, 1-β is the detection probability as required in the Criteria, and n is the number of items that must be verified to satisfy the Criteria. Implicit in this formula is the assumption that if a defective item is included in the sample, it will be detected. It has been shown that for inspections in which sample sizes are calculated according to this formula, the scheme can also detect the diversion of 1 SQ distributed throughout the year with the same detection probability, 1-β.

The Basic Randomized Inspection Scheme

First, the overall required detection probability, 1-β, remains the same (medium detection probability, about 50%). The timeliness requirement, or interim inspection schedule, is also preserved. A few days before the scheduled inspection, inspectors will decide if the actual inspection will be carried out, with probability p, $0<p\leq 1$. (As shown below, p must lie between 1-β and 1, and a different value can be selected for each scheduled inspection.) After p is selected by the inspectors, they then use a random number generator which generates random numbers uniformly between 0 and 1. If the random number obtained is less than or equal to p, the scheduled inspection will actually be carried out; if it is greater than p, the actual inspection will be omitted.

Let n' be the number of random samples selected for verification during the inspections actually carried out, N the total number of items in the population and m the number of defective items making up 1 SQ. The total non-detection probability, B, for the randomized inspection scheme can then be expressed as :

$$B = 1 - p + p\,\frac{{}_{N-m}C_{n'}}{{}_{N}C_{n'}} \qquad (2)$$

where ${}_{N}C_{n}$ is the number of combinations of N items taken n at a time. The non-detection probability, B, at this inspection is the sum of two terms:

(i) the probability that the inspection is not carried out, 1-p, and

(ii) the product of the probability that the actual inspection is carried out and the probability that none of the defective items is included in the n' samples.

For the IAEA inspections, B is required to be less than values of β as specified in the Criteria. The exact sample size in this scheme can then be calculated from Eq. (2).

After a variable transformation,

$$\beta' = \frac{\beta+p-1}{p} = 1 - \frac{1-\beta}{p} \qquad (3)$$

one sees that, analogous to the derivation of the original sample size formula, Eq. (1), the sample size in the randomized inspection scheme can be approximated by:

$$n' = N(1-\beta'^{1/m}) \qquad (4)$$

Thus, in this scheme the sample size depends on the probability of actually carrying out an inspection, p. The higher the p, the smaller the sample size. When p = 1, the formula reduces to the original IAEA formula, Eq. (1), as it should when all the scheduled inspections are carried out. However, in the new, more general scheme, not all the scheduled inspections need be carried out. Instead, whether a scheduled inspection is to be actually carried out is determined randomly before the inspection, with a probability p for actually carrying out the inspection. The cost of this scheme is that when an inspection is actually carried out, more items than in the original approach must be sampled for verification. This will most likely be counter balanced by the benefits from carrying out fewer actual inspections.

It can be seen from Eq. (2) that p must be greater than 1-β. Thus, if one would like to have a 1-β detection probability, then p (the probability for carrying out an inspection) must be at least 1-β. Within this constraint, inspectors are free to choose a different p at each scheduled inspection.

The basic randomized inspection scheme described above can be carried out easily and it will, on the average, reduce the resources required for IAEA inspections. A fraction, 1-p, of the scheduled inspections may not be carried out, at a cost of some additional sampling effort at the fraction p of the scheduled inspections which are carried out. This latter fractional increase of inspection effort will be (at most) pf, where f is the fractional increase in sampling effort. Under most circumstances, the net effect should save resources for the IAEA[1]: the average fractional net savings become

$$(1 - p) - pf = 1 - p(1 + f).$$

The definitions and relations contained in Eq. (3) above have a useful physical interpretation. Just as β and 1 - β = D represent the originally stipulated non-detection and detection probabilities, respectively, so do β' (given by Eq. (3)) and

$$D' = 1 - \beta' = \frac{1 - \beta}{p}$$

represent the "effective" non-detection and detection probabilities, respectively, in the randomized inspection scheme described here.

Distributed Diversions

The randomized inspection scheme satisfies the Criteria at each inspection. A desirable feature of the current IAEA scheme is its ability to detect with the same probability, 1-β, a diversion of 1 SQ distributed throughout the year. We show below that the proposed randomized inspection scheme preserves this feature.

In the case of the proposed randomized inspection scheme, the sample size, n_i', at the i-th inspection with total population N_i is:

$$n_i' = N_i(1 - \beta_i'^{\,1/m}) \qquad (5)$$

where, again, $\beta_i' = (\beta + p_i - 1)/p_i$, and $p_i \geq 1-\beta$. It follows that:

$$1 - \frac{n_i'}{N_i} = \beta_i'^{\,1/m} \qquad (6)$$

When the sample size is determined as described, and when the total number of defective items is m_i (instead of m) during this inspection period, the non-detection probability, B_i, for the i-th inspection can be expressed as:

$$B_i = 1 - p_i + p_i \prod_{j=1}^{m_i} \left(1 - \frac{n_i'}{N_i - j + 1}\right) \qquad (7)$$

$$\leq 1 - p_i + p_i \prod_{j=1}^{m_i} \left(1 - \frac{n_i'}{N_i}\right)$$

$$= 1 - p_i + p_i \beta_i'^{\,m_i/m}$$

$$\leq (1 - p_i + p_i \beta_i')^{m_i/m} \quad \text{(Sanborn 1987 /7/, Lemma 3)}$$

$$= \beta^{m_i/m}$$

Summing over all the inspections with

$$\sum_{i=1}^{T} m_i = m,$$

(where T is the total number of inspections), the total non-detection probability is

$$B = \prod_{i=1}^{T} B_i \leq \prod_{i=1}^{T} \beta^{m_i/m} = \beta$$

Thus the proposed randomized inspection scheme could also detect the diversion of 1 SQ distributed randomly throughout the year, with the same probability, 1-β.

Estimate of Savings

We can estimate the savings in the following way. For each facility, the expected number of inspection trips not undertaken is the product of total number of inspections scheduled, T, times the probability of not conducting the inspection, 1-p. For now we assume that the application of the randomized scheme is limited to interim inspections only, while the PIV is still carried out as usual; this is a conservative approach. Let the inspection man-days (MDI) per interim inspection be d, and the fraction of time spent on measurements during an interim inspection be q. Then the expected savings per year for this scheme is at least:

$$S = [\,1 - p - p\,q\,(\frac{n'}{n} - 1)]\,T\,d \qquad (8)$$

where n' and n are the samples sizes calculated according to Eqs. (4) and (1), respectively, for a total population N. The savings depend on the value of p. The limit on p is p≥1-β. The estimates can be calculated for each type of facility and the savings for the IAEA as a whole then can be estimated by summing Eq. (8) over all facilities under safeguards.

Any value of p≥1-β will do. However, if p=1-β, then at least N-m+1 items need to be measured to ensure that at least one defect will be detected. This approach might not be desirable under some circumstances (e.g., many small items). To get a feeling of the estimated savings, the following estimates were calculated with

$$p = \sqrt{1-\beta}, \qquad (9)$$

and

$$p = (1-\beta)^{0.9} \qquad (10)$$

respectively; both equations satisfy the requirement that p is not less than 1-β.

For LEU facilities or light water reactors (LWR) with spent fuels, about 30% of the scheduled inspection trips may be omitted when Eq. (9) is used to determine the probability

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[1] Since $f \leq \frac{1-p}{p}$ in all practical cases.

of carrying out an inspection, while for each of the inspections actually carried out, the sampling fraction is increased from approximately 50% to 70%. When Eq. (10) is used, about 45% of the inspection trips may be omitted, while the sampling fraction is increased from about 50% to 93%.

Thus, the estimated savings in total inspection resources for large-scale LEU conversion and fabrication facilities are about 12% to 20%, and for LWRs with spent fuel but without fresh MOX, the savings are between 20% and 30%. Since there are about 100 LWRs and 8 LEU facilities, the total savings are about 250 man-days for LWRs, and 60 man-days for LEU facilities. The savings would allow IAEA more opportunities for interim inspections, and thus, more complete coverage of flow verifications.

Another possible formula for the selection of p_i is:

$$p_i = (1-\beta)^{(1-N_i/N)} \qquad (11)$$

The model fits the criterion that $p_i \geq 1-\beta$. Here, the more material available for inspection, the higher the probability for actually carrying out that inspection, and hence, the smaller the sample size required during that inspection.

Selection of Strata or Facilities

In some situations, it might be preferable to select a few strata for verification instead of verifying all of them. The methodology presented here can be readily extended to such applications. Consider first the current IAEA regime where all inspections are actually carried out; this corresponds to p=1 in Eq. (2).

However, instead of selecting n (Eq. (2) with p=1) samples from all strata for verification, each strata is assigned a probability q for verification. Then n_q samples need to be chosen from the strata randomly selected for verification with probability q. The non-detection probability for each stratum can be expressed as:

$$B_q = 1 - q + q\,\frac{{}_{N\text{-}m}C_{n_q}}{{}_{N}C_{n_q}}$$

Since B_q is required to be less than β, n_q can be approximated by:

$$n_q = N(1-\beta_q{}^{1/m}) \qquad (12)$$

where

$$\beta_q = \frac{\beta-1+q}{q} \qquad \text{with } q \geq 1-\beta.$$

The random selection of strata for verification is also applicable in the basic randomization scheme. The only modification required is to substitute β' for β in the definition of β_q.

The basic randomization scheme also can be applied to the randomization over facilities (e.g., within a country), in order to select facilities for actual inspections, in a manner similar to the randomization over material. The formalism is exactly the same as that described above.

Summary

To summarize, our approach has the following features:

- it satisfies the stipulated quantitative criteria for detection probability and timeliness;
- it uses the inspection resources more efficiently by decreasing the indirect (overhead) costs, while intensifying the actual inspections;
- it reduces interference with plant operations because of the lower frequency of actual plant inspections; and
- it has an increased deterrence on potential diversions because the inspection times are unpredictable and enhanced actual inspection intensity.

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Optimal Randomization Strategies for Timely Verification

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ABSTRACT

Game-theoretical models of randomized inspections for timely verification of compliance are investigated. Guaranteed average detection times comparable to or shorter than those obtainable without randomization are calculated.

1. Introduction

In this paper we examine the following idealized model of interim inspections for timely verification of non-diversion: A reference time period is subdivided into n equal intervals, at the end of each of which an interim inspection can take place. An inspection is assumed *always* to take place after the nth interval, i.e. at the end of the reference period, and the total number of inspections, including the last, is defined to be $k \leq n$. It is assumed, furthermore, that *at most* one diversion will occur, and if it occurs, it will be detected at the next inspection.

For the case $k < n$, we formulate a two-person zero-sum game between plant operator and inspector with the payoff to the former, in case of illegal behavior, being the expected time to detection as measured from the time of diversion. In case of legal behavior, since no diversion occurs and since we exclude the possibility of false alarms, the time to detection is undefined. Formally, therefore, the operator's optimal strategy may well be simply to behave legally. Rather than introducing subjective utilities into the payoff function to deal explicitly with this option (and thereby complicating the analysis unnecessarily) we can give the following technical justification for considering the illegal game: we seek a ***subgame perfect equilibrium*** of the overall two-person game (see e.g. /1/), i.e. a strategy for the inspector which is also optimal in the case in which the operator, perhaps against his better judgement, chooses to act illegally within the reference period.

Thus we consider only the illegal subgame. In order to stress this we shall, in the sequel, refer to the operator as the 'diverter'.

Two versions of the game are considered. In the first, k is interpreted as the precise number of inspections to be carried out, so that an inspector's strategy is to choose a subset of $k-1$ interim inspections out of $n-1$ possibilities. We also require that the diversion strategy be chosen prior to the reference period, and that it be irrevocable. The resulting ***matrix game***, by virtue of von Neumann's ***minimax theorem*** /2/, possesses a saddlepoint in the domain of mixed strategies, i.e. for randomized inspections. Numerical solutions may be calculated using the simplex algorithm.

In the second version, k is treated as an expectation value for the number of inspections, when the interim inspections are performed according to a probability distribution over the n periods. That is, if p_i is the probability that an inspection takes place after interval i, then $k = \sum_1^n p_i$. It is demonstated that, if both players are ignorant of their opponent's strategies, the game has no saddlepoint. The related ***inspector leadership game***, in which the inspector announces his strategy in advance, does however have an equilibrium, which is derived.

The game theoretical results are presented in Section 2. The solutions are discussed in relation to applicability to interim inspection problems in Section 3.

2. Game Theoretical Results

Let the total number of inspections that are to take place in the reference time n be $k < n$. As indicated in the introduction, we suppose that inspections are such that prior diversions will be detected with certainty at the first subsequent inspection and that an inspection *always* takes place at $i = n$. Suppose further that the diverter is aware of this and of the number k of inspections that will be performed.

Since detection is inevitable, the diverter can at best choose his diversion time in such a way as to *maximize* the time to detection. He has precisely $n - 1$ pure strategies at his disposal, namely to violate just after time $i, i = 0 \ldots n-2$. (Time $n - 1$ can be excluded, since this will always lead to the minimum detection time of one period.)

2.1. THE MATRIX GAME

We treat the problem first as a finite game. The diverter chooses his strategy irrevocably prior to the start of play, i.e. at time $t < 0$. He cannot change his mind during the course of the game. (We shall return to this point later.) The value k is taken to be exact, that is *precisely* k inspections must take place within the reference time n. Thus the inspector has $\binom{n-1}{k-1}$ pure strategies, recalling that one inspection is always reserved for time n, and he will choose his strategy such as to *minimize* the time to detection. Each strategy combination of diverter and inspector leads to a unique detection time which is an integral number of inspection opportunity intervals. This suffices to define a *two-person, zero-sum matrix game* between diverter (player I) and inspector (player II) with the payoff to the diverter being the time to detection after diversion. The payoff matrix is a matrix whose rows and columns are labelled by the pure strategies of players I and II, respectively. The corresponding matrix elements are the detection times for each strategy combination.

The payoff matrices constructed in this way do not possess saddlepoints, that is, there is no pure strategy combination (i^*, j^*) for players I and II, respectively, which satisfies

$$L_{i,j^*} \leq L_{i^*,j^*} \leq L_{i^*,j} \qquad \forall i,j$$

where $L_{i,j}$ is an element of the payoff matrix. However the *minimax* theorem of von Neumann /2/ guarantees the existence of a saddlepoint solution in the domain of mixed strategies, where a mixed strategy for a player is a probability distribution over his set of pure strategies. A mixed strategy for player II, the inspector, is thus an *inspection randomization* strategy. For small matrices, the optimal mixed strategies (solutions of the game) can generally be calculated by hand. A graphical solution for the simplest non-trivial case $n = 3, k = 2$ is shown in Figure 1. For many cases of interest, however, the matrix can be quite large and only numerical solutions are possible. For example for $n = 12, k = 6$ the game matrix is of dimension 11 × 462.

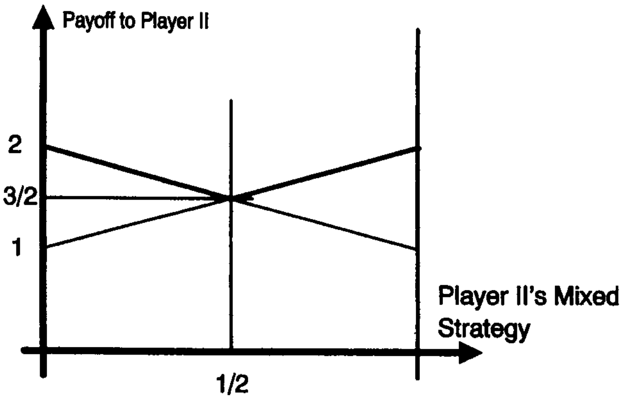

Fig. 1: Graphical solution of the matrix game $n = 3, k = 2$.

The optimal inspection randomization strategies and the associated guaranteed detection times for all values $3 \leq n \leq 12$ with $2 \leq k \leq n-1$ have been calculated numerically using the simplex algorithm and are tabulated in /3/. The guaranteed detection times L are interesting from the point of view of inspection efficiency since they satisfy

$$\begin{aligned} L &= \frac{n}{k} & k &= n-1 \\ L &< \frac{n}{k} & 1 &< k < n-1 \end{aligned} \tag{2.1}$$

This means that, by performing k random inspections over the n opportunities, the inspector can obtain *shorter* detection times (at least for $k < n-1$) than by splitting the reference period into k equal intervals and inspecting *with certainty* after each interval. (This latter strategy gives of course a detection time of exactly $\frac{n}{k}$.) For fixed inspection effort, a randomization strategy is thus superior to a deterministic one. We shall discuss a practical application of this in Section 3.

The disadvantage of the matrix game approach is that it requires, as we have already indicated above, that player I choose his strategy prior to the reference period without taking into account any information about the inspector's strategy gained in the course of the game. One could imagine that, knowing both n and k, the diverter would wait for a few periods in order to observe how many inspections had been 'used up', or identify the pure strategy pattern chosen, and then act. Using such a *behavioral strategy* (which is of course also selected prior to the reference period) the diverter might, with some justification, hope to push the detection time beyond the saddlepoint value of the game considered here. This is not to say that the solution has no practical application. Depending on external circumstances the diverter may have no choice in timing his action (e.g. he may have to plan well in advance).

Alternatively, the diverter may be ignorant of the number k of inspections. This would be the case if, for instance, not the precise value but only the expectation value of k is known to the operator. This situation is considered in the following section.

2.2. THE INSPECTOR LEADERSHIP GAME

Suppose the inspector chooses to inspect at the ith opportunity with probability p_i, whereby the p_i are independent. Then the expected value of the number of interim inspections carried out (i.e. excluding the last one) is

$$\sum_{i=1}^{n-1} p_i = k-1. \tag{2.2}$$

Thus a fixed k is to be interpreted now as an *expectation value* and need no longer be an integer. Again assuming that an inspection, if it takes place, will always detect a prior diversion with certainty, we can write down the following recursive expression for L_i, the expected time to detection of a diversion at the beginning of the ith interval:

$$\begin{aligned} L_i &= 1 + (1-p_{i+1}) \cdot L_{i+1}, \qquad 0 \le i < n-1 \\ L_{n-1} &= 1. \end{aligned} \tag{2.3}$$

It is convenient to consider first the special case $n = 3$. We will generalize to arbitrary n later. For this case, equation (2.2) becomes

$$p_1 + p_2 = k-1, \qquad 1 \le k \le 3 \tag{2.4}$$

with the restriction

$$\max(0, k-2) \le p_{1(2)} \le \min(1, k-1). \tag{2.5}$$

From equation (2.3) we obtain

$$\begin{aligned} L_0 &= 1 + (1-p_1)L_1 \\ L_1 &= 1 + (1-p_2)L_2 \\ L_2 &= 1. \end{aligned}$$

Defining $p_1 = p$, and with (2.4), $p_2 = k - p - 1$,

$$\begin{aligned} L_0 &= 1 + (1-p) \cdot (3-k+p) \\ L_1 &= 3-k+p. \end{aligned} \tag{2.6}$$

Let $q = q_0$ be the probability that the diversion occurs at time 0. Then the probability of diversion at time 1 is $q_1 = 1 - q$, since, as already noted, it would not be sensible for player I to time his diversion at $i = n - 1 = 2$, this leading to the minimum detection time of one period. The average time to detection can therefore be written

$$\begin{aligned} L(q,p;k) =& q \cdot L_0 + (1-q) \cdot L_1 \\ =& q \cdot (1 + (1-p) \cdot (3-k+p)) \\ &+ (1-q) \cdot (3-k+p). \end{aligned} \tag{2.7}$$

We can now formulate the problem of determining the optimal inspection strategy as solving a two-person, zero-sum *infinite* game $< L, q, p >$ where the payoff function L is given by equation (2.7) and the strategy sets of players I and II satisfy, respectively, $0 \le q \le 1$ and $\max(0, k-2) \le p \le \min(1, k-1)$. (The game is infinite since the inspector has a continuum of pure strategies p available to him.) We seek q^* and p^* such that the *equilibrium* or *saddlepoint conditions*

$$L(q,p^*;k) \le L(q^*,p^*;k) \le L(q^*,p;k) \tag{2.8}$$

are satisfied.

Unlike the finite matrix game considered before, there is no guarantee that a saddlepoint exists. In fact a saddlepoint exists if and only if

$$\min_p \max_q L(q,p;k) = \max_q \min_p L(q,p;k)$$

and we shall show that this is in general not the case. We begin by stating

Theorem 1: *Define* $L(q^*, p^*; k) = \min_p \max_q L(q, p; k)$, *where* $L(q, p; k)$ *is given by (2.7). Then for* $3/2 < k \leq 3$

$$\begin{aligned} L(q^*, p^*; k) &= \frac{2}{k - 3 + \sqrt{(k-3)^2 + 4}} \\ q^* &= 0 \text{ or } 1 \\ p^* &= \frac{1}{L} \end{aligned} \tag{2.9}$$

and for $1 \leq k \leq 3/2$

$$\begin{aligned} L(q^*, p^*; k) &= 5 - 2k \\ q^* &= 1 \\ p^* &= k - 1. \end{aligned} \tag{2.10}$$

Proof: From (2.7) we can write L(q,p;k) as

$$L(q, p; k) = q(1 - p(3 - k) - p^2) + 3 - k + p. \tag{2.11}$$

Since L is linear in q, it will be maximized for $q = 1$ or $q = 0$ depending upon whether the coefficient of q in (2.11) is positive or negative, respectively. This in turn depends upon the value of p. The coefficient of q in (2.11) changes sign for $p = p^*$ satisfying

$$1 - p(3 - k) - p^2 = 0$$

or

$$p^* = \frac{k - 3 + \sqrt{(k-3)^2 + 4}}{2} \tag{2.12}$$

being positive for $p < p^*$ and negative for $p > p^*$.

Thus we have

$$\begin{aligned} L(q^* = 0, p; k) &= 3 - k + p, \quad p \geq p^* \\ L(q^* = 1, p; k) &= 2 + (2 - k)(1 - p) - p^2, \quad p \leq p^*. \end{aligned} \tag{2.13}$$

For $3/2 < k \leq 3$ this function takes its minimum wrt p at $p = p^*$. (See Fig. 2.) Substituting for p^* in either of the expressions (2.13), we get

$$L(q^*, p^*; k) = \frac{2}{k - 3 + \sqrt{(k-3)^2 + 4}} = \frac{1}{p^*}$$

which is (2.9) as required.

For $k \leq 3/2$ the situation is as shown in Fig. 3 where, by virtue of (2.5), $0 \leq p \leq k - 1 < 1/2$. Now the violator will always choose $q^* = 1$ (upper curve) and the minimum wrt p occurs with p set to its maximum allowed value of $p^* = k - 1$. Substituting this value into $L(q^* = 1, p; k)$ in equation (2.13), we get

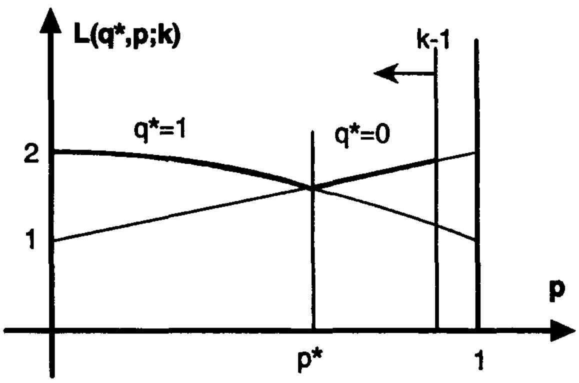

Fig. 2: Illustrating the situation of Theorem 1 for $3/2 < k \leq 3$.

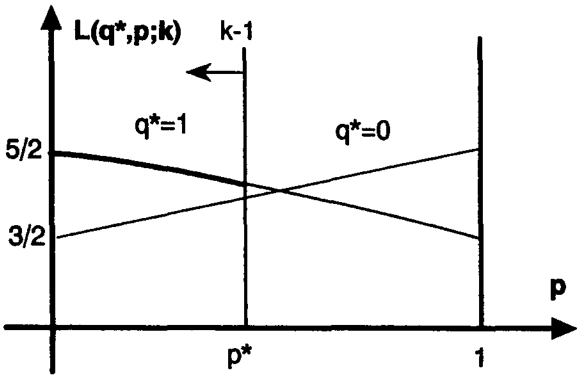

Fig. 3: Illustrating the situation of Theorem 1 for $1 \leq k \leq 3/2$.

$$L(q^* = 1, p^* = k - 1; k) = 5 - 2k$$

which is (2.10), and the proof is complete.

We note the following properties of Theorem 1:

i) The condition for validity of solution (2.9) can be written

$$n/2 < k \leq n/1$$

where $n = 3$. For k satisfying this condition, the diverter is *indifferent* to strategies $q = 1$ and $q = 0$, i.e. to diversion at times 0 or $1 = n - 2$.

ii) Similarly, the condition for solution (2.10) can be written

$$n/n \leq k \leq n/2$$

and now the diverter chooses $q = 1$, that is he does not consider diversion at time $n-2$, knowing that k is so small that he can do better than achieving a detection time of 2 periods. We shall see shortly that these results can be generalized.

That a saddlepoint need not exist can be easily seen by considering the special case $n = 3, k = 2$. From (2.7)

$$L(q,p;2) = q \cdot (2 - p^2) + (1 - q) \cdot (1 + p).$$

Since this equation is quadratic in p with nonnegative slope at $p = 0$,

$$\left[\frac{dL}{dp}\right]_{p=0} = 1 - q \geq 0,$$

it can take its minimum wrt p only at $p = 0$ or $p = 1$. At these points we have

$$L(q,0;2) = 1 + q$$
$$L(q,1;2) = 2 - q.$$

and it follows that

$$\min_p L(q,p;2) = \begin{cases} 1 + q, & \text{for } q \leq 1/2 \\ 2 - q, & \text{for } q \geq 1/2 \end{cases}$$

The maximum of this function wrt q occurs for q satisfying $1+q = 2-q$. Thus $q = 1/2$ and $\max_q \min_p L(q,p;2) = 3/2$, whereas from (2.9) we obtain

$$\min_p \max_q L(q,p;2) = \frac{2}{\sqrt{5}-1} > 3/2 = \max_q \min_p L(q,p;2)$$

and there is no saddlepoint.

In non-cooperative games, equilibrium (or for a 2-person zero sum game the saddlepoint) is the only available solution concept. In order to calculate the inspector's randomization strategy and his associated guaranteed detection time, we therefore need a new game theoretical model. This is provided by a change of rules. Rather than the players not informing one another of their strategies, we assume that the inspector ***announces his strategy in advance***, that is his inspection probabilities $p_i, i = 1 \ldots n$. We call this the ***inspector leadership game*** and it is obviously realizable in an actual inspection regime. In this model, the equilibrium solution for both players is the *minmax* solution given by Theorem 1. This can be seen by means of backward induction as follows: Player II (the inspector) announces his strategy, p^0 say, in advance. Player I (the diverter) will then choose his strategy q^0 so as to maximize the detection time, $q^0 = \arg\max_q L(q,p^0;k)$. Knowing this, player II will choose his p^0 so as to minimize $\max_q L(q,p;k)$, i.e. he announces $p^0 = \arg\min_p \max_q L(q,p;k)$.

We see then that the inspector leadership game, which is solved in Theorem 1 for $n = 3$, provides a satisfactory solution to our problem. We clearly wish to generalize Theorem 1 to arbitrary n, leading us to

Theorem 2: *Given the function*

$$L(\underline{q},\underline{p};k) = \sum_{i=0}^{n-1} q_i \cdot L_i, \qquad n \geq 3$$

where L_i satisfies

$$\begin{aligned} &L_i = 1 + (1 - p_{i+1}) \cdot L_{i+1}, \qquad i < n-1 \\ &L_{n-1} = 1 \end{aligned} \tag{2.14}$$

and where

$$0 \leq q_i \leq 1, \qquad \sum_{i=0}^{n-1} q_i = 1$$

and

$$0 \leq p_i \leq 1, \qquad \sum_{i=1}^{n} p_i = k, \qquad p_n = 1$$

then for k in the range

$$\frac{n}{r+1} < k \leq \frac{n}{r}, \qquad r = 1, 2, \ldots n-1$$

the quantity

$$L(\underline{q}^*, \underline{p}^*; k) \equiv \min_{\underline{p}} \max_{\underline{q}} L(\underline{q}, \underline{p}; k)$$

is the positive solution of the quadratic equation

$$L^2 + (kr - 2r - 1) \cdot L - r \cdot (n - r - 1) = 0 \tag{2.15}$$

with the vector $\underline{p}^* = (p_1^* \ldots p_n^*)$ given by

$$\begin{aligned} &p_i^* = \frac{1}{L}, \qquad 1 \leq i \leq n-r-1 \\ &p_{n-r}^* = \frac{(r+1-L)}{r} \\ &p_i^* = 0, \qquad n-r+1 \leq i \leq n-1 \\ &p_n^* = 1 \end{aligned} \tag{2.16}$$

and the vector $\underline{q}^* = (q_0^* \ldots q_{n-1}^*)$ by

$$(1,0,0\ldots0) \text{ or } (0,1,0\ldots0) \text{ or } (0,0,1\ldots0) \text{ or } \ldots \tag{2.17}$$

the last $n - r$ components being all 0's.

Proof: See Ref. /3/.

We note the following properties of the inspector leadership solution:

i) For $k = n/r$, that is when n is an integer multiple of k, equation (2.15) has the solution $L = n/k$. The optimal inspection strategy for the inspector leadership game gives an average detection time equal to the detection time for the deterministic strategy of splitting the reference time n into k equal intervals and inspecting with certainty after each interval (compare with Section 2.1 above). If n is not an integer multiple of k, then L exceeds n/k very slightly (see the numerical example in Section 3 below).

ii) Since according to (2.17), the diverter is ***indifferent*** as to which of the first $n-r$ periods in which to act, he cannot improve his payoff by delaying his decision. This game thus avoids the objection to the matrix game solution at the expense of longer detection times.

3. Discussion

The inspector's guaranteed detection times for the two approaches discussed in Section 2 are compared in Table 1 for the case $n = 10$ and all values of $k = 1, 2 \ldots 10$. Included in the table are the average run lengths that would be obtained if, on the average, $k-1$ interim inspections are distributed with equal and independent probabilities over the $n-1$ opputunities. From equation (2.3) one can calculate that it is then optimal for the diverter to act in the first interval. Equation (2.3) is also used to obtain the figures in the last column. Except for the trivial cases $k = 1$ and $k = 10$ the matrix game solution always gives the best result. Both game theoretical solutions are better than for equally distributed probabilities.

TABLE 1 *Comparison of solutions for* $n = 10$.

k	Matrix	Leadership	Equal Prob.
1	10.00	10.00	10.00
2	3.987	5.000	6.228
3	2.558	3.359	4.135
4	1.946	2.531	2.948
5	1.645	2.000	2.244
6	1.444	1.702	1.799
7	1.333	1.464	1.500
8	1.222	1.275	1.286
9	1.111	1.123	1.125
10	1.000	1.000	1.000

In addition to the equilibrium solutions in terms of average run lengths, the distributions of the run lengths about the average value are of interest. Monte Carlo simulations of typical distributions are shown in Figure 4. The matrix game solution is seen not only to have the lower mean detection time, but also the narrower distribution. Note also that in the inspector leadership game there is always a small but finite probability that detection will occur only after the nth period. In both cases, the probability that a detection occurs *before* the deterministic detection time n/k is greater than 50 % (although the *mean* of the distribution is less than n/k only for the matrix game solution.)

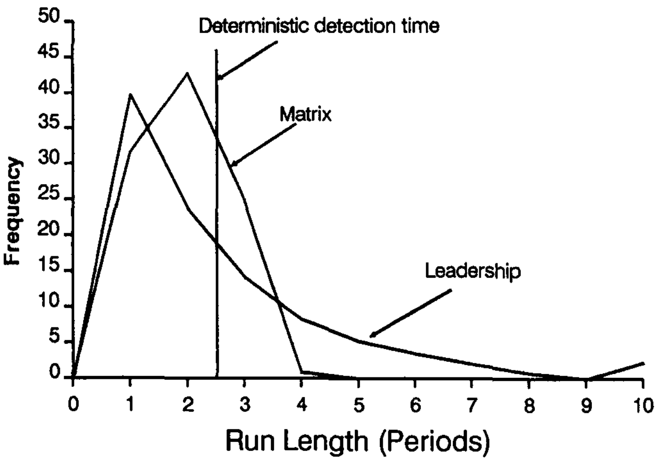

Fig. 4: Simulated probability distribution of run lengths to detection for $n = 10$ *and* $k = 4$ *for the two games. For the inspector leadership game, diversion was assumed to be at time* $i = 0$.

We conclude with an example of a real application. The detection time goal for diversion of irradiated fuel elements at light water reactors is currently 3 months /4/. The International Atomic Energy Agency achieves this goal by sending inspectors to the reactor sites at 3 month intervals, with a major inspection (physical inventory verification) occurring about once per year during reactor refuelling and maintenance. This corresponds to $n = k = 4$ in our models and a period length of 3 months. The interim inspection activities generally involve the checking of seals and reviewing of surveillance films, with virtually 100 % detection probability for diversion of a complete fuel assembly.

Figure 5 shows a plot of detection time in months as a function of increasing n, as the reference time of one year is subdivided into smaller and smaller intervals, with the number of interim inspections held constant at $k = 4$ and $k = 3$, respectively. The curves correspond to the matrix game saddlepoint solutions of Section 2.1. The guaranteed detection times are seen to decrease substantially with n, so that for $n = 12$ the timeliness goal can be achieved with $k = 3$, that is with only two random interim inspections as opposed to the current practice of three deterministic interim inspections. The prerequisites for realization of such a scheme are that the reactor operator agree to an inspection opportunity once each calendar month, and that the simultaneity assumption (advance commitment to a diversion strategy) is valid.

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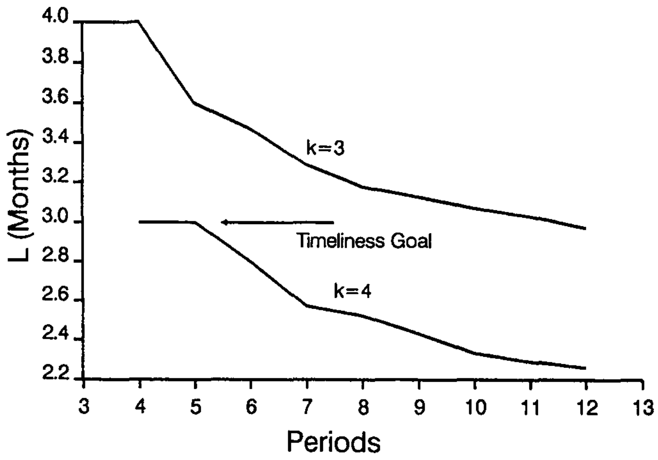

Fig. 5: Guaranteed detection times for a one year reference period, as a function of the number n of inspection intervals, for two values of k.

DEVELOPMENT OF A KNOWLEDGE-BASED SOFTWARE SYSTEM FOR INSPECTION STRATEGY

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ABSTRACT

Assessing inspection strategies against diversion plans can be made easier by using knowledge-based software tools, which add flexibility and allow for qualitative reasoning and simulation (of strategies).

Prior to developing a knowledge-based software system for inspection strategy, a feasibility study was performed on a simple diversion path analysis with a Prolog based software tool named SPIRAL developed by the CEA. The example chosen involves a spent fuel assembly diverted from a spent fuel pond.

The next step covers diversion path analysis involving flows of nuclear material between facilities within a comprehensive fuel cycle, using the same tools, with temporal aspects in the reasoning.

1 - INTRODUCTION

The objective of this study is to develop a software system to help to improve nuclear material inspection strategies.

It is being carried out under the auspices of the French Support Programme to the IAEA.

The purpose of all strategy is first and foremost to determine the frequency and scope of inspections which, as regards the IAEA, are of three types:

- annual inventory verifications,
- interim verifications (for timeliness),
- material flow verifications.

All of the above should be represented in the software. The important word qualifying it is "assistance". There is no question of this software finding the exact solution to the problem of inspection strategy optimization, but its use should make it possible to compare and improve different inspection strategies.

Inspection strategies cannot be determined unless the expected effectiveness of the inspections is estimated. We consider that the only way of seriously doing this iis to determine diversion paths or scenarios which cannot be detected. Hence, we consider that a software system must have one fundamental objective: it must be capable of determining diversion paths which cannot be detected by the safeguards approach set up by the IAEA or which are poorly covered by inspection activities corresponding to a given inspection strategy. As a consequence, its use must assist in improving or optimizing the inspection strategy, i.e. determine the strategy for a minimum efficiency level at least equal to the current level. In addition, the proposed strategies should lead to a reduction in means. In order to fulfil this objective we use new techniques: object-centred representation and backward chaining logic-based, reasoning with backtrack. All of this is included in the knowledge-based system tool SPIRAL and is described in §2.

In §3, in order to demonstrate SPIRAL's ability, we present the results of a study of the detection of diversion of a spent fuel assembly from a PWR spent fuel pond.

In §4 we present the next stage in the development of the task, a software system called STRASSY (STRategy ASsistance SYstem), in which diversion path analysis involving the flow of nuclear materials between facilities within a complete fuel cycle will be performed.

Finally in the conclusions, we discuss various possible applications of STRASSY.

2 - THE SOFTWARE TOOLS

To show a facility in sufficient detail, and indeed a full nuclear cycle, it is necessary to make a detailed description of the numerous parameters, such as the nature of the facility and of the transformation carried out, the plutonium and uranium-235 contents etc.

The following summary of the principles of object-oriented programming [1] shows how suitable it is for the problem in question:

- an object is a discrete entity used in a computer-based process of reasoning,
- as a number of objects can have common structures and forms of behaviour, it is helpful to group them in classes,
- for each class, there may be a number of sub-classes which inherit its properties,
- a class can be thought of as an environment in which objects, referred to as instances of the class, can be generated,

By using an object representation, one can generate a **knowledge base**. The next step is to use an **inference engine** in order to infer knowledge from the knowledge base and create a Knowledge Base System (KBS). If we limit our investigations to first order logic based on clauses — facts or rules — (with no more than one predicate), we have to select between logic with forward chaining and logic with backward chaining [2]. In the later case, the inference engine starts from an initial goal (for our problem of finding a diversion path) and examines the conditions that enable the goal to be reached.

Among the various KBS tools available today, we have selected SPIRAL [3], which features object-centred representation associated with rules, Prolog language (first order logic and backward chaining) and the possibility of running under the UNIX operating system with powerful graphical interfaces. (SPIRAL was initially developed at the French Commissariat à l'Energie Atomique and is marketed by the company CRIL) [3].

3 - SIMULATION OF THE DIVERSION OF A SPENT FUEL ASSEMBLY

We consider a fuel pond associated with a pressurized water reactor subject to international safeguards implying the following inspection activities:

a) Video retrieval from one video camera, for follow-up video reviewing (this camera is used for surveillance of the pond, i.e. enabling undeclared assembly movements to be detected).

b) Counting of the spent fuel assemblies present in the pond.

c) Spent fuel assembly serial number identification.

d) Night Vision Device (NVD) measurements for verifying the irradiated state of spent fuel assemblies, which is done only in the case of knowledge loss through camera breakdown.

In Figures 1 and 2, we give the classes and rules that allow us to formalize the problem.

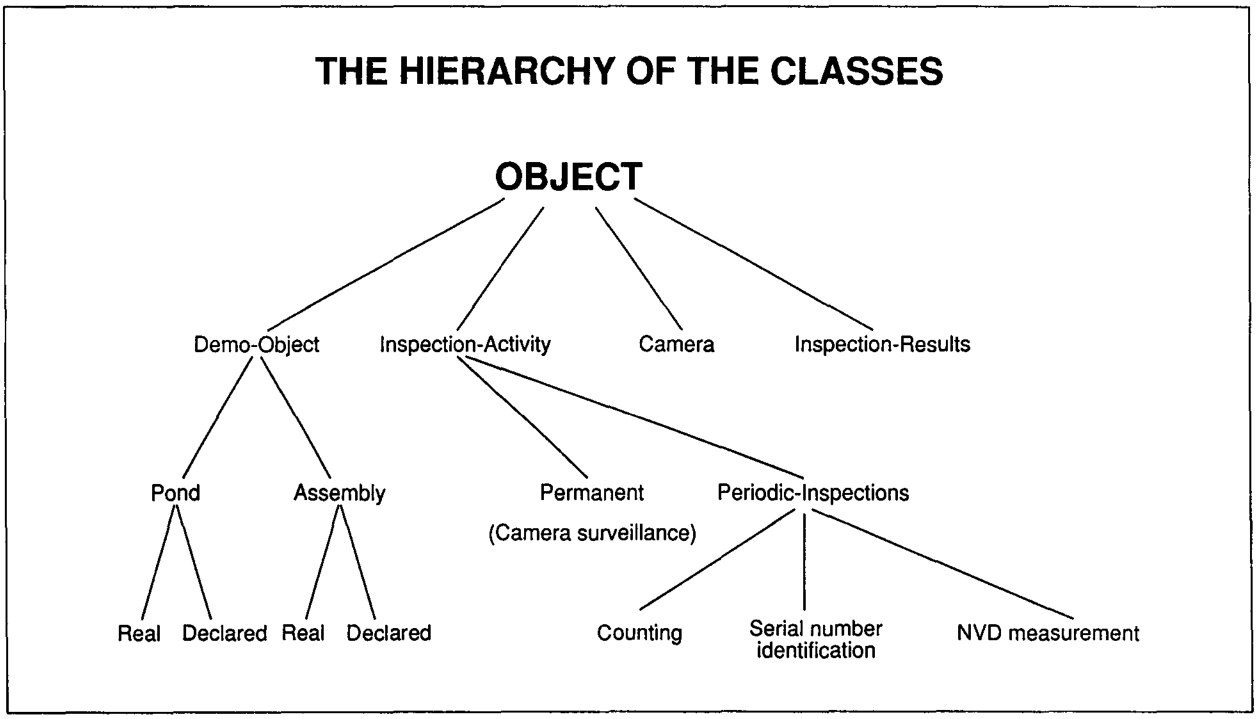

Figure 1

THE MAIN RULES

- **Operator strategy**
  - Try to find an undetected means of diversion
  - If the diversion is detected, refine the means of diversion
- **Inspector strategy**
  - Periodic inspections
  - Detection of assembly movements by camera
- **Assembly displacement**
- **Modification of assembly numbers**
- **Camera switched on/off**
- **Utilities for checking numbers of assemblies in real or apparent pond**

Figure 2

The subject of diversion is a spent fuel assembly (containing about half a significant quantity of uranium, i.e. 4 kg). The questions asked of the system are "for a given inspection strategy, what are the diversion paths?" and "are there any which have not been detected?".

For a given inspection strategy, the inference engine selects a number of the existing rules and arranges them to form action plans (diversion paths). In the event of failure, i.e. detection of the diversion path, the system backtracks and produces a more complicated action plan.

The first inspection strategy used includes activities a), b) and c). The results obtained give four action plans of increasing complexity. The first three end in detection while the fourth escapes detection.

- **The first plan.** The screendump in Figure 3 shows the end of this plan. The graphical windows give a true representation known to the operator (on the right-hand side) and a declared representation known to the inspector of the situation (on the left-hand side). The upper windows show these two situations in full. The middle windows correspond to observations made with the camera. The lower windows show other inspection activities b), c) or d). The plan features transfer of fuel assembly No. 1 to the hiding place and detection of the movement by the camera.
- **The second plan.** The camera is made unserviceable before transfer of the fuel assembly and returned to service afterwards. This is detected on counting the fuel assemblies (activity b) during the interim inspection.

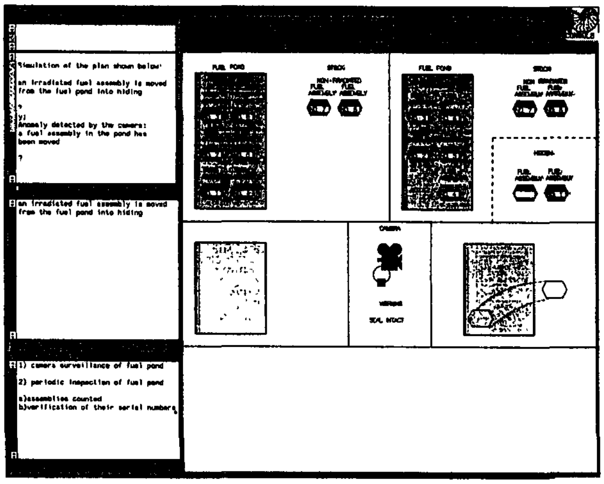
```
Simulation of the plan shown below'

an irradiated fuel assembly is moved
from the fuel pond into hiding

+
y;
Anomaly detected by the camera:
a fuel assembly in the pond has
been moved

?
```
```
an irradiated fuel assembly is moved
from the fuel pond into hiding
```
```
1) camera surveillance of fuel pond

2) periodic inspection of fuel pond

a)assemblies counted
b)verification of their serial numbers
```

Figure 3 - Diversion detected by camera

- **The third plan.** This is identical to the second except that the missing fuel assembly is replaced by a fake assembly. This is detected on checking the serial numbers (activity c).
- **The fourth plan.** This is identical to the third except that the serial number of the deviated fuel assembly is engraved on the fake assembly. In this case, deviation is not detected. However, analysis of the results reveals a minor anomaly (failure of the camera), indicating that the inspection strategy needs to be altered.

The new inspection strategy then includes activities a), b), c) and d). The final result of the fourth plan, as shown in the screendump in Figure 4, is detection of the diversion as measurement of the Cherenkov effect, indicating that one of the fuel assemblies in the pond has not been irradiated and is therefore a fake.

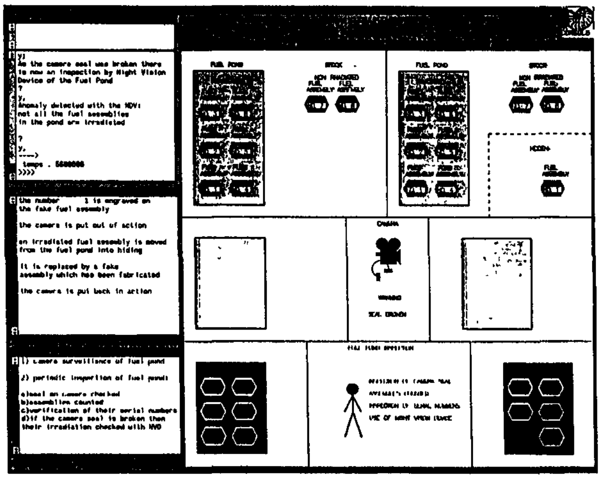
```
y;
As the camera seal was broken there
is now an inspection by Night Vision
Device of the Fuel Pond
?
y,
Anomaly detected with the NDV:
not all the fuel assemblies
in the pond are irradiated

?
y,
---->
 temps . 5600000
>>>>
```
```
the number      1 is engraved on
the fake fuel assembly

the camera is put out of action

an irradiated fuel assembly is moved
from the fuel pond into hiding

it is replaced by a fake
assembly which has been fabricated

the camera is put back in action
```
```
1) camera surveillance of fuel pond

2) periodic inspection of fuel pond:

a)seal on camera checked
b)assemblies counted
c)verification of their serial numbers
d)if the camera seal is broken then
their irradiation checked with NVD
```

Figure 4 - Diversion detected by NVD measurements

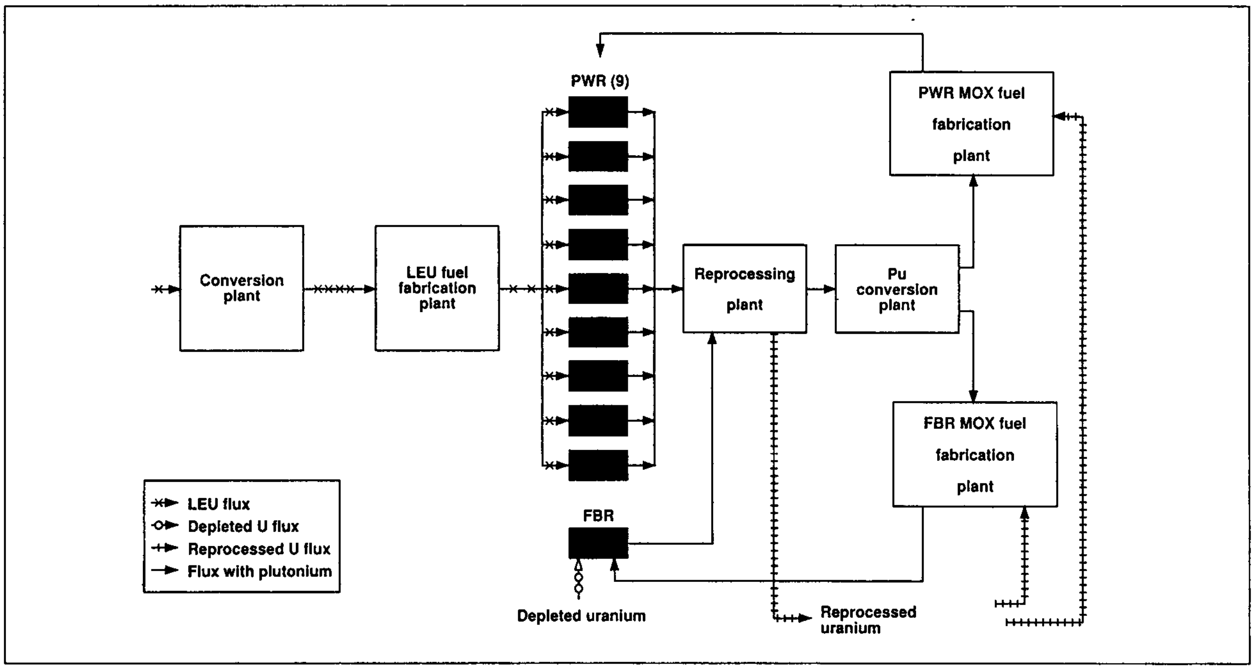

Figure 5 - Reference cycle

4 - DESCRIPTION OF A SOFTWARE SYSTEM FOR DIVERSION PATH ANALYSIS IN A REFERENCE CYCLE

After having demonstrated the ability of SPIRAL to help in the detection of diversion in a simple situation, we are preparing a demonstration prototype which should represent a complete reference cycle with the corresponding types of basic inspection activities and the frequency of these inspections.

We have decided not to make a general description showing all the existing fuel cycles but to concentrate on one which is sufficiently representative of the cycle complexity. This we call the reference cycle. It is designed to address a number of requirements:

- using the experience acquired during earlier studies [4 and 5],
- obtaining adequate cycle representation, particularly as concerns the recycling of plutonium,
- simplifying as much as possible the description of the cycle facilities; in this phase of the study we are essentially concerned with material flows between facilities and with object-centered representation, it is an easy matter to subsequently enhance the descriptions of the facilities.

The entire cycle is schematically shown in Figure 5, in which it can be seen to consist of sixteen facilities:

- nine pressurized water reactors with associated storage ponds,
- one fast reactor with a storage pond,
- one reprocessing plant,
- one plutonium conversion plant,
- one low-enriched uranium conversion plant,
- one low-enriched uranium fuel assembly fabrication plant,
- one pressurized water reactor MOX fuel assembly fabrication plant,
- one fast reactor MOX fuel assembly fabrication plant.

The cycle is assumed to be well established and we do not consider the period in which an equilibrium is established but only the events during one year, using quantified time intervals of one day. The nuclear materials used during this cycle are:

- the plutonium contained in the spent and MOX fuel assemblies,
- the low-enriched uranium,
- the uranium derived from reprocessing which is partially used in fabrication of the MOX fuel,
- the depleted uranium used in the fertile fuels of the fast reactor and the natural uranium, which we assume will not be the subject of diversion.

For each nuclear material, the significant quantities, the "detection times" and the residence times in the facilities are different.

The inspections in this cycle are of three types:

a) annual inventory verifications,

b) interim verifications, for timeliness,

c) flow verification, whenever material is shipped or received.

The frequencies of these inspections are predetermined as regards types a) and b) and depend on material shipment and receipt for type c). We assume here that, in the case of reactors, that type a) inspections are carried out at the time of refuelling (if a zone approach was to be considered later, simultaneous annual inventory verifications would not require simultaneous refuellings).

We are aiming to represent only those inspection activities which arise from the cycle aspect, i.e. we are not seeking to detail the activities inside the facilities. Therefore we consider each facility as just one single MBA. But the representation of these activities should be designed in such a way that it would be easy, at a later stage, to add or remove activities from the software package.

The temporal aspect should be taken into account in these activities, with a timeliness goal of one month, associated with the MOX fuel.

Time delays for arrival of declarations of shipments or receipts, at IAEA Headquarters are either set at a normal value (about 1 month) or, artificially at zero (in order to tighten the corresponding constraint). In the demonstration prototype, we take two months as the time limit for these shipments and receipts to appear in the IAEA accounting system.

A **diversion path or scenario** is defined as being a series of operator actions leading to diversion, and their corresponding durations, of which arrangement and chronology constitute a **diversion plan**, under the constraints present, in particular the usual inspection activities.

For the results of inspections, it is necessary to make a distinction between:

- "minor" anomalies, which do not necessarily imply diversion (e.g. some delay in sending a shipment or receipt declaration).
- "major" anomalies, where there is no doubt about diversion (e.g. a proven concealment or a very large unexplained MUF in a bulk facility).

A diversion path will be considered successful if the various corresponding operator actions make it possible to avoid the detection of major anomalies which can be easily detected by the safeguards set up by the Agency for a length of time linked to the category of nuclear material (timeliness goal).

The **temporal evolution aspect** should be well represented in the model since it is fundamental to the success (or failure) of diversion paths; it is therefore essential to adequately model the durations of the different actions or activities, as well as the times when they begin (or end). These are parameters which govern the sequence of activities. The temporal techniques needed for STRASSY are described in greater detail in [6] and [7].

5 - CONCLUSIONS

Knowledge-based computer systems appear to be able to make a decisive contribution to solving the problem of improving international nuclear material safeguards. As proof of their effectiveness, we have applied the SPIRAL tool to the inspection of a spent fuel storage pond.

Development work is currently being carried out on a prototype system containing descriptions of the inspection activities carried out in a fuel cycle. This protototype is capable of investigating all diversion paths for a given inspection strategy.

This prototype may be developed further — depending on the results obtained with it — in order to produce a complete operational software package capable of:

- incorporating the various inspection strategy techniques to allow comparisons, parametric studies and optimization, and eventually connections with other software programs which generate random or game-theory strategies,
- tracking down anomalies (implying a far more detailed representation inside the facilities).

At the prototype manufacturing stage, allowance should therefore be made for accommodating all the interfaces which will be required in the future, particularly for communicating with databases.

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NUCLEAR MATERIAL ACCOUNTANCY FOR AND CONTROL OF IN CZECH AND SLOVAK FEDERAL REPUBLIC

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Abstract

The Czechoslovak State System of Accounting for and Control of (SSAC) is described. It is discussed the organizational chart and role of the Czechoslovak Atomic Energy Commission as the State Authority in the Safeguards as well as its functions in the related fields (nuclear safety, physical protection) are mentioned. The individual nuclear facilities from the nuclear material accountancy point of wiew are shortly described and the necessity of well functioned facility level accountancy system is expressed. The cooperation between the SSAC and IAEA is mentioned and experience gained is briefly summarized.

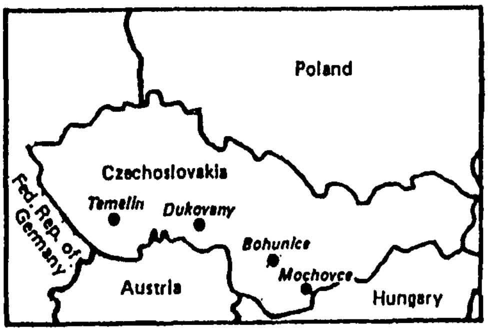
1. Introduction

Czechoslovakia is a non-nuclear weapons state. It signed the Non-Proliferation Treaty (NPT) in 1968. The Safeguards Agreement with the IAEA was signed in 1972. The State System of Accounting for and Control of Nuclear Materials (SSAC) is based on INFCIRC/153 and full scope safeguards are applied in the CSFR.

The "real" nuclear activity in Czechoslovakia has started in 1955 when the Nuclear Research Institute at Rez near Prague was founded.

The first prototype nuclear power plant A-1, 150 MWe was in operation from 1972 to 1978. It was an on-load reactor, moderated by heavy water, cooled by carbon dioxide and metallic natural uranium was used as fuel. Operation and experience have demonstrated the perspectives but also the performance limitation for this type of nuclear reactor. The construction of reactors with higher power would require a time consuming research and development programme and therefore it was decided to decommission the A-1 nuclear power plant.

At present, our nuclear programme is undergoing rapid industrial development (see Fig. 1). Due to the negative balance, CSFR urgently needed a reliable source of energy. The construction of power plants using brown coal as a fuel was stopped due to the high pollution effect and therefore it was decided to construct a number of industrially developed PWRs of the WWER type USSR design in the second stage of our nuclear power programme. It is estimated that the nuclear programme will include 12 nuclear reactors with an output of 440 MWe each. Eight units are already in operation in Jaslovske Bohunice and Dukovany, the remaining four units are scheduled to start operation gradually during the next five years. Construction has started on the first PWR (also WWER type) with an output of 1000 MWe. The site for 1000 MWe units is licensed for 4 units. Additionally two other sites have been selected and further sites are under consideration.



Facility	Type	Site	Status
1. NPP EBO	4xVVER 440	Jaslovske Bohunice	in operation
2. NPP EDU	4xVVER 440	Dukovany	in operation
3. NPP EMO	4xVVER 440	Mochovce	under construction
4. NPP ETE	2xVVER 1000	Temelin	under construction
5. LVR-15	research reactor	NRI Rez	in trial operation
6. LR-0	research reactor	NRI Rez	in operation
7. SR-0	research reactor	Skoda Plzen	in reconstruction
8. VR-1P	research reactor	Technical University Prague	in commissioning
9. AFR SFSF	storage facility	Jaslovské Bohunice	in operation
10. Regional waste disposal facility		Mochovce	under construction
11. Regional waste disposal facility		Dukovany	under construction
12. NPP A-1	HWGCR	Jaslovske Bohunice	in decommissioning
13. District heating system Trnava		Jaslovske Bohunice	in operation

Fig. 1 - Czechoslovak Nuclear Facilities

At the present time all fresh fuel for nuclear power plants is imported from the USSR. The away-from-the reactor storage is used with enough capacity to store spent fuel from all our 440 MWe NPPs up to five years.

Such an extensive nuclear programme cannot exist without nuclear research and industry. Fundamental research facilities include 4 research reactors, a small fuel fabrication plant and various laboratories. The industry is producing and exporting reactor components including reactor vessels, steam generators, piping, pumps, valves, etc.

All regulatory activities related to the nuclear safety of the peaceful use of nuclear energy are performed by the Czechoslovak Atomic Energy Commission (CAEC) namely by the Nuclear Safety Inspectorate headed by the Inspector General (see Fig.2). According to the Act No. 28 of March 22, 1984 on State Supervision of the Nuclear Safety of Nuclear Facilities CAEC supervises the nuclear safety of nuclear facilities, physical protection measures, transportation of nuclear materials etc.

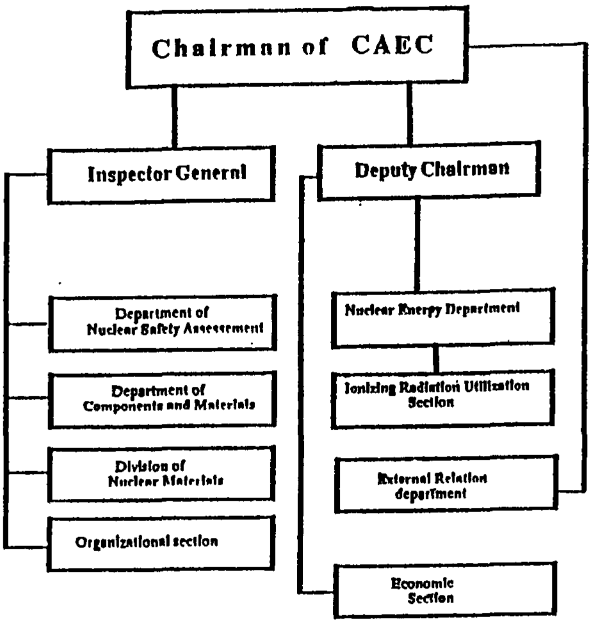


Fig. 2 - Organization scheme of the CAEC

Apart from the other regulatory activities the CAEC authorizes the use of nuclear material at the facilities, its export from and import to the CSFR, and accounting for and control of the nuclear material in Czechoslovakia. Therefore, CAEC is responsible for the SSAC. Licensing procedures are established and the national inspection of nuclear materials is carried out by CAEC inspectors.

The basic objectives of the SSAC are to prevent the use of nuclear material for purposes other than for which it has been specified, a timely detection of loss, should it occur, and assistance in returning the retrieved material to its former purpose. The SSAC operates in the way which facilitates the goal attainment.

Close linkage between the departments dealing with Nuclear Safety and Safeguards is very useful and important because exchange of information gained during inspections may lead to a timely detection of unauthorized use of nuclear facilities or installation or of misuse of nuclear materials. Even part of safeguards inspection activity may be carried out by the CAEC inspectors located at the nuclear power plants, whose main task is to supervise the nuclear safety of the facility.

2.SSAC - General Information

As a basic legal tool the accounting for and control of nuclear material folow Regulations for the Nuclear Material Accounting and Control issued by CAEC in 1977. The above mentioned regulations establish the CAEC as a regulatory body for nuclear material accounting and control. CAEC inspectors are authorized by the statute to enter any location where nuclear material is processed, used or stored, examine the accounting records, demand information essential for the performance of their duties, and to verify the physical state of all material present in the relevant location. If the CAEC inspector finds that the use of the nuclear material is not consistent with its designation or that it is not correctly accounted for, he may ask the manager of the facility to correct their findings. In case of repeated shortcomings in the nuclear material accounting, CAEC chairman may withdraw the nuclear material license from the facility.

During inspection the CAEC verifies the information reported by the facility and makes comparisons with the operation records. The inspector is authorized to carry out measurements, using CAEC's equipments, to verify the validity of the instruments and to order control measurements using facility equipment and to take samples and to apply seals as necessary.

The CAEC's inspections are divided into the following categories:

Routine Inspections: The number and scope of routine inspections in the facility is not limited. The frequency depends on the nuclear material in the inventory and on the effectiveness of the accounting and control system at the facility.

Physical Inventory Inspections: These inspections are usually carried out during a physical inventory taking. The inspector supervises the physical inventory taken by the operator.

Special Inspections: The CAEC is authorized to carry out an inspection upon dispatch of a special report or if it has reason to believe that a situation has arisen which would require the facility to send a special report. The CAEC also makes a special inspection if the facility does not comply with the provisions of the regulations. The scope of the special inspection is limited to those inventory items which are related to the special report or to the shortcomings discovered in the facility.

The CAEC is obliged to give advance notice to the facility, at least 24 hours in advance, about routine and physical inventory inspections. Special inspections do not require advance notification.

The basic material balance covers organizations which use, produce or consume nuclear material. The organization also accounts for material which is owned by another organization but is stored or used within its premises. If the organization has several nuclear facilities, the CAEC may divide it into several MBAs. This is dependent on the agreement with the IAEA. Each MBA is considered as an independent organization in this instance.

The manager of the facility is responsible for complying with the regulations concerning accounting for and control of nuclear material. The methods are prescribed in the instructions issued by the manager. The manager is obliged to prepare these instructions in consultation with the CAEC. The manager also appoints an expert (chief accountant for nuclear material) who assumes responsibility for the accounting for and control of nuclear material. For each MBA a deputy chief accountant is appointed as well.

The facility is obliged to inform the CAEC prior to receiving nuclear material, and declare the use of the material. Based on evaluation of this information the CAEC may issue a license to use the material. The nuclear material license is usually restricted to certain periods of time, certain categories or quantities of nuclear material, and may be subject to other conditions. A facility not having the CAEC license is not allowed to receive nuclear material.

Special attention is paid to waste disposal conditions. Before the nuclear material is disposed of as scrap, the facility must have approval from the CAEC. The CAEC can order control measurements and it has the right to take part in the disposal operations. After disposal, the material is in nonrecoverable form. Approval is also required in the case of termination or exemption of nuclear material.

Each MBA maintains the operating and accounting records in a form agreed with the CAEC. All movements of nuclear material have to be entered into the records and every record has to be signed by the responsible person.

The information system of the SSAC contains a number of reports. Any inventory change of location inside a facility which uses nuclear material must be reported to the chief accountant and entered within two days after the inventory transfer occurs. This is valid not only for changes which represent "real" inventory change between two MBAs but also for "internal" inventory changes representing transfers of nuclear material inside the facility. The chief accountant must submit the "real" inventory changes in the form of reports (ICRs) to the CAEC not later than five days after each change has occurred.

The communication between the facility and the CAEC is maintained by mail or by telex. Matters of urgency may be reported by telephone, and must be confirmed immediately in writing.

Communication with the IAEA is maintained, in accordance with the Agreement, through the CAEC only. All reports, advance notifications and other information from the MBA is collected in the Nuclear Material Division of the CAEC. The information is validated, checked with the design information, corrected as necessary and transcribed in the form agreed upon with the IAEA. Standardization of the information and its correctness is thus ensured. Similarly, the information from the IAEA is communicated to the MBA through the CAEC.

The increasing number of nuclear facilities in CSFR caused a similar high increase in the information flow from the MBAs to the CAEC and from the CAEC to the IAEA. During the period from 1972 to 1976 about 500 entry lines in the forms of ICRs, PILs and MBRs, were reported to the IAEA annually. This number gradually increased to about 10,000 entry lines during the last two years. Such a large amount of information represented a large workload and created many problems in performing the necessary cross-checkings and design information verification. The number of mistakes during the typing of reports also increased and the amount of paper work exceeded the time originally allocated for nuclear material verification. The only way to maintain high standarts and efficiency in the SSAC was to introduce a computerized accounting system. This was accomplished in 1979. Now the computerized system is an intrinsic part of our SSAC and anyone who remembers the "old times" with reports being typed would not want to do anything without the computer now. Today there are more than 90,000 entry lines in the computarized data base. Our computer, is compatible with the computers of the nuclear power plants and the data are transferred on magnetic diskettes or tapes. A number of programmes can be run for data processing. Automatic cross-checking between MBAs and design information checks are carried out. The accounting office is promptly informed about all inconsistencies in the reports and is able to correct them easily and without delay. Reports to the IAEA are prepared on magnetic diskettes and are transmitted to the IAEA every month. Besides routine reports to the IAEA various printouts supporting the inspection activity for each MBA can be requested. The book inventory listing (BIL) is very useful. This list of batches is prepared by entering inventory changes to the last physical inventory list and includes more information than is requested for physical inventory listing. It provides good basis for performing a physical inventory and also helps during both CAEC's and IAEA's inspections. The general ledger is also a very important document as well as the list of sums stratified according to key measurement points, material description codes, etc.

For small MBAs with low material flow, a simple accounting system using hard copy form is still used. Information from hard copies are checked at the CAEC and used as input data for the national computerized system.

The regulatory activities of the CAEC are supported by a number of laboratories of the Nuclear Research Institute. Assistance in the field of nuclear material accounting and control is provided by the Central Control Laboratory. The main task of this laboratory is to analyse samples taken during inspections. The laboratory also arranges consultations with the MBAs to assist in establishing effective accounting and control systems, to develop measurement systems for nuclear material movements and for physical inventory takings, etc. The laboratory is also involved in the IAEA's laboratories network programme and has been performing successful chemical analyses of a number of IAEA's samples taken from various types and forms of nuclear material, especially of plutonium and uranium in spent fuel.

As mentioned above, the Nuclear Material Division of the CAEC is responsible for accounting for and control of nuclear material. At the present time it accounts for 13 MBAs including 4 research reactors, five nuclear power plants and one away-from-reactor-storage. The Division is responsible to the Inspector General who is the head of the Nuclear Safety Inspectorate. All regulatory activities of the CAEC are concentrated in one department and a high efficiency in both inspection and licensing activities is achieved.

3. SSAC - Practial Information about Nuclear Material Accountancy at Different Types of Facilities

Research Reactors: There is a very low material flow. The accounting systems are very simple and easy to verify. The critical assembly LR-0 is an exception. This facility is loaded with dismountable fuel assemblies and fuel pins are often reshuffeled during testing. The basic accounting unit is one fuel pin instead of one fuel assembly and each fuel pin has its own identification number. A computerized accounting system was installed to track every single pin in the facility.

Bulk Facilities: These are small facilities in CSFR. The most important is the Nuclear Fuel Institute. It has a very limited production of low enriched uranium fuel in the form of uranium dioxide pellets and shielding containers from depleted uranium are fabricated there. Additionally, there are also other locations such as laboratories, research institutes, glass works which use nuclear material in bulk form. Their throughput and inventory of nuclear material is very low. Therefore, the accounting application is very simple. In bulk facilities the MUF values are establishsed every year. They represent 0.1 - 0.5 % of the material inventory. This value is fully acceptable from the safeguards point of view.

Nuclear Power Plants: This group of facilities consists of four MBAs with twin WWER 440 units, one MBA is away from reactor storage for WWER 440 spent fuel, and one MBA is spent fuel storage of the A-1 power plant. The WWER 440 unit is comprised of two reactors and two spent fuel stores in a common reactor hall. Easy access to the fresh and irradiated fuel and the application of seals and surveillance cameras is provided. Fuel assemblies maintain their identities after receipt from the USSR. The accounting system at the NPPs is computerized. The A-1 power plant is a complicated facility from the safeguards point of view. The facility design does not allow easy access to the spent fuel and it was necessary to install a surveillance system in order to verify that no fuel is shipped from the MBA without control.

IAEA safeguards are applied at the individual facilities in accordance with the relevant Facility Attachment. Each facility attachment is carefully analyzed and the accounting system is tailored so that all requirements are fulfiled.

In 1990 the IAEA carried out 49 inspections in the CSFR. Most inspections were in NPPs (4-5 inspections per year per MBA). IAEA inspectors are always accompanied by CAEC inspectors and it provides for assurance that all safeguards requirements are fully understood and fulfilled. The number of person-days used is normally lower than stated in the Facility Attachment. This reflects the correctness and operational efficiency of our SSAC.

Of course, from time to time, some kind of discrepancy is found during the IAEA or internal inspection. When it occurs the manager of the facility is informed and asked to clarify the situation. Follow-up inspections are carried out by the CAEC inspectors to confirm that all problems have been solved.

The CAEC keeps on good term with Department of Safeguard of IAEA. Every year the working visit, where the operational problems are solved in very short way, is organised and every two or three years the visits on higher level are arranged, where the general views, opinions and evaluations related to performance of safeguards and nuclear programme of Czechoslovakia are discussed.

The Czechoslovak experts participate in IAEA's advisory groups and technical committees playing very active role. Very interesting is e.g. the proposal to implementation of so called "selective safeguards" which was raised by Czechoslovak member of SAGSI.

The IAEA takes the advantage of very short distance among Vienna and the Czechoslovak nuclear facilities and according to the offer of CSFR the IAEA realises some field tests of new developed safeguards instruments there. At the Czechoslovak NPPs some TV surveilance systems, underwater telescope and spent fuel counter have been tested, some measurements on the fresh fuel have been performed and some research contracts are running.

The Introductory Course of Agency Safeguards 1990 for new IAEA inspectors was organised by SSAC in cooperation with the Nuclear Research Institute and Nuclear Power Plant Dukovany in CSFR.

Another example of good cooperation among the SSAC's should be mentioned. Since about 1976 the meeting of safeguards experts from the CMEA countries dealing with the technical aspects of international safeguard is held every year. During that meeting various problems concerning the computerised and standardised safeguards systems at both SSAC and facility level, the export and import of nuclear materials, evaluation of safeguards goal attainment in SIR etc. have been discussed and often solved. At present the organisation of these meetings is interrupted but we hope that on a new basis, with participation of all interested countries, similar meetings, which we consider very useful, would be organised again.

4. Closing Remarks

The Czechoslovak SSAC accounts for all nuclear material in the nuclear facilities and other locations using nuclear material and it complies with all IAEA's safeguards reguirements. Its approach to international safeguards reflects our international policy, to utilize nuclear energy solely for peaceful purposes. Also, in the future we will suport every effort to incerease the effectiveness of safeguards and to improve the reliability of the nuclear material verification.

In our opinion, good cooperation between the operator and the CAEC will result in further progress in this field. We conduct regular meetings with the chief accountants at individual MBAs in order to explain the goals of the SSAC and the IAEA safeguards. These meetings are a very good basis for exchange of information and often result in new ideas for improvement to our SSAC.

Also the close cooperation between the IAEA and SSAC increases the efficiency of safeguards inspections activity and the safeguards goal attainments without increasing the total inspection effort and is advantegeous for both the IAEA and CSFR.

ELECTRONIC DATA INTERCHANGES IN BANKING AREA WITH EDIFACT LANGUAGE AND ETEBAC 5 PROTOCOLE

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Banking area was for many years, very involved in EDI (Electronic Data Interchange): in 1990, the French banking system, quite as U.K. banking system, cleared more than 1,700 x 10^6 electronic payment messages: the same year, 3,000 banks in 65 countries exchanged 300 x 10^6 electronic payment messages throught SWIFT, the international banking network.

All these electronic transactions need safeguards systems for information which have to be understood by specific people in the world as a Japanese buyer who has to pay invoices to a Spanish seller via their respective banks.

Two challenges had to be solved.

1. <u>An international Language Understandable by any Partners: EDIFACT</u>
(Electronic Data Interchange for Administration, Commerce and Transport)

This EDI language is already used for messages like: Orders, Invoices, Payments ... Five regional rapporteurs are reporting twice a year in Geneva (United Nation Organisation) for common agreement on messages including segments, data elements and codes. These rapporteurs, from Eastern Europe, Western Europe, USA and Canada, New Zeland and Australia, Japan and Singapore with support of Message Development group are building, together, this new language which already contains about 50 messages, 1,000 data elements and quite 10,000 codes. One main message in the banking area is called the "PAYORD" the fund transfer order initiated in EDI by the buyer to this bank for paying his seller.

2. <u>Security in the EDI Teletransmission</u>

French banks defined several ETEBAC protocols (stands for data communication between banks and their customers) in order to provide the French banking community with telecommunication standards. This standardization is important to favorize such exchanges, reducing the data processing costs and delays which is a valuable benefit for both customers and banks.

Today, ETEBAC 5 standard introduces new and most significant improvements:

- *ETEBAC 5 uses a reliable and efficient file transfer protocole PeSIT, close to international standard FTAM.*

- *ETEBAC 5 allows totally secure transfers, including a strong mutual authentication of communicating partners and a real digital signature.*

- *ETEBAC 5 complies with international standards for telecommunication (OSI model) and security (ISO TC 68 standards). ETEBAC 5 standard also permits the transfer of EDI files.*

Security services provided

One of the main features of ETEBAC 5 standard is the availability of a wide range of security services:

- *mutual partners authentication,*
- *file content integrity,*
- *file content confidentiality,*
- *mutual transfer non-repudiation.*

These services imply usage of symmetric algorithm DES (Data Encryption System) and of asymmetric algorithm RSA (Rivest, Shamir, Adleman).

Mutual authentication relies on each partner possessing an RSA secret key and a certificate issued by a Certification Authority.

File content integrity is obtained by verification of a Message Authentication Code (MAC) computed using the Cypher Block Chaining (CBC) mode of DES algorithm. This MAC is appended to the transmitted file by the sender, the receiver checks that this received MAC is the same as the one he computes from the data he actually received.

File content confidentiality is obtained by enscription of the file content using the CBC mode of DES algorithm.

Mutual transfer non-repudiation is obtained by transmission of a digital signature of the file from the sender, and of an acknowledgment, including a digital signature, by the receiver. The sender signature and the receiver acknowledgment are based upon RSA encryption, using the owner secret key, of file caracteristic data including MAC file.

Keys management

ETEBAC 5 security has been designed in order to avoid the management of any kind of key directory:

- *RSA public key are dynamically exchanged during the hand-shakes related to the mutual authentication.*

- *DES encryption and MAC keys are generated when needed, and transmitted RSA crypted.*

Security usage

Mutual authentication may be provided only with partners engaged in an ETEBAC connection.

Usage of confidentiality is optional.

File signature and acknolwedgment may be transmitted, as protocol parameters, during the file transfer, or may be recorded in a specific file, called "Execution Order" that may be transmitted indipendently from the main file. So, whatever the communication medium used to transmit the main file may be, its Execution Order may be sent using ETEBAC 5.

Signature and acknowledgment must be performed by Customer and Bank entities. Bank and Customer Operators may use the same security mechanisms, but, thus, will not engage the Bank nor the Customer responsibility. Two different Customer entities may perform double signature of the same file.

So, ETEBAC 5 standard may satisfy any organizational scheme on both partner sides.

Before the end of this year, French banks are going to provide this new customer EDI service that we call EDI with EDIFACT language and ETEBAC 5 protocole to secure all the electronic messages transmitted from the corporate to the bank.

Thank you for your attention.

STATE SYSTEM OF ACCOUNTING FOR AND CONTROL OF NUCLEAR MATERIALS IN POLAND

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Abstract

The paper presents the operation of the State System of Accounting for and Control of Nuclear Materials (SSAC NM) in Poland. The structure of the system, legislation, inspection activities, data maintenance and computerized system of data processing and reporting to IAEA are presented in the paper. The nuclear materials control in the field by the NDA methods performed by state inspectors is also described.

1. History

Poland, as the party member of the Treaty on the Non-Proliferation of Nuclear Weapons (1970), concluded the Agreement with the International Atomic Energy Agency for the application of Safeguards, basing on the IAEA INFCIRC/ 153 document. This Agreement entered into force on the 11th of October 1972. Since then safeguards is applied on all source nuclear materials and special fissionable materials within the territory of Poland.

2. National and International Sources of Regulations on Nuclear Material Control in Poland

The basis for the legal regulations on nuclear material safeguards in Poland is the NPT Treaty, Agreement for Application of Safeguards signed with the IAEA and Subsidiary Arrangements to this Agreement specifying all detailed requirements and procedures on international safeguards application in Poland. Each facility is controlled by IAEA inspectors according to the Facility Attachements documents.

Two main legal documents regulate the operation and the requirements of nuclear material safeguards in Poland:

- "Atomic Law" - Act of Parliament of 10th April, 1986
- "Principles of Accountancy and Control of Nuclear Materials" - Regulation issued by President of the National Atomic Energy Agency (NAEA) on 20 October, 1987

President of NAEA in Poland is authorized to enforce regulations on nuclear safety and radiological protection.

3. Structure of the State System of Accounting for and Control of Nuclear Materials in Poland

Nuclear materials accountancy and control as well as the physical protection of nuclear materials is under supervision of the National Atomic Energy Agency (NAEA).

Nuclear materials control is performed by nuclear safety inspectors from the National Inspectorate of Nuclear Safety and Radiological Protection, which is one of the departments of the NAEA. The State Office for Nuclear Materials Accountancy and Control (SO-NMAC) is situated in the Central Laboratory for Radiological Protection, Warsaw which among it's other activities is responsible for the accountancy of radioactive materials.

Nuclear materials are accounted for in 6 material balance areas:

- PL-A - research reactor EWA (10 MW)
- PL-B - critical assemblies ANNA and AGATA
- PL-C - research reactor MARIA (30 MW)
- PL-D - research laboratories of the Institute of Atomic Energy and the Institute of Nuclear Problems (Świerk)
- PL-E - miscellaneous locations (small quantities of nuclear materials in different research laboratories and institutes - eg. in medicine, Universities) in 51 locations over the whole country
- PL-F - research laboratories in the Institute of the Nuclear Chemistry and Techniques (Warsaw).

4. State Office for Nuclear Materials Accountancy and Control (SO-NMAC)

The SO-NMAC is responsible for collecting and maintenance of all accountancy data and other informations relevant to nuclear materials balance and movements between MBA-s and from/to the country.

This documentation is based on periodical informations and reports from users of nuclear materials as well as on results of control activities of nuclear safety inspectors from NINS & RP.
The SO-NMAC cooperates directly with the IAEA Department of Safeguards and prepares all ICR, MBR, PIL and other standard reports to the IAEA. Each potential user of nuclear materials must fulfill special requirements prior he is allowed to buy nuclear materials. He is obliged to organize internal system of nuclear materials accountancy and control - describe its operation with specifying all internal accountancy documentation and control procedures. This document is subject to the approval of the President of the NAEA after its verification by the SO-NMAC. Such documentation must be prepared 12 months in advance before expected date of receiving of nuclear materials in the case of nuclear facilities and 3 months in advance in the case of the localization outside nuclear facilities. Any change in the organization of nuclear materials safeguards within nuclear facility must be announced to the SO-NMAC 3 months in advance and is subject to be licenced by the President of NAEA.

Advance notification on the foreseen import of nuclear materials should be sent to the SO-NMAC by importer 30 days in advance and the notification on planned export 60 days in advance, if its quantity exceeds 1 eff kg in total during 3 months. If this quantity is less than 1 eff kg then the notification on foreseen shipment/receipt should be sent 7 days before the time of the transaction. In the case of international transfers this documentation should contain:

- name and address of shipper/receiver,
- quantity and type of nuclear materials,
- foreseen date of shipment/receipt,
- foreseen date and place of nuclear materials packing/unpacking,
- foreseen date and place of taking the responsibility for nuclear materials by Poland and on transfer of this responsiblility to the other state in case of export,
- formal certificate of competent authorities of foreign receiver that exported from Poland nuclear materials will be subject to the IAEA safeguards (with exception of nuclear countries as specified by NPT Treaty).

Each shipment on receipt of nuclear material is reported to the SO-NMAC not later than 5 days from the date of its shipment/receipt on standard "Notification on Change" document which is one of basic accountancy documents.

There are 4 people involved in the work of the SO-NMAC:

- 2 nuclear safety inspectors
- 1 programmist
- 1 technician (book keeping)

The responsibilities of the SO-NMAC cover following activities:

- maintenance of accountancy records on all nuclear materials within the territory of country,
- inspections and control of nuclear materials safeguards,
- participation in the inspections of IAEA inspectors,
- control measurements of nuclear materials,
- research and development in the field of nuclear materials accountancy and control.

Nuclear materials accountancy at facility level is performed by the facility safeguards officer.

5. Inspection Activities
(state and IAEA)

State Inspection activities concentrate on the:

- control of the presence of nuclear material and verification of the completness and correctness of the nuclear materials safeguards accountancy documents at nuclear facilities and other users of nuclear materials,
- control measurements of nuclear materials enrichment (NDA) and quantity (NDA, weighting),
- control of the fulfilling by the nuclear material user the requirements and principles of nuclear materials safeguards according to approved "Safeguards System Organization" at the facility or at the other user.

There are 3 types of safeguards inspections which are typically performed by the SO-NMAC:

- normal inspection - routine inspection to control nuclear materials balance and correctness of safeguards accountancy data in comparison with reports received by SO-NMAC;
- ad hoc inspections - to control data which can not be verified basing on received reports and to control the fulfillment of requirement from previous inspections;
- special inspections - performed in the case of unusual events, as eg. unauthorized movement of nuclear materials or its loss.

The IAEA inspections are performed routinely on monthly basis at research reactors. In total 26 mandays for each of 2 reactors. Other MBA-s are controlled at least once per year after PIT for PIV. In addition to the Physical Verification of the nuclear materials and verification of accountancy data, the following measures are being applied for safeguarding of and control of nuclear materials:

- IAEA metallic and fiber glass seals on fresh and spent fuel containers;
- State Office - paper seals on storage areas or containers with nuclear materials;
- Measurements with NDA-methods (state - NDA spectrometry with portable germanium detector; IAEA - SAM II, PMCN, PMCG, HM-4, ICVD .

Few fuel elements of high enrichment were measured by IAEA inspectors with Neutron Collar and since then they are used as secondary standards for the measurements with NaI or HPGe detectors by IAEA inspectors. Measurements are being done by IAEA inspectors - 4 times a year in research reactor and in each of MBA-s during PIV.

6. Nuclear Material Under Safeguards in Poland

Safeguards is being applied in Poland according to the provisions of the Agreement with the IAEA on:

- special fissionable materials (Pu-239, U-233, enriched uranium in U-235 and any material containing one or more of these radionuclides);
- source nuclear materials - natural, depleted uranium and thorium (in metallic form, alloy, chemical compound or concentrate);
- uranium and thorium ores - in the case and the amounts subject to be processed for separation of uranium and thorium.

Nuclear materials in Poland are used mainly at research reactors which are operating for research, training and isotope production purposes. The fuel cycle in Poland is not closed as there are not uranium enrichment or processing/reprocessing facilities in Poland. The construction of the first nuclear power station, which was supposed to be the WWR-440 reactor, was stopped in 1990 and is actually not supposed to be continued.

Nuclear materials used at research reactors are highly enriched uranium (80% and 36% at MARIA and 36% at EWA reactors). Fuel elements are accounted for basing on shipper certificates (USSR) data. Each fuel element has its own certificate and engraved identification number. Spent fuel is stored at the place of reactors in water pools.

In localisations outside facilities - PL-D,E,F small quantities of uranium, thorium compounds and plutonium (Pu-Be neutron sources) are used for research purposes in nuclear physics, chemistry and industry. Depleted uranium is used mainly as the shielding for highly radioactive gamma sources.

7. Computerized Nuclear Materials Accountancy System

Accountancy system at State Office was computerized in 1988/89. D'Base III Plus data base system operating at IBM PC/AT computer was used for computerized system of nuclear materials accountancy data. For the processing to the IAEA - PIL, ICR and MBR documents are created with the programme and transmitted on 5 1/4" diskettes. The total number of reports submitted the IAEA per year exceeds usually 40.

The system utilises one main data base which is organized in sequential files. Additionally all reports which have been sent to the IAEA are kept as separate files. Following reports can be created with the system:

- ICR, PIL, MBR, Concise Note (for transfer to the IAEA)
- General Ledger
- Book Inventory Listing
- Seals List (subtotals for sealed, unsealed materials)
- Nuclear materials stratification.

Quality control of data entries to the system is performed, what assures correctness and consistency with requirements of Facility Attachments and Code 10. Actually fixed format of Code 10 is applied for data transmission to the IAEA. Anyway parallelly, from several months, the labelled code 10 format is used and tested in cooperation with the IAEA. In the nearest future after testing period we intend to switch only for labelled code 10 format. Reports created with our system are used also for support of inspection activities of IAEA inspectors and the prepared hard copy form is suitable to fill in IAEA inspection logsheets.

Accountancy at facility level is based mainly on book keeping. For research reactors computerized accountancy is performed.

8. Nuclear Materials Measurements. Research and Development Activities on Nuclear Materials Safeguards

There are two types of control measurements performed by nuclear safety inspectors during their inspection activities. Measurement procedures were elaborated by SO-NMAC in the framework of governmental research programme 1988/90 - Nuclear Safety and Radiological Protection. NDA method of gamma spectrometry with HPGe portable detector is used for uranium enrichment control. In the case of nuclear fuel - because of its standard geometrical form - enrichment and the total amount of uranium is being checked basing on reference calibration standards. Such measurements are based on the 185.7 keV energy for U-235 and 1001 keV for U-238 (Pa-234M).

There are 2 fuel elements which were measured in the past for total uranium by IAEA inspectors with AWCC. These elements we use as secondary standards for control of highly enriched fuel assemblies. These measurements are usually done parallely to IAEA measurements of fresh fuel for the same elements - "from the other side". Portable HPGe detector and portable Canberra-10 (4096 K) MCA is used for measurements. Spectrum is elaborated at the place with portable Toshiba personal computer. As MR-6 type fuel used for reactor MARIA is of 2 different enrichments (80% and 36%) its distinguish can be sometimes difficult because of the same geometrical shape of the fuel. To distinguish these two types of fuel - photopeak ratio Pa-234M : U-235 (1001 keV : 185 keV); is used for quick recognition of the enrichment. Such measurements of fuel can be done at any distance of fuel from the detector.

The same "qualititative" method for uranium enrichment control is used for control of materials in bulk or liquid form, which are kept in different type of containers (steel, plastic, glass). The principle of enrichment measurements in the field is to measure with no opening of the container and never moving the material from its own container to the standard one at the place of control. Typically several batches of nuclear material in bulk form are measured at the place of control. The enrichment is estimated basing on the following equations:

$$E_1(\%) = a_1 \cdot \left[\frac{N(766keV)}{N(185keV)}\right]^{b_1}$$

$$E_2(\%) = a_2 \cdot \left[\frac{N(1001keV)}{N(\ 185keV)}\right]^{b_2}$$

where $a_{1,2}, b_{1,2}$ factors are calculated as for the set of measurements of uranium compounds (UO_2 - powder) of different enrichments - from depleted up to 80% enriched uranium.

As these measurements are used for the "estimation" of enrichment not its precise measurements, then no corrections are applied for the attenuation of gamma radiation in different uranium compounds or container materials. This effects can be controlled and estimated basing on the automatic printout of the spectrum elaboration which calculates following photopeak ratios and presents it in tables:

Relations of photopeak ratios of U-238 daughters and U-235 calculated for the enrichment estimation :

$$\frac{\text{keV}}{\text{keV}}\ \frac{63,\ 766,\ 1001}{185,\ 205}$$

Relations of U-235 and Pa-234M photopeak ratios used for the estimation of autoabsorption effect :

$$\frac{\text{keV}}{\text{keV}}\ \frac{84,\ 143,\ 163,\ 194,\ 205}{185}$$

$$\frac{\text{keV}}{\text{keV}}\ \frac{63,\ 258,\ 766,\ 786,\ 883,\ 946}{1001}$$

Photopeak ratios (766/185; 1001/185 keV/keV) when presented in graphic form for the set of controlled materials indicate clearly possible uncorrect operators data on uranium enrichment .

Such samples are later on sent to the SO-NMAC for precise measurements of U-enrichment with the 185.7 keV photopeak measurements and calibration of the germanium detector with EC-NRM-171/NBS-SRM-969 set of standards. Special collimators and plastic containers for bulk form materials nuclear control are then applied in spectrometrical laboratory of the SO-NMAC. All correction factors are then applied as specified in the instruction manual of these calibration sources utilization.

9. Final remarks

Referring the activities of the SSAC NM in Poland and the implementation of IAEA safeguards for last 19 years the provisions of the Safeguards Agreement between Poland and the IAEA were always met. The cooperation between State Office and IAEA inspectors was always very constructive and correct. Poland accepts all designated by IAEA inspectors with no restrictions.

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NUCLEAR MATERIALS ACCOUNTANCY IN AN INDUSTRIAL MOX FUEL FABRICATION PLANT SAFEGUARDS VERSUS COMMERCIAL ASPECTS

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Abstract

In a modern MOX Fuel Fabrication Plant, with a large throughput of nuclear materials, computerized real-time accountancy systems are applied. Following regulations and prescriptions imposed by the Inspectorates EURATOM-IAEA, the State and also by internal plant safety rules, the accountancy is kept in plutonium element, uranium element and ^{235}U for enriched uranium.

In practice, Safeguards Authorities are concerned with quantities of the element (U_{tot}, Pu_{tot}) and to some extent with its fissile content. Custom Authorities are for historical reasons, interested in fissile quantities (U_{fiss}, Pu_{fiss}) whereas owners wish to recover the energetic value of their material (Pu equivalent).

Balancing the accountancy simultaneously in all these related but not proportional units is a new problem in a MOX-plant where pool accountancy is applied. This paper indicates possible ways to solve the balancing problem created by these different units used for expressing nuclear material quantities.

1.Introduction

In the last 30 years BELGONUCLEAIRE developed a "real time" nuclear materials accountancy system which was applied first in its research laboratories and later in the MOX Fuel Fabrication plant in Dessel. This computerized system fulfils the requirements put forward by the Inspectorates IAEA and EURATOM, and in particular those defined in the Commission Regulation EURATOM n° 3227/76 of 19 th. October 1976 concerning the application of the provision on EURATOM Safeguards.

In principle this accountancy is kept in plutonium element (Pu_T), uranium element (U_T) and in enriched uranium (^{235}U).

Although Safeguards authorities are directly concerned with quantities of PU_T, U_T and ^{235}U; the Inspectorates have an interest in the fissile content, as verification measurements are made to determine the isotopic composition as well as the total content.

When crossing of State borders occurs, the customs authorities verify and follow the fissile content (Pu_{fiss}) of the material, as for historical reasons, it is considered by them as a good unit for the financial value of the material. To a certain extent the EURATOM Supply Agency wants to know the plutonium fissile content of nuclear material transfers when the borders of the Community are crossed.

But last but not least, the customer of the MOX plant, who is using the plutonium in order to produce electricity is interested in recovering the energetic value of the material he supplied, value or quantity expressed in $Pu_{equivalent}$ (Pu_{eq}).

In order not to keep separate accountancy systems running in parallel for Safeguards, for clients and for national and international authorities, BELGONUCLEAIRE tried to solve this problem by using the system of "current accounts" in which all quantities are recorded in Pu_T, Pu_{fiss} and Pu_{eq} simultaneously.

In this system the requirements of Safeguards and commercial nuclear material management are met.

The title of this presentation should in fact not be "Safeguards versus commercial aspect" but well "Combining Safeguards with commercial aspects".

2.Pool Accountancy

2.1. The basis of the good functioning of a MOX-Fuel Fabrication Plant lies in the fact that there is a continuous flow of plutonium through the process without long hold-ups or storage during which ^{241}Am builds up. This good flow is needed because all plutonium batches which are processed nowadays have a high ^{241}Pu content, a situation which is different from the one occurring 15 or 20 years ago when the basis rules of material follow up were laid down.

Indeed in the past there was a relative low percentage of ^{241}Pu eg. 2 to 4 %. In recent years however this ^{241}Pu content went up to 12 %. Taking into account the relative short half-life of this isotope namely 14.4 years there is a rather high ^{241}Am build up in a short time lapse. Higher ^{241}Pu means faster ^{241}Am build up and consequently higher radiation (gamma, alpha and thus neutron) which has to be minimized during material handling.

2.2. An other important factor which has to be taken into account is the fact that the plutonium batches (60 to 80 kg Pu_T) are sometimes very different from each other in isotopic composition. In other to obtain the uniform isotopic composition in the MOX-product which is required by some customers it is necessary to perform cross-blending. This cross-blending between batches, not only of the same client, but also between batches supplied by different customers led to the need of instituting a pool accountancy system for the BELGONUCLEAIRE MOX-Fuel Fabrication Plant.

2.3. So, during the last 5 years, BELGONUCLEAIRE operates its Dessel plant as a pool of fissile materials; i.e. that when entering the plant the fissile material looses its ownership and its obligation (fungibility principle). However accurate balances are kept by BELGONUCLEAIRE in terms of :

- debt towards each customer;

- liability towards Safeguards Authorities, Belgian Authorities, EURATOM and IAEA following the Commission Regulation (EURATOM) n°3227/76 of 1976 in terms of nuclear material content per material categories as well as per obligation and also towards the EURATOM Supply Agency in term of total as well as fissile material component per campaign or sub-campaign.

When leaving the BELGONUCLEAIRE plant, normally after transformation from raw material into mixed oxide fuel, the fissile material regains ownership and obligation. Change of obligation can only with the agreement of both parties and by Euratom. It must however be well understood that a customer supplying fissile material for a fabrication (or a storage) campaign will get in return an equivalent amount of fissile material (decay taken into account), but must not expect to receive back the same material (quantity, isotopic vector, etc ...).

This pool accountancy system is beneficial for both customers and fabricator because :

- it avoids expensive segregation and traceability between fabrication and recycle materials from different customers;
- it allows for the utilization of material belonging to one customer for the supply to another customer so that overageing of materials awaiting utilization for the next campaign for the same customer does not occur. Overageing would lead to products that have to be purified by the expensive liquid route because their Americium content has become excessive for fabrication;
- it allows for a wider choice for cross-blending operations with as consequence a reduced spread of isotopic compositions within the product.
- it avoids interruptions of fabrication in case of temporary technical or administrative problems regarding the timely supply of fissile material to the fabricator's plant, thus helping in obtaining a reliable mixed oxide fuel supply to the customers' reactors.

In order to be sure that each customer receives back its due quantity and quality of material the system of current accounts for the fissile material accountancy is applied.

3.Fissile material accountancy "Current account"

3.1.Units used

3.1.1. For Uranium. The nuclear decay of uranium isotopes occurs with such a long period that this decay can be neglected. As enriched uranium is only used exceptionally for mixed oxide fabrication, it will not be considered here.

The units used are :

- total uranium (U_{total}) : quantity of uranium element contained in the material, expressed in grams.
- isotope ^{235}Uranium (^{235}U) : quantity of the 235 isotope of uranium in the material, expressed in grams.

It is evident that a simultaneous balance in both these units can only be achieved if the isotopic composition of the incoming and outgoing materials is exactly the same. The pool type management of nuclear materials in the BELGONUCLEAIRE plant makes this very unlikely.

Three ranges of ^{235}U content are thus defined :

- near natural uranium : $0.65 < \% \, ^{235}U < 0.75$
- slightly depleted uranium : $0.35 < \% \, ^{235}U < 0.65$
- depleted uranium : $\% \, ^{235}U < 0.35$

Within each of these ranges, only the U_{total} is taken into account for material balance.

3.1.2.For Plutonium and Americium. All the isotopes of plutonium as well as americium, its daughter product, decay with periods that have to be taken into account for the material accountancy. The loss of material due to radioactive decay is always attributed to the customer who supplied the material. Summing and subtracting of quantities of plutonium and/or americium, whatever the unit, can only be done when all parts have been calculated to the same date.

The units used are :

- Total Plutonium (Pu_{tot}) : quantity of plutonium element (excluding americium) contained in the material, expressed in grams. This unit is used for Safeguards purposes in the accounting for the EURATOM Supply Agency accountancy which is done on the basis of the contracts concluded for one or several campaigns.
- fissile Plutonium (Pu_{fiss}) : quantity of the isotopes ^{239}Pu and ^{241}Pu contained in the material, expressed in grams. This unit is the one traditionally used for customs purposes, possibly for Safeguards purposes.
- equivalent Plutonium (Pu_{eq}) : quantity of element summing the content of each plutonium isotope and of americium, multiplied by the energetic equivalent coefficient of the corresponding isotope. The formula is of the form :

$$Pu_{eq} = (a \times {}^{238}Pu) + (b \times {}^{239}Pu) + (c \times {}^{240}Pu) + (d \times {}^{241}Pu) + (e \times {}^{242}Pu) + (f \times {}^{241}Am)$$

where : - ^{238}Pu, ^{239}Pu, ... are the weights, of each isotope; expressed in grams;
- a, b, ... f, are the energetic equivalence coefficients for each corresponding isotope.

NB : Pu_{fiss} is a particular case of Pu_{eq} where : $a = c = e = f = 0$
$b = d = 1$

In practice the energetic equivalence coefficients are more complex and can vary for various types of reactors. It is however proposed to use the following standardized average set of values for commercial accountancy purposes :

$a = -1$
$b = 1$
$c = -0.4$
$d = 1.3$
$e = -1.4$
$f = -2.2$

As in the case of Uranium, a simultaneous balance in all units is impossible.

NB : This effect would be observed even without the pool type of fissile management. It would be due to :
- the fact that all the by the customer supplied lots of Pu will not have been used in the same proportion;
- analysis inaccuracies.

Therefore one of these units must be used on a commercial basis for balancing inputs and outputs towards each customer. Pu_{eq} is the unit representing the energetic value of the fissile material and must thus be chosen for commercial accounts balancing purposes.

3.2.Commercial in/out accountancy

An example of balance for Plutonium is given in appendix 1. Inputs to the plant (material supplied by the customer) are positive, outputs from the plant are negative. Waste and archives are considered as outputs and are supposed to have the same isotopic vector as the product delivered by the BELGONUCLEAIRE plant. All data are recalculated at a same date (reference date of the current campaign), inputs and outputs in the three units are cumulated. The balance of the Pu_{eq} is the quantity that has to be transferred to the next campaign for the same customer, or returned to the customer (last campaign).

3.3.Transfer from one campaign to the next one, for the same customer

The balance of Pu_{eq} at the end of a campaign must be transferred to the next one which has usually a different reference date. Decay must thus be calculated from one reference date to the other and for this purpose the material to be transferred must be attributed an isotopic vector and a quantity of Pu_{tot} which correspond to the to be transferred quantity of Pu_{eq}.

It is supposed that the material transferred has the same isotopic composition as the product delivered by the BELGONUCLEAIRE plant for the completed fabrication campaign; the quantity of Pu_{tot} is calculated to give the required Pu_{eq} quantity.

The so defined transferred material is the first input in the Pu material balance of the next campaign.

3.4.Balancing the other plutonium units

Balancing the Pu_{eq} unit settles all energetic and thus financial accounts but for Safeguards and customs purposes the Pu_T and Pu_{Fiss} must also be balanced, this is physically impossible. The balance in those two units must thus be brought back to zero by the transfer, by mutual agreement of the parties, of fictitious material from the debtor to the creditor at zero cost.

Remarks

a) The settlement of the accounts must preferably occur at the end of each campaign, occasionally the balances for the Pu_{tot} and Pu_{fiss} of several campaigns can be cumulated in such an agreement.

b) The ratio between the quantities of Pu_{tot} and Pu_{fiss} can never correspond to a physically plausible Plutonium isotopic vector as they have a Pu_{eq} value of zero.

4.Conclusion

In order to harmonize different units in use in the nuclear material accountancy of BELGONUCLEAIRE the system of "current accounts" expressed in Pu_{tot}, Pu_{fiss} as well as Pu_{eq} was applied during the last 5 years. The data are used for safeguards as well as for commercial purposes.
During this period about 6 tons of plutonium were processed in the MOX-Fuel Fabrication plant in Dessel for different customers and for a different fabrication campaigns. This "current account" system proved to work successfully.

5.Reference

1. R. Lefèvre "Commercial Nuclear Material Accountancy" private communication
BN-reference -/-/46/n/285D

2. H. De Canck - R. Ingels - J. Beckers
A "real time" nuclear accountancy system in application at the BELGONUCLEAIRE plutonium fuel fabrication plant.
Proc. ESARDA-Symposium on Safeguards and Nuclear Material Managements, p. 499-503, April 1979 Brussels.

APPENDIX 1

"CURRENT ACCOUNT"

1) IN- AND OUTPUTS

CUSTOMER X

	LOT NUMBER	NR.	TRANSPORT	PuTot(g)	PuFiss(g)	ANALYSE DATE	%238	%239	%240	%241	%242	ppm Am	SOM ISOTOPEN
CAMP.1	M220-1	1	23-Feb-89	100000.0	74880.0	21-Jan-87	0.200	70.330	23.950	4.550	0.970	360	100
CAMP.1	503-A	2	18-May-89	116000.0	77352.3	02-Oct-87	1.330	56.837	26.722	9.846	5.265	229	100
CAMP.1	X-01		23-Mar-90	-200000.0	-143188.0	31-Jul-90	0.786	64.766	24.403	6.828	3.217	7500	100
CAMP.1	ARCHIVES		23-Mar-90	-67.1	-48.0	31-Jul-90	0.786	64.766	24.403	6.828	3.217	7500	100
CAMP.1	WASTE		23-Mar-90	-287.3	-205.7	31-Jul-90	0.786	64.766	24.403	6.828	3.217	7500	100

2) RECALCULATION TO REFERENCE DATE

CALCULATION DATE	PuTot(g)	PuFiss(g)	%238	%239	%240	%241	%242	ppm Am
31-Jul-90	99268.6	74163.1	0.1959	70.8410	24.1175	3.8685	0.9771	7490
31-Jul-90	114497.6	75893.2	1.3177	57.5781	27.0645	8.7056	5.3341	12898
31-Jul-90	-200000.0	-143188.0	0.7860	0.6477	24.4030	6.8280	3.2170	7500
31-Jul-90	-67.1	-48.0	0.7860	0.6477	24.4030	6.8280	3.2170	7500
31-Jul-90	-287.3	-205.7	0.7860	0.6477	24.4030	6.8280	3.2170	7500

3) CALCULATION OF PU EQUIVALENT

WEIGHT Pu238	COEFF. Pu238	Pu.eq. Pu238	WEIGHT Pu239	COEFF. Pu239	Pu.eq. Pu239	WEIGHT Pu240	COEFF. Pu240	Pu.eq. Pu240	WEIGHT Pu241	COEFF. Pu241	Pu.eq. Pu241	WEIGHT Pu242	COEFF. Pu242	Pu.eq. Pu242	WEIGHT Am241	COEFF. Am241	Pu.eq. Am241
194.47	-1	-194.47	70322.87	1	70322.87	23941.10	-0.4	-9576.44	3840.21	1.3	4992.27	969.95	-1.4	-1357.93	743.52	-2,2	-1635.74
1509.73	-1	-1509.73	65925.54	1	65925.54	30988.20	-0.4	-12395.20	9967.70	1.3	12958.01	6107.42	-1.4	-8550.38	1476.79	-2.2	-3248.98
-1572.00	-1	1572.00	-129532.00	1	-129532.00	48806.00	-0.4	19522.40	-13656.00	1.3	-17752.80	-6434.00	-1.4	9007.60	-1500.00	-2.2	3300.00
-0.53	-1	0.53	-43.46	1	-43.46	-16.37	-0.4	6.55	-4.58	1.3	-5.95	-2.16	-1.4	3.02	-0.50	-2.2	1.11
-2.26	-1	2.26	-186.07	1	-186.07	-70.11	-0.4	28.04	-19.62	1.3	-25.51	-9.24	-1.4	12.94	-2.15	-2.2	4.74

4) PuTot, PuFiss, PuEq.

	LOT NUMBER	TOTAL PuTot	TOTAL PuFiss	TOTAL Pu.eq.
IN	M220-1	99268.6	74163.1	62550.6
IN	503-A	114497.6	75893.2	53180.2
OUT	X-01	-200000.0	-143188.0	-113882.8
OUT	ARCHIVES	-67.1	-48.0	-38.2
OUT	WASTE	-287.3	-205.7	-163.6

5) CURRENT ACCOUNT CUSTOMER "X"
BALANCING CAMPAIGN 1

CURRENT ACCOUNT "X"					31-Jul-90				31-Jul-90							
				**	CREDIT TO THE CUSTOMER			**	DEBIT TO THE CUSTOMER			**	BALANCE OF CREDIT TO THE CUSTOMER			
			LOT	**				**				**				
	NR.	TRANSPORT	NUMBER	**	PuTot(g)	PuFiss(g)	PuEq(g)	**	PuTot(g)	PuFiss(g)	PuEq(g)	**	DATE	PuTot(g)	PuFiss(g)	PuEq(g)
CAMP.1	1	23-Feb-89	M220-1	**	99268.6	74163.1	62550.6	**	0.0	0.0	0.0	**	31-Jul-90	99268.6	74163.1	62550.6
CAMP.1	2	18-May-89	503-A	**	114497.6	75893.2	53180.2	**	0.0	0.0	0.0	**	31-Jul-90	213766.2	150056.3	115730.8
CAMP.1		23-Mar-90	X-01	**	0.0	0.0	0.0	**	200000.0	143188.0	113882.8	**	31-Jul-90	13766.2	6868.3	1848.0
CAMP.1		23-Mar-90	ARCHIVES	**	0.0	0.0	0.0	**	67.1	48.0	38.2	**	31-Jul-90	13699.1	6820.3	1809.8
CAMP.1		23-Mar-90	WASTE	**	0.0	0.0	0.0	**	287.3	205.7	163.6	**	31-Jul-90	13411.8	6614.6	1646.2
CAMP.1			TRANSFER	**	0.0	0.0	0.0	**	2891.0	2069.8	1646.2	**	31-Jul-90	10520.8	4544.8	(0.0)
CAMP.1			REGULARIS	**	0.0	0.0	0.0	**	10520.8	4544.8	0.0	**	31-Jul-90	(0.0)	(0.0)	(0.0)
CAMP.2			TRANSFER	**	2891.0	2069.8	1646.2	**	0.0	0.0	0.0	**	31-Jul-90	2891.0	2069.8	1646.2
CAMP.2		29-Nov-89	532(021R)	**	6968.2	5275.6	4413.8	**	0.0	0.0	0.0	**	31-Jul-90	9859.2	7345.4	6060.0

SAFEGUARDING LARGE PLUTONIUM STORES

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Abstract

Within the European Communities there are presently under construction or commissioning several large plutonium stores. A generic Pu store design is described which takes into account typical features of importance to safeguards. The different safeguards measures used, or to be used in the near future, to safeguard such stores are reviewed.
Different store designs are characterized according to a number of important parameters, i.e. storage location design, handling machines and routines, access routes for personnel, nuclear and non-nuclear material, etc. and each safeguards measure is presented and analysed in order to derive a clàssification of the aptness of each measure.
Particular emphasis is placed on the role of "inter-locking" safeguards measures which treat as a specific requirement the avoidance of unnecessary re-verification through a judicious selection of complimentary techniques and instruments.

1. Introduction

The purpose of this paper is to examine the problems associated with the safeguarding of the large plutonium stores in the commercially operating plutonium handling plants that are presently under construction or have been recently commissioned. These stores have been designed to incorporate the necessary radiation protection measures for the reduction of radiation doses to the plant operating staff and include remote handling techniques to reduce the need of human presence inside the stores to a minimum. As a consequence of the design and plant automation in the storage areas the classical approach of individual item or location sealing together with NDA measurement systems using mobile NDA equipment in the storage area had to be fundamentally re-examined. This together with the incorporation of the new instrument technology available led to the development of a safeguards system compatible with the new automated handling sequences and systems in the storage areas.

The layout of the paper is based upon the sequence and iterative development processes in the approach to safeguarding these large plutonium stores (see Fig. 1). The following areas are discussed in some detail :

a) Technical and accounting information about the plant
b) Safeguards approach
c) C & S measures
d) NDA measures
e) Project management
f) Inspection activities

The results of the studies and work carried out by the Inspectorate, as described in this paper, will show the evolution of the safeguarding framework arising from the approach considerations utilising the available technology for both NDA and C & S aspects.

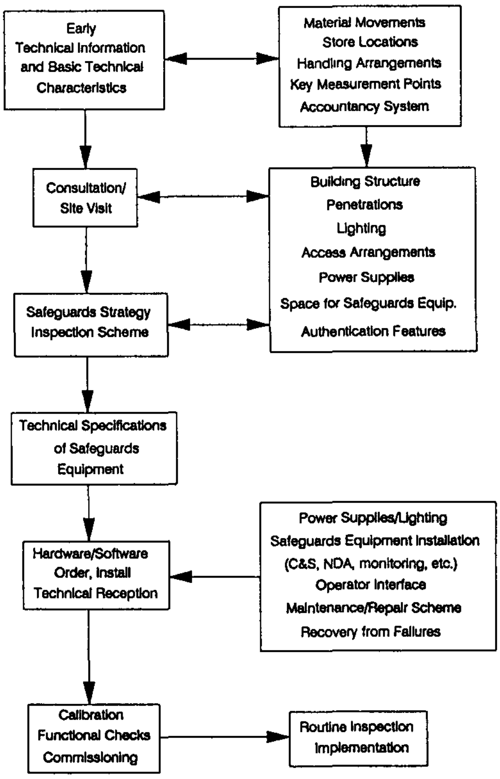

Fig. 1 Flow Diagram of Principal Decision Points in the Establishment of a System for Safeguarding Large Plutonium Stores

2. Examination and Discussion of Technical and Accounting Information of the Plant

A consultation/discussion with the plant operator at an early design and construction stage was considered vital and necessary for a satisfactory development and implementation of the safeguards approach . This consultation was essential in the identification of the material flow routes, key measurement and verification points, the possible locations for non-routine access, the understanding of the container handling and transfer, the storage location layout, and the interaction of the material access routes in relation to the processing areas of the new plants. During these consultations the identification of possible locations for equipment to be installed was made, together with the expected location of the inspectors office, to ensure that in the subsequent deliberations as many options as possible were examined for the safeguards approach.

During the first discussion and subsequent discussions more exact details concerning the quantities of material to be stored and configuration of the individual storage locations were confirmed together with the storage container design. These details were essential for the design of the C & S system and the measurement configuration for any proposed installed NDA system.

In addition to the verification of the mechanical and physical details of the store, questions concerning the reliability of the electrical power available to the inspectors and linkage to the operators emergency back-up system were examined and discussed with the operator in some detail. This was deemed absolutely crucial to ensure that there was the necessary security of electrical power supply. In the absence of certainty of guarantee of electrical supply there was a need for the inspectors to design and incorporate a certain level of power back-up in their safeguards system.

At this early stage of the discussions on the plant design the question of branching from the operators installed equipment, e.g. for NDA, merited examination for the inspectors use. The problems of authentication had to be studied and examined in detail before the construction and installation was completed. These discussions, defining the interface between operators/inspectors signals was considered the most important and the most useful by all the operators involved.

Based upon this preliminary information derived from the consultations and the ensuing discussions with the operator an approach scheme for safeguarding the plant was drawn-up and discussed.

3. Safeguards Approach

The large plutonium stores recently commissioned or presently under construction differ in some distinct relevant safeguards features from previous fuel stores for the following reasons:

1) The necessity for personnel access to the store has been significantly reduced for physical protection, health physics, and resource reasons

2) As a consequence of (1) above, the new stores have multiple containment systems and complete remote operations of fuel transfers

3) The classical approach for (manual) verification of cans with mobile equipment and the subsequent sealing of items in their individual storage positions has therefore become obsolete.

4) The re-establishment of an inventory in such stores would be a very resource and cost intensive exercise for operators and inspectorates. Seals, which have proved to be very reliable cannot, in general, be used on the material locations inside the store. This places a new and highly significant emphasis on the design of very reliable alternative systems.

5) The increasing use of unattended safeguards systems in these stores necessitates more effort from technical services to maintain a high level of reliability, but it reduces, on the other hand, the need for continuous presence of inspectors in some facilities. The transmission of data to a central office significantly reduces the amount of time spent by the inspectors passing through controlled zone areas and following the necessary change of clothing routines.

The general scheme designed by Euratom for these large stores is based upon the following basic principles:

- 100% verification of flow into and out of the store by unattended Non Destructive Assay neutron and gamma measurements

- 100% identification of flow into and out of store either by optical surveillance and/or number reading techniques

- Highly reliable Containment and Surveillance measures to maintain continuity of knowledge for the material in the store and follow-up of any internal movements of cans/containers inside the store. The design goal with respect to reliability is to have a C & S system where the failure probability is at the maximum of 1 per life time of the plant.

- The physical inventory verification consists essentially of examination of C & S results and the verification of some randomly selected items from the inventory and this activity can bee distributed over a material balance period. A schematic layout of such a store is shown in figure 2.

The scheme for safeguarding these large plutonium stores was therefore based upon built-in redundancy, diversity, and separate interlinking systems to preserve the continuity of knowledge of the material inside the stores. Account was taken of the need for timeliness, with respect to the material residency time within the store and the frequency of shipment/exports from the storage area. From the inspectors point of view this was acknowledged by the requisite for the necessary regular review of the results of the surveillance equipment and diagnostic examination of the performance of the installed instrumentation.

One of the most important features taken into consideration for detailed examination in the development of an approach was the difficulty of access to the material once inside the storage locations of the store. This aspect of the facility design and the logistics of handling arrangements inside the store had an impact on the cost/time penalty for the re-establishment of inventory in the case of surveillance failure.

The overall approach taken was that of an object cocooned by a number of protective shells, interlinked but at the same time independent for redundancy purposes. These shells of containment would be linked by monitored passageways from the outside to the inner sanctum of the store area and vice versa. The concept of the establishment of a number of containment barriers with sensors monitoring for penetration was therefore established and instituted.

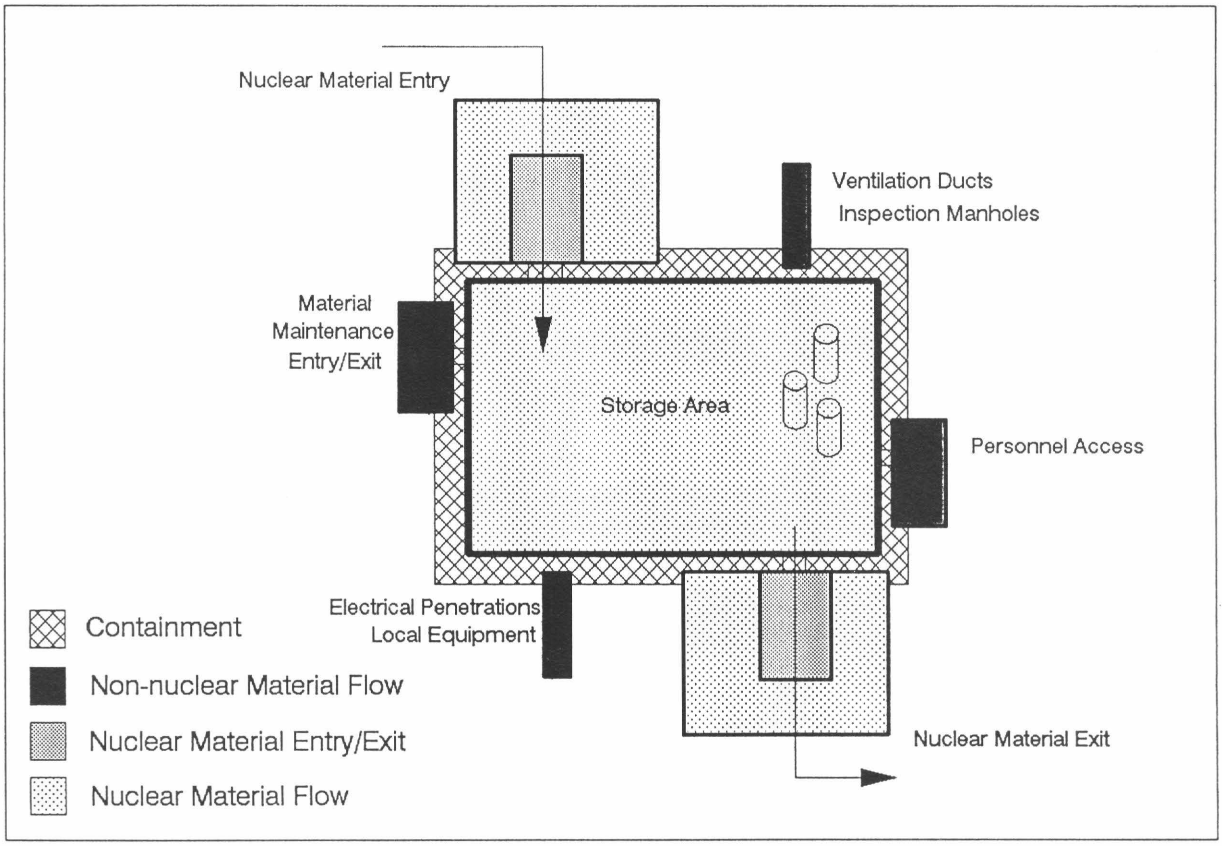

Fig. 2 Physical Layout of Store

The overall C & S system was designed to achieve at least a category **B** type C & S system. This was based upon the measurement of material entering the storage area and using several methods for C & S, including the installed NDA as a monitoring device outside the measurement activities, to ensure the continuity of knowledge.

The proposed safeguards measures for a store are shown in Fig. 3.

4. C&S Considerations

One of the main considerations for the application of C & S in these large stores was the absence of the realistic opportunity due to operational limitations of placing classical seals on the individual storage locations. An alternative sealing method based upon different methods of containment were necessary for study under the umbrella of the safeguards approach as discussed in the previous section. Having established the overall safeguards approach, and identified/specified the equipment to be utilised, the store was examined in detail by the safeguards inspectorate for the optimum siting of the equipment.

The necessity for sealing, using conventional seals was not abandoned but the use of seals was restricted to accessible areas so that inspectors could readily verify them without undue intrusiveness to the operator.

The C & S system installed was designed to provide the necessary surveillance containment inside the store and to cover the different access routes in/out of the store namely:

a) nuclear material flow into and out of the store

b) Personnel access

c) Ventilation ducting

d) maintenance hatch/doors

e) inspection manholes

as well as an overview of the store and storage locations.

The C & S systems utilised were based upon the standard Euratom video system, together with front-end motion detection, active infra red for intrusion monitoring, and neutron and gamma counting for the installed NDA equipment. The systems were incorporated into an overall scheme so that any failure of the video system would be backed-up by techniques such as intrusion monitors and neutron detectors.

Due to the problems associated with the spiking in the operator's power supply great importance was attached to the need for the inclusion of electrical filtering into the inspector's system.

The camera system installed was based upon a standard commercially available CCD camera linked through a multiplexer and motion detection system to GYRR time lapse recorders. The cameras and recording systems were designed with an appropriate

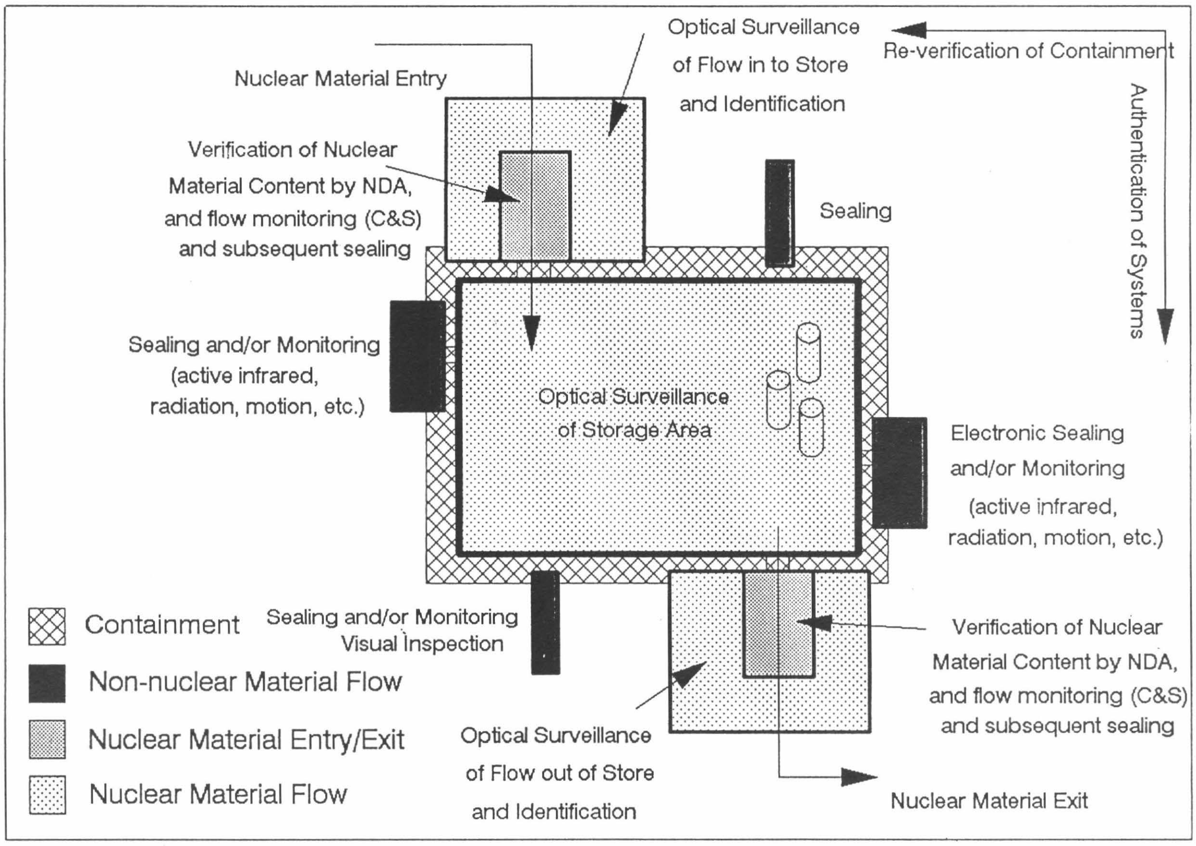

Fig.3 Safeguards Measures for the Store

redundancy based upon experience and historical data from similar installed video systems.

The infra-red detection system utilised was the curtain type that was capable of detecting an object greater than 5cm passing through the beam.

As previously stated the inclusion of the installed NDA operating as a monitor outside the programmed measurement cycles was an important feature for the linkage into the overall C & S strategy.

It was important that the interlinking of the different C & S systems into an overall scheme ensured that the likelihood of a total loss of continuity of knowledge was reduced to an absolute minimum. As previously mentioned recovery from a total or even a partial failure of surveillance will be extremely difficult and time consuming for both the inspectorate and the operator.

The reliability of the electrical power supply, lighting, and adequate emergency power back-up were also important factors in the design of the overall C & S system. This aspect prompted considerable discussion throughout the realisation of the projects and sufficient back-up power or emergency supply through the operator had to be available to power an adequate C & S system during the periods of normal power outage.

5. Input and Output Measurements

The decision on the installation location of the NDA instrumentation for the measurement of the material entering or leaving the store was dependent upon the engineering layout of the store, the operational sequences, and the space available. The location of the inspector's own equipment was established during the subsequent discussions to the early plant design visit. The siting of the equipment was linked to the proposed C & S measures and would cover the material flow into and out of the store. The design of the neutron measurement system was based upon the HLNCC design of instrumentation with an accuracy of 1-2% and the gamma measurement system on high resolution spectroscopy using the MGA code.

The specification of the installed NDA system included the monitoring function step to dove-tail into the other C & S devices included in the overall safeguards scheme. As previously discussed this provided one of the protective containment shells to the inner store area. The measurement procedure was established on the cycle time of the material container transfer and the geometry of the container presented for the inspectors measurement. The signal transfer of the neutron and gamma signals was transmitted either to a local sited PC or the inspectors central computer system for recording and evaluation. The NDA measurement seqûence would be triggered as part of the monitoring cycle to permit measurements both going in and out of the store.

In the case of the branched option for the NDA measurement from the operator's equipment, appropriate authentication modifications had to be made to the branched signal to ensure that the equipment was triggered in the correct sequence and at the correct time. Internal monitoring of the NDA equipment through the inspectors software would alert the inspectors to unusual events or detector failure.

Emphasis on the NDA measurement evaluation and data interpretation with reference to the operators declaration should be made towards the independence of the results rather than a comparison of two sets of derived figures, particularly in the case of the branched measurement system.

6. Project Management

Due to the complex nature of the proposed C & S and NDA installations associated with the safeguarding of the stores it was necessary to establish a proper project management scheme. This entailed the drafting of appropriate specifications for the different items for installation and the incorporated NDA equipment. With particular reference to the NDA installations this involved exploring new areas for unattended mode operation and data acquisition for both neutron and gamma measurements.

After the establishment and agreement of the equipment specifications tenders were sought for the installation of the safeguards equipment. The offers in comparison with similar projects enabled the inspectorate to evaluate the reasonableness of the offers in relation to the overall costs of installation, the design and engineering component of the total cost figure, and the associated services installation. The capital costs presented about 30-40% of the overall investment in safeguarding these large plutonium handling plants. The remainder of the money invested being absorbed in the installation and design costs. Based upon the capital and installation costs an estimate was made for the forecast running costs and maintenance of such equipment over the next 5/10 year operating period.

After detailed scrutiny and examination of the offers, together with further discussions with the suppliers and tenderers concerning the justification of the offer figures, approval for the expenditure was sought from the budget authority. After acceptance contracts were placed and the project for installing safeguards equipment was instituted to dovetail into the overall construction project of the new plants.

7. Inspection Activities

The routine inspection activities would include:

- Check of internal consistency of operator records
- Check of consistency of records with other MBAs
- Check of consistency between records/reports

The internal consistency check would include the comparison of weight/identity/content records of storage items against data gathered from the inspector's system. This would involve the frequent assessment of the C & S results together with the correlation of the NDA results and monitoring information to confirm and reconcile the declared movements into and out of the store.

The inspector would compare the NDA results obtained from either from branched or independent measurement to the operators declared figures. The establishment of this comparison is essential for the judgement on the operator's materials accounting system.

Where problems have been identified with respect to measurement results, these should be followed up, possibly implying repeat measurement.

On the occasion of the Physical Inventory Taking (PIT), sufficient items should be remeasured to provide confidence that the safeguards system is indeed functioning as envisaged.

8. Further Remarks and Future Developments

The major new technical features for safeguards equipment in remotely operated fuel stores are the unattended operation mode and the integration of the equipment in the operators fuel handling, transfer, measurement or surveillance equipment.

Due to the restrictions in the use of seals, system reliability is a fundamental requirement in remotely operated stores. The new generation of the twin recorder multiplexed video systems in operation reach reliability values between 96% and 98%. (The reliability of the twin Minolta camera system has now reached a figure of 99.2%)

A video system with a reliability of 95% would not be acceptable as the only C & S system in such a store, given that approximately each 1.5 years an equipment failure can be expected. If this video system was combined with an NDA/monitoring system, with a 95% reliability, covering the same safeguards objective the remaining total failure rate would be about 1 in 30 years, the operating lifetime of the plant. Careful attention is required in the design of such systems to avoid common mode failures, such as total power or light failures. The use of a properly electrically filtered Uniteruptable Power Supply unit would overcome the problems of total power failure.

Further development work is required in the following areas:

1) Where sensors are used to trigger optical surveillance or measurement equipment, means are required to check in an un-attended way the performance of the sensor in question (i.e. normalisation, calibration or diagnostic check). In the case of an unattended neutron detector system, 3 very small Cf-sources were integrated and sealed into the detectors to provide an independent test/reference signal.

2) A systematic fault tree analysis should be performed for the safeguards systems used in plutonium stores to optimise the reliability of systems and to determine the optimum combination of measures.

3) A systematic performance evaluation needs to be carried out to monitor and maintain the reliability of the installed systems.

4) The equipment status needs to be transferred to the inspectors office on or off-site to permit quick interaction in case of diagnostic messages or failures.

5) Authentication techniques need to be further developed as proposed by the Euratom Safeguards Directorate in the paper presented at the INMM in 1990.

9. Conclusions

The conclusions of this paper are that:

a) Early access for plant design information is required to ensure that the necessary equipment and cabling can be installed before the plant becomes active.

b) In the case of branching operators NDA equipment early access is also required so that the necessary authentication can be made.

c) The whole scheme must be properly project managed to ensure that the equipment and installation is specified in detail.

d) The equipment installed must be either capable of lasting the lifetime of the plant or the cabling accessible for equipment change/upgrading during the lifetime of the plant.

e) The NDA system should be designed to act in a monitoring function and be interlinked into the overall C & S system to maintain continuity of knowledge.

f) The overall safeguards system must be designed in such a way that the loss of continuity of knowledge for the whole system is reduced to an extremely low probability of once in the operating lifetime of the plant.

g) The electrical power supply system must have either the necessary guarantees for non-interruption or be backed-up by a sufficiently large power supply to cover interruptions to the electrical supply.

h) The inspection activities will place a much greater reliance on the installed instrumentation than in previous similar locations due to large number of individual items and the complexity of moving and individually identifying them.

A SURVEY OF NDA INSTRUMENTATION USED ROUTINELY BY THE EURATOM SAFEGUARDS DIRECTORATE

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Abstract

A systematic review of the different nuclear material categories, physical and chemical forms, and container descriptions held by nuclear installations in the European Communities is presented along with the non-destructive instrumentation (NDA) currently used for their verification. Material categories will be identified where presently used NDA verification techniques are insufficiently accurate or where no adequate NDA technique yet exists. Based on this analysis the future needs for NDA instrumentation are discussed, particularly with respect to the new large-scale nuclear facilities presently under construction or in commissioning.

1. Introduction

The purpose of this paper is primarily to outline the present and future needs for NDA instrumentation. This is done by comparing the types of nuclear material held in the European Community with the capabilities of existing NDA instrumentation. The aim of the paper is to present those areas where improvements are required in NDA instrumentation; it does not attempt to assess the needs in terms of, for example, detector numbers. The paper begins with a description of the nuclear installations in the Community and then gives the types and forms of nuclear material likely to be found in a set of generic installations which cover the most important types. This is followed by a very brief description of the NDA instruments in use by the Euratom Safeguards Directorate Luxembourg. The paper concludes with a list of areas where new or improved NDA instrumentation is needed.

2. European Community Nuclear Installations

The number of each type of facility in the European Community is given in table 1.

Type	Number
Research Laboratories	58
Research Reactor and Critical Assemblies	68
Mines and Concentration plants	26
Enrichment plants	5
Fuel Conversion/ Fabrication plants	27
Reprocessing plants	11
Power Reactors	128
Storage Installations	52
Others	420
Total	795

Table 1 European Community Nuclear Installations

NDA measurements are carried out at all of the major types of installation as part of the Euratom inspection activities.

The various types of installation, representing the most important in terms of nuclear material usage, are now presented in more detail; The types of material expected to be present in seven different types of generic facilities are given. Material types are relevant to NDA requirements if they appear in the inventory of the plant or in transfers to and from the plant in non-negligible quantities. Each of the subsequent sections also gives the NDA measurement capability associated with each material type; the detector type and expected operator/inspector difference of the measurement are given. The detectors are identified in table 9. Unless otherwise stated, the quantity measured with the neutron equipment is the plutonium or U-235 mass. In the case of the gamma instruments the measured quantity is normally the enrichment in the case of uranium or the isotopic composition for plutonium. Only neutron and gamma techniques have been included in the tables, which means that weighing, for example, is excluded even though it may play a major part in the verification of some materials.

(i) Research Reactors

The description of the expected material in a research reactor is given in table 2.

Material	NDA Method
Fresh HEU fuel assemblies:	AWCC 3%, Davidson+Ge 0.5% Gamma scanner enrich. 1%
Irradiated HEU fuel assemblies:	none
Fresh scrap	AWCC 20% Davidson+Ge 1%

Table 2 Research Reactor material types.

The AWCC measures the bulk U-235 content and the Davidson + Ge detector measures the uranium enrichment. The gamma scanner measures the enrichment and the U-235 content. In the case of scrap, it should be noted that the interpretation of the enrichment measurement will depend to a large extent on the nature of the sample. Similar techniques would be used at a HEU fabrication plant.

(ii) Enrichment Plants

The description of the expected material in an enrichment plant is given in table 3.

Material	NDA Method
UF6 cylinders:	
Depleted U	CIND 15-20% Davidson+NaI 10%
LEU	CIND 8-10% Davidson+Ge 3%
Natural U	CIND 15-20% Davidson+NaI 8%
In-process material	Cascade meter Go/NoGo for LEU
UF6 waste	none

Table 3 Enrichment plant material types.

The categories in this case are limited by the nature of the enrichment process to UF6 feed, product, tails and waste. Out of the process, the UF6 is generally stored in cylinders. It should be noted that the CIND measures the enrichment in a small fraction of the volume of the cylinder. The Davidson MCA plus detector measures the uranium enrichment in each case. The cascade meter is used to confirm that the uranium enrichment is less than 20% (within 3 standard deviations of the measurement).

(iii) Power Reactors

The description of the expected material in a power reactor is given in table 4.

Material	NDA Method
Fresh LEU fuel assemblies	NCC (active) 2-5%; Davidson+NaI 3%
Fresh MOX fuel assemblies	NCC (passive) 2% Davidson+NaI 3%
" " (underwater)	Mod. Fork 5%
Irradiated fuel assemblies	Ion-fork attribute
Irradiated pins	none

Table 4 Power Reactor material types.

Power reactor material types are very limited. They are fresh fuel assemblies (LEU or MOX) and irradiated assemblies. Sometimes pins are removed from irradiated assemblies and stored in boxes on site. The NCC measures the U-235 content of a certain length of fuel, which is combined with an active length measurement to give the total U-235 content.

(iv) LEU Fuel Fabrication plants

The description of the expected material in an LEU fuel fabrication plant is given in table 5. (All the material is unirradiated).

Material	NDA Method
Powders:	
LEU and Natural U	PHONID 2% Davidson+NaI 2%
Ceramic pellets:	
LEU and Natural	PHONID 2% " "
Fuel rods:	
LEU and Natural	Operator rod scanner; Davidson+NaI 2%
Fuel assemblies:	
LEU	NCC active 3-5% Davidson+NaI 2%
Waste, scrap and small quantities	PHONID 20%

Table 5 LEU Fuel Fabrication plant material types.

PHONID measures the bulk U-235 content of the samples.

(v) MOX Fuel Fabrication plants

The description of the expected material in a MOX Fuel Fabrication plant is given in table 6. (All of the material is unirradiated).

Material	NDA Method
Powders:	
UO2	PHONID 2%
PuO2	HLNCC 2% Cicero/Pu meter 1%
MOX	HLNCC 2% Cicero/Pu meter 1%
Ceramic pellets:	
MOX	HLNCC 2% Cicero/Pu meter 1%
Fuel rods:	
MOX single pins	Cerceuil <2%; HM4 for active length
MOX pin trays	UFBC 2% Cicero/Pu meter 1%
Fuel assemblies:	
MOX LWR	NCC passive 2% HM4 for active length
FBR	UFBR 2%
Scrap and waste	HLNCC Octagone 20%

Table 6 MOX Fuel Fabrication plant material types.

The Cicero or the Pu meter are used with MGA (reference 1) which gives uncertainties of less than 1% on all plutonium isotopic abundances depending on the burn-up and the measurement time. For high burn-up material the errors increase to 2-3% if the Pu-242 is calculated from correlations. Improved correlations are presented at another paper in this conference (reference 2).

(vi) Reprocessing Plants

The description of the expected material in a reprocessing plant is given in table 7.

Material	NDA Method
Irradiated fuel elements:	
LEU (Pu)	Ion-Fork attribute
Irradiated fuel rods	none
Irradiated nitrate solutions	Hybrid K-edge Pu 0.7% U 0.3%
Fresh nitrate solutions	
Depleted U	none
Pu	HLNC INVS 2%; Cicero/Pu meter 1% K-edge 0.2-0.3%
Fresh powders:	
PuO2	HLNC INVS 2%; Cicero/Pu meter 1%

Table 7 Reprocessing plant material types.

Due to the active nature of the material at such a plant, a large amount of material is difficult to access directly (e.g. large tanks of active solutions) therefore even when NDA techniques are available, sampling is required. The Hybrid K-edge measures the concentration of materials in solution and has an accuracy of 0.3% for uranium and 0.7% for plutonium. The K-edge has an accuracy for plutonium of 0.2-0.3%.

(vii) Research Laboratories

The description of the expected material in a research laboratory is given in table 8.

Material	NDA Method
Fresh fuel rods:	
HEU	AWCC 2-3%; Davidson+Ge 0.5%
LEU	NCC active 2%; Davidson+NaI 3%
Irradiated fuel rods:	
LEU and Natural	none
Fresh ceramic pellets:	
LEU and Natural	AWCC 2%; Davidson+NaI 3%
Fresh metal:	
Depleted U	none
HEU	AWCC 3%
Natural U	AWCC 3%
Fresh powders:	
UO2	AWCC 5%; Davidson+NaI 3%
Thorium	Davidson+NaI attribute
Scrap and waste	AWCC 30%

Table 8 Research Laboratory material types.

This list does not attempt to give all possibilities of material types which exist in research laboratories but gives an indication of the diversity of the material types. In general the more unusual forms exist in relatively small quantities.

3. Euratom Safeguards NDA Equipment

Figure 1 shows the breakdown of current usage of NDA equipment by installation type. Figure 2 shows how the usage of NDA equipment has increased over the past 7 years. Table 9 lists the types of NDA equipment which are currently in use in the Euratom Safeguards Directorate.

Fig. 1 Usage of NDA by Installation

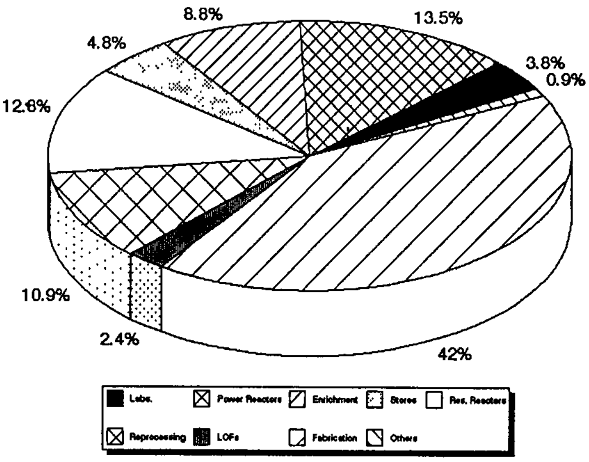
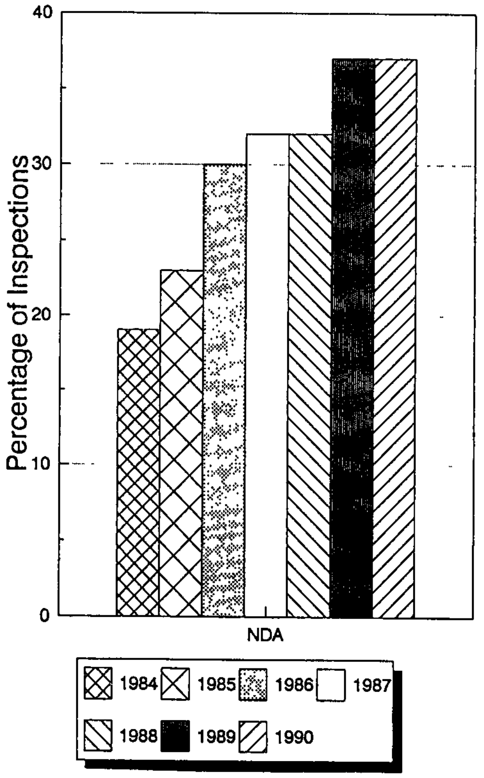


Fig. 2 Usage of NDA 1984-1990



It is not possible in the space available to describe the method of operation and the applications of each of the pieces of equipment. Therefore only a brief description of each class of equipment will be given here.

(a) Passive Neutron
The bulk of the passive neutron measuring equipment consists of well-known detectors developed at Los Alamos Laboratory: HLNC, UFBC, INVS, and NCC(passive). The performance of these systems is very good for well-characterized material giving an

Equipment	#in use	Typical usage	ref
Passive Neutron			
HLNC	13	Fresh Pu and MOX, powders, pellets, solutions	3
INVS	2	as HLNC for small samples	4
NCC passive	4	Fresh MOX fuel assemblies	5
UFBC	4	as NCC + large MOX powders FBR assemblies	6
Cerceuil	4	MOX fuel rods	7
Ion-fork	6	Spent fuel assemblies	8
Ion-fork (mod)	1	Fresh MOX assemblies (under water)	9
Octagone	1	Pu waste	10
Active Neutron			
AWCC	5	Fresh MTR fuel, HEU	11
NCC active	3	Fresh LEU fuel assemblies	12
CIND	3	UF6 cylinders	13
PHONID	3	UO2 powders, pellets, metal	14
Gamma			
Cascade meter	1	U enrichment	15
Cicero + HPGe	7	Pu isotopics	16
Enrichment meter	2	U enrichment	17
Gamma scanner	2	MTR fuel active length	18
HM4	7	U, Pu, values	19
Davidson + NaI	20	U enrichment	20
Pu meter	6	Pu isotopics	21
Rod scanner	1	Rod active length	18
K-edge	1	Fresh Pu and U solutions	22
Hybrid K-edge	2	Irradiated Pu, U solutions	23

Table 9 List of Euratom Safeguards NDA equipment

overall accuracy of 1 - 2%. This is as good a performance as is likely to be achieved in the medium term for such sample materials as clean PuO2 powder, pellets, clean MOX powder pellets, plutonium nitrate solutions, fuel rods and assemblies. One important feature of nuclear material which is not included in the description above and which is important in particular cases is the storage location. This can make access with certain instruments difficult or impossible. For example the usage of underwater storage for fresh MOX fuel elements at reactors has made the use of the standard neutron coincidence collar impossible and created the need for a new measurement method. The problem has been treated with good effect by the use of a modified ion-fork.(reference 9).
The standard ion-fork is used to verify the burn-up and cooling time of spent fuel assemblies in cooling ponds. The accuracy on the burn-up is about 5%.

(b) Active Neutron
The active neutron systems which are in use, comprise some Los Alamos systems: AWCC and NCC (active mode) which allow satisfactory measurements of fresh MTR fuel elements and fresh LWR elements. Other important active neutron instruments are CIND (Cogema) for measuring UF6 cylinders and PHONID (Ispra) which is used for uranium samples.

(c) Gamma Measurements
These instruments supply information on plutonium isotopics and uranium enrichment. In some situations of fixed geometry, they can be used to measure the nuclear material content of items.

Further information on the status of these instruments as used by Euratom can be found in references 24 and 25.

4. Non-destructive Assay Needs

NDA capabilities have been improved over a number of years and therefore have been able to play a wider role in the overall safeguards strategy. Further improvements such as greater accuracy or decreased measurement time could always be utilised to expand their role even more, giving the benefits of improved timeliness of the results and the reduced need for destructive analysis. Inspection of the tables 2-8 shows a number of areas where Euratom could benefit from improved NDA performance:

- the measurement of the bulk fissile content of irradiated HEU assemblies.

- Irradiated LEU fuel assemblies can only be measured in 'attribute' mode; i.e. operator values of burn-up and cooling time can be verified but no measurement of the fissile content is made. A method to improve this situation (using complex fixed instrumentation) is mentioned below.

- the measurement of the bulk U-235 content of UF6 cylinders.

- the measurement of irradiated fuel rods or non-liquid samples of irradiated material.

- suitable techniques for the measurement of waste and, in particular, scrap are very limited. In new plants, however, the fraction of waste material is much reduced and tends to be better segregated making measurement less difficult.

Other points which may be mentioned regarding the status of NDA instrumentation are as follows:

There is a need for more sophisticated portable instrumentation such as a hand-held multi-channel analyser for rapid, simple, qualitative measurements of plutonium and uranium.

The performance of the passive neutron measurement systems is not adequate for samples with unknown impurities (such as moisture or fluorine). This makes the measurement of waste in particular difficult. Work is under way (references 26,27) to extract more information from the pulse train from the neutron detectors, and this may be one way to fulfil this requirement.

There is a need for neutron coincidence detector systems having larger custom-designed measurement cavities.

The calibration of the neutron collar when used in active mode for the measurement of LWR fuel assemblies is not readily transferable between different fuel assembly designs. This necessitates a calibration for many fuel element designs, consuming significant effort in each case. In addition for a single fuel assembly design the effect of different burnable poisons also requires individual assessment. It also means that the measurement of a 'scrap' fuel assembly (assuming some change in the configuration of the nuclear material) would not be possible by NDA without dismantling it back to pins. This is an area where neutron transport calculations may enable the extension of existing calibrations to new situations. However it would be necessary to verify the input parameters (e.g. the number of burnable poison pins) for such a method to be usable.

It should be borne in mind that there are requirements in terms of size and ease-of-use in order for non-installed equipment to be suitable for inspection use.

The special requirements for large-scale facilities are dealt with in the following section.

5. Large-scale Facilities

Large-scale bulk-handling facilities present special requirements in terms of NDA equipment. Large amounts of material are processed which makes a large number of measurements necessary. At the same time, the design of the facility is such that the material is not readily accessible. In the latest generation of bulk-handling facilities many of the operations are automated and Euratom has taken the opportunity, at the design stage, to include automated NDA stations. These have the advantage of being able to measure 100% of the material flow at various parts of the plant. In addition, because they are no longer required to be transportable, it is possible for them to be much more sophisticated. For example, whereas at the output of a reactor the spent fuel can be monitored with an ion-fork, at the input to a modern reprocessing plant, the fuel can be measured with a high resolution gamma detector and an active neutron interrogation technique.
The automated approach involves a large amount of preparatory work before the equipment becomes operational. In general the design of each detector system is unique to the particular part of the plant to which it is applied which leads to a large collection of different detectors to commission and calibrate. In terms of the control software for these systems, it is an important consideration to ensure standardisation of the user interface and not to underestimate the developments required to ensure data security and performance monitoring.

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THE "MAILBOX" FOR RANDOMISED SAFEGUARDS INSPECTIONS*

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Abstract

Enhancements in both the effectiveness and efficiency of international safeguards verifications of inventory changes at natural or low-enriched uranium conversion and fuel fabrication plants are possible by means of short-notice random inspections. Accordingly, a plant operator would declare the contents of nuclear material items involved in transfers before knowing if a safeguards inspection would occur to verify them. Such declarations would be deposited into a literal or figurative "mailbox", whose characteristics are important for sustaining the validity of the randomisation procedure. An inspection performed successfully in accordance with an approved randomisation procedure should permit the extrapolation of verification results to unverified batches included in the operator's declarations.

1. Introduction

The necessity for statistical inference procedures in connection with safeguards inspection activities at a nuclear facility arises because of time and cost limitations imposed on these activities. During an inspection, the inspector records data consisting of numerical values associated with randomly selected items that he or she measures by non-destructive assay or obtains chemical samples for later analysis. The set of all possible data is called the population, and inferential statistical theory is used to make statements about the population based on the data derived from the randomly selected items, i.e., the sample. The term "population" is also used to refer to the group of items from which the sample is drawn. (This paragraph is in part directly quoted from /1/).

Of course the underlying goal of these procedures is to verify the plant operator's declarations of values for nuclear material contained in inventory or transfer items, i.e., the state of control of the plant material accountancy system /2/. Random selection of items from a population permits this verification to be done efficiently.

In possible additional applications of randomisation, the population can consist of possible dates of inspection and possible facilities to inspect. In most cases, it may be more appropriate to think of these extensions as more complicated ways to choose the sample of nuclear-material items. Thus the problem generally becomes one of a two-stage sampling procedure. The first stage is to choose, for example, dates to carry out an inspection to verify inventory changes, and the second stage is to sample items for actual verification.

As an example, verification of the plant material balance for a bulk-handling facility depends upon verification of the feed to the plant process (or receipts at the plant) and the plant production (or shipments from the plant) as well as the beginning and ending inventories. If the plant handles indirect-use material (natural or low-enriched uranium) exclusively, then frequent inspections for inventory change verification are not feasible in a situation of constrained safeguards inspection resources, including both inspectors and travel.

Under such circumstances, a regime of short-notice random inspections (SNRIs) for inventory change verifications can permit a valid conclusion about the state of control of the plant operator's material accountancy system based on verifications of a limited number of items. The regime would be carried out by having short-notice inspections occur at randomly selected times throughout the year guided by the plant's operational programme.

It is the purpose of this paper to explain the importance to SNRIs of declarations of nuclear material prior to the initiation or deliberate foregoing of an Agency safeguards inspection.

2. Conditions for the Application of Randomisation

For the valid application of randomisation to safeguards verifications, three conditions must be met /3/ (see also Annex F of /4/). These apply both to simple sampling from a stratum as well as to the additional applications of randomisation:

a) All items in the population must have a non-zero (generally equal) likelihood to be chosen for verification.
b) The plant operator must declare to the Agency values for the nuclear-material content of items before knowing if they are to be verified.
c) The operator must not alter item identity or content after learning that an item is chosen for verification and before the verification actually occurs.

The second condition, a material accountancy declaration before notice of inspection, is encompassed in the notion of a "mailbox".

The procedure for SNRIs and the "mailbox posting" must be such that the operator does not know, prior to his posting of the material accountancy values to the mailbox,

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* This paper reflects the opinion of the authors, not necessarily those of the IAEA or its Member States

whether the Agency inspectors will come to verify the material batches in question. If they do come, this would happen during some residence time - possibly very short - after receipt or production at the facility and after the posting of material accountancy values, when the batches are available for verification.

For random inspections requiring a mailbox, the mailbox information would ideally be "posted" as soon as the plant operator knows the content of items. This could mean immediately upon receipt, production or measurement, as appropriate. The "mailbox" must be equipped with a time clock, enabling a verification after the operator declared the consignment but before the actual date of the shipment or feeding into the process.

The information posted should be that necessary for verification purposes for those items newly available: dates, nuclear material content, and identification number. Information which is of relevance only for the measurement method used for verification, e.g., zones of enrichment and the distribution of neutron poisons for fuel assemblies, can be supplied after the random selection of items for verification.

Given the existence of a mailbox procedure, the first condition is satisfied if there is an agreed residence time for item verification after the mailbox posting about the item in question. Safeguards inspections can be planned in advance on the basis of an average residence time for verification with a realistic distribution of values for different items. However, satisfying this condition leads to the need for an absolute minimum value of this residence time to permit verification at an SNRI: one full day after the posting.

Regarding the third condition, the time between notification of an inspection and the initiation of verifications would have to be established on the basis of practical implementation procedures, but it should not be longer than the time it would take the operator to carry out "credible" diversion activities.

If the three randomization conditions are met, verification results can be extrapolated to unverified batches in the operator's declaration.

3. Practical Realisation of a "Mailbox"

Several possible realisations of the mailbox concept are possible.

Existing methods include, first, the ordinary mail. This would involve either paper or computer disk declarations and transmission to another location. Second would be data transmission by telefax.

Methods requiring minor development include, first, a sealed box at the plant site that records the date and time of submission of records by the plant operator. Recording of the date and time could also be by means of a built-in surveillance system. Accordingly, the plant operator would "post" declarations in, say, serially numbered envelopes. The surveillance system would periodically make images of the top of the stack of envelopes and include date and time information by means of a self-contained clock.

Other methods requiring minor development include a remote Agency computer to which a plant computer or terminal communicates data, or a tamper-proof Agency computer at the plant site to which the plant computer or an operator directly inserts information. In these situations involving a computer, the information, once in memory, must be unalterable; amending information can, however, be submitted. Because of their flexibility and compatibility with other needs, the computer solutions are the most desirable over the long term.

4. Advantages of Randomisation of Inspections for Inventory Change Verification

In contrast to the situation with verification of inventory changes when all available items in a stratum available at an inspection constitute the population for sampling /4/, inspection randomisation permits an efficiency of benefit to the Agency, the plant operators, and any accompanying personnel of the State System of Accounting for and Control of Nuclear Material (SSAC).

Because the plant operator would be regularly submitting inventory change data to the mailbox and because each batch of inventory change items would be available for verification for some agreed residence time, the population of items to be sampled for actual verification need only consist of the batches whose residence time overlaps with the beginning of the inspection. Items from earlier batches that happen still to be present at the facility need not be part of the population subject to random selection for verification. The reason is that the randomness in timing of inspections means that the earlier batches were subject to verification at an earlier date and data giving their nuclear material contents were submitted to the mailbox. Of course this batch distinction makes item identification particularly important.

This point is more graphically explained in the situation illustrated in Figure 1 for a fuel fabrication plant. After mailbox posting of their nuclear material content batches 11 through 13 of fuel assemblies were in residence for possible verification at the time of the beginning of the second inspection. These three batches would constitute the population for random sampling during the second inspection. Any assemblies still present from earlier produced batches, however numerous, need not be part of the population for sampling, because they were either subject to verification at earlier times during randomly timed inspections (such as the first inspection), irrespective of the number of such inspections actually conducted, or they were available for verification even if an inspection did not occur. Batches 14 (a borderline case) and 15 were also in residence for verification during the second inspection, but randomization condition 2 (see Section 2) would not apply; the fabrication of these two batches and posting of their mailbox data did not precede the beginning of the inspection.

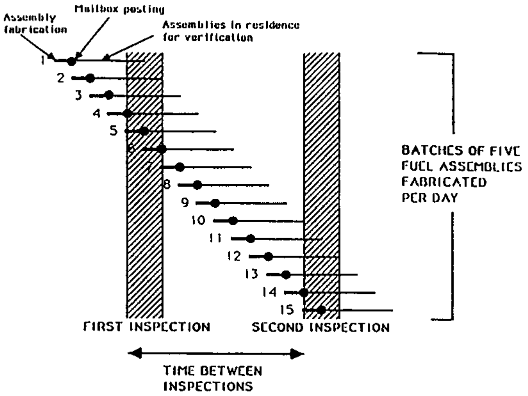

Figure 1. Schematic diagram of the availability for verification of fuel assembly batches at randomly timed safeguards inspections

In comparison to a situation where far from all inventory change items during a material balance period are available for verification, the inspection randomisation regime offers a dramatic increase in effectiveness by virtue of the complete (100%) coverage. (The quantitative defect detection probabilities for inventory change strata would depend on item and hypothesized defect sizes, inspection frequency and residence times for verification. Detailed quantitative considerations are not addressed here.)

In comparison to a situation where all inventory change items are available for verification at regularly scheduled inspections, usually achieved only in facilities handling unirradiated direct-use material, the randomisation regime yields a decrease in inspection effort and a decrease in detection probability.

5. Practical Problems of Randomisation of Inspections for Inventory Change Verification

First and foremost, implementation of the inspection randomisation scheme described here is not possible without the full cooperation of the plant operator, the SSAC, and the inspectors. This would be based on mutual understanding and agreement concerning the benefits to the three parties.

Practical aspects associated with randomised inspections can mitigate their value, or, more precisely, can call into question the necessary conditions for the validity of the inspections.

There would be the need to modify facility attachments to accommodate the additional aspects of randomisation. One key need would be advance notice of domestic transfers to facilitate SNRIs for inventory change verification.

Implementation of any randomisation scheme requires advance discussions and clear understandings of procedures by the SSAC and the facility operator, with detailed instructions and working papers, and provision for certain notifications as required, in particular, by the mailbox.

The inspection schedule for SNRIs must be maintained secret. Given this secrecy, then the whereabouts of the inspectors must not be known to the facility or SSAC before any given inspection. This requires that inspectors not be required to go through extraordinary border formalities /5/.

Within the facility, practical verification difficulties may occur if the plant operator is not prepared for the inspection. This could lead to much waiting time by inspectors. However, arrangements could be codified in advance based on the (announced) general programme of inspections.

Some practical inspection problems are the same for both announced inspections and SNRIs. One problem is that a fuel fabrication plant with one crane for moving finished fuel assemblies could probably not operate routinely if that same crane were required to move assemblies for safeguards verification.

Another problem is that items are sometimes stored during their residence time in ways inconvenient for verification. Fuel assemblies may be in crates and powder cans, in drums. Opening the containers for verification may be timeconsuming or may contravene quality-control regulations.

The requirement for a minimum residence time may be in conflict with the effort by fabrication plant operators to shorten material residence time for cost-saving reasons. Shorter residence times would reduce the achievable defect detection probability for a given inspection frequency. In the limit of very short residence times (one day), this could mean that certain batches might systematically be unavailable for verification: for example, those produced on Friday if inspections are not feasible on weekends. Systematic unavailability of items would violate the first condition for the application of randomisation.

6. Potential for Field Trials

Agency inspectors carried out SNRIs for inventory change verification at the General Electric LEU Fuel Fabrication Plant in the USA for more than two years /6/. Though these inspections demonstrated the workability of the short notice feature, prior mailbox declarations associated with agreed residence times were not part of this field trial.

Field testing of the mailbox concept remains necessary, and it is expected that this will be done either in the framework of a Member State Support Programme or as a supplement to actual Agency inspections of an appropriate facility.

7. Acknowledgement

One of the authors (LGF) is grateful to Mr Jonathan Sanborn for a discussion about certain points in the paper.

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Simulation of Inspection Randomization Models

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ABSTRACT

A computer simulation of a model light water reactor fuel cycle is described which allows a realistic estimate of the impact of alternative inspection randomization models on inspectorate resources and safeguards effectiveness and efficiency. The program simulates the flows and inventories of nuclear material through the fuel cycle dynamically, as well as basic safeguards activities carried out by the operators and the inspectorate. Results of simulation experiments involving current IAEA inspection procedures and different randomization schemes are presented.

1. Introduction

A number of global, as opposed to facility-oriented, approaches to IAEA safeguards implementation have been proposed as a means to maintain effectiveness under manpower and resource constraints. They include the super-MBA concept (or zone appraoch, see e.g. /1/), randomization over interim inspections /2/, over inspection activities /3/ and randomization over entire fuel cycle facilities (the RFUV model /4,5/). In addition to the more formal problems of compatibility with safeguards agreements and IAEA effectiveness criteria, these 'fuel cycle approaches' also raise practical problems with regard to efficient planning, coordination and implementation of inspections.

Field experiments involving alternative global safeguards strategies would be very difficult to carry out, partly because of the IAEA's own performance criteria, which tightly constrain the required inspection activities, and also due to boundary conditions imposed by facility attachments.

In the present work, a computer simulation of a nuclear fuel cycle was developed to allow a more realistic estimate of the impact of alternative inspection regimes on inspectorate resources and safeguards efficiency, without the cost and inconvenience of actually implementing such regimes. The program simulates the flows and inventories of nuclear material through the fuel cycle dynamically, as well as the basic safeguards activities of closing and reporting material balances, verification by routine on-site inspection, application of C/S, etc. The program is written in an object-oriented style and can therefore be easily modified to accomodate new fuel cycles and safeguards strategies.

The simulation program and the associated reference fuel cycle are described in Section 2. Section 3 illustrates the use of the simulator for investigation of some typical inspection strategies. Results are discussed in terms of inspector resources, travel costs, quantification of effectiveness, consistency with existing safeguards agreements and practicability.

2. The Simulation Model

2.1. THE REFERENCE FUEL CYCLE

Fishbone and Higinbotham, in their investigation of zone fuel cycle approaches /1/, presented a reference fuel cycle (referred to in their report as the 'second reference cycle') which proved to be very convenient as a basis for our simulation program. Their fuel cycle is based on an electrical power generating capacity of about 20 Gigawatts with associated uranium-conversion and fuel fabrication facilities. A large away-from-reactor store and a

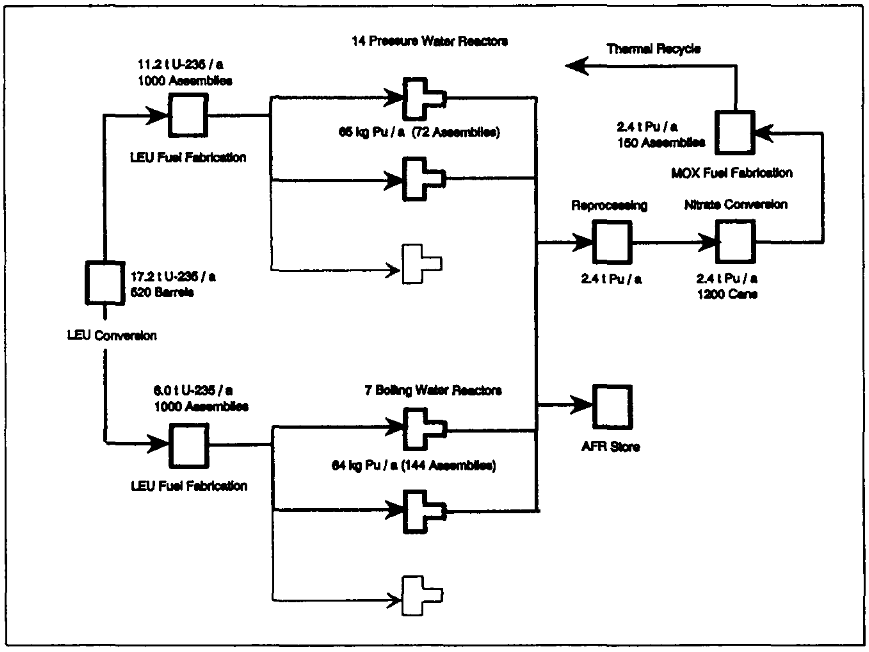


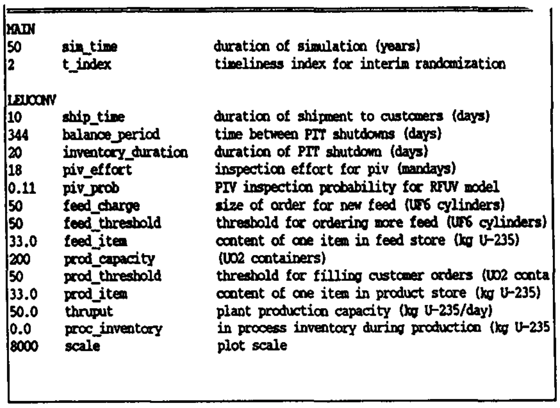
Fig. 1: The reference light water reactor fuel cycle, as modified from /1/.

single reprocessing facility feeding mixed oxide fuel production plants constitute the back-end. As this reference cycle evolved from prior studies within the IAEA, rough estimates of required inspection efforts for verification activities for the various facility types are also available /6/. In the original version of the fuel cycle /1/, reprocessed plutonium was merely stored for future use in an unspecified fast breeder system. In order to make the present investigation more contemporary, all reprocessed plutonium is thermally recycled to some of the light water reactors. Figure 1 shows an overview of the modified reference fuel cycle, including yearly throughput data. A portion of the input data file for the program is shown in Figure 2.

2.2. THE SIMULATION PROGRAM

The fuel cycle simulator runs on an IBM PC and is programmed in Pascal. In order to allow for process-oriented, discrete event simulation in the style of the classical simulation language SIMULA-67, a front end system was written, partly in machine language, to emulate the basic simulation class constructs of SIMULA /7/. The object-oriented facilities are provided by the Pascal compiler itself /8/.

The process-oriented discrete event simulation paradigm used in the present work consists of the creation and scheduling of (pseudo)parallel processes which represent, for example, fuel cycle facilities, inspectors, transport vehicles, etc. Control is always given to the currently active process. An active process will typically update its state variables, perhaps create, activate or reschedule other processes, and then reschedule itself. The passage of time in the simulation corresponds to control passing from the current process to the next one scheduled. Simulation terminates with the exhaustion of all actions of the last process on the time axis. The encapsulation and class inheritance offered by object-orientation and the parallel-



```
MAIN
50      sim_time              duration of simulation (years)
2       t_index               timeliness index for interim randomization

LEUCONV
10      ship_time             duration of shipment to customers (days)
344     balance_period        time between PIT shutdowns (days)
20      inventory_duration    duration of PIT shutdown (days)
18      piv_effort            inspection effort for piv (mandays)
0.11    piv_prob              PIV inspection probability for RFUV model
50      feed_charge           size of order for new feed (UF6 cylinders)
50      feed_threshold        threshold for ordering more feed (UF6 cylinders)
33.0    feed_item             content of one item in feed store (kg U-235)
200     prod_capacity         (UO2 containers)
50      prod_threshold        threshold for filling customer orders (UO2 conta
33.0    prod_item             content of one item in product store (kg U-235)
50.0    thruput               plant production capacity (kg U-235/day)
0.0     proc_inventory        in process inventory during production (kg U-235
8000    scale                 plot scale
```

Fig. 2: An input data file for the simulation program.

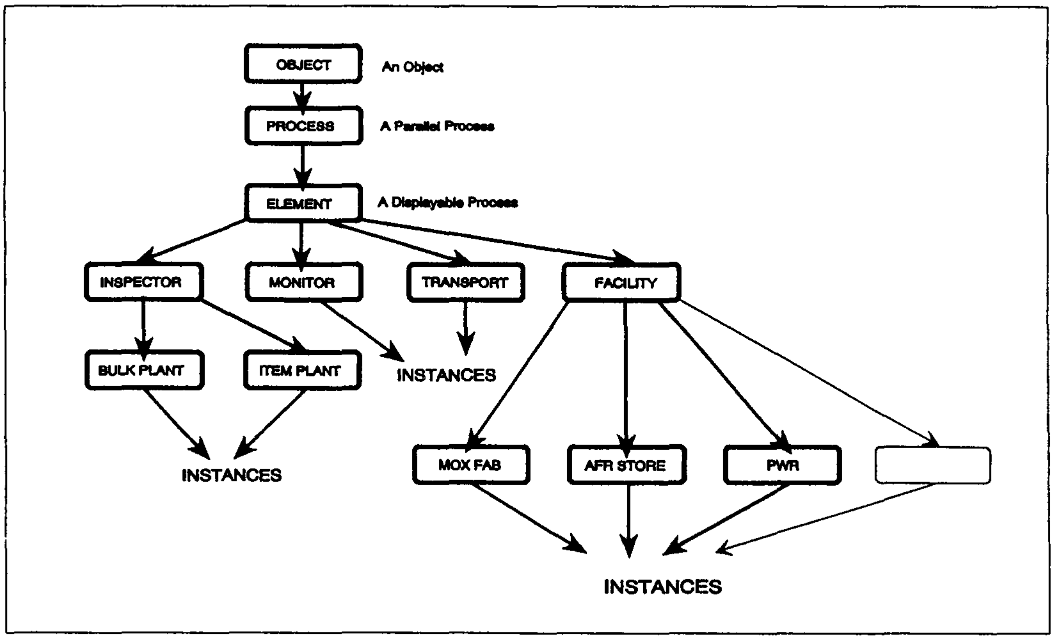


Fig. 3: The object class hierachy of the simulation program.

lism of the SIMULA constructs allow for programming of very complicated activities with relative ease. Program modification is also very simple. For example, a new nuclear facility of a given pre-defined class can be created, woven into the fuel cycle and activated with just a few lines of code.

The class hierarchy stucture of the program is shown in Figure 3. The processes which take part in the simulation are instances of the various object classes defined in the heirarchy. For example there are 14 instances of the class PWR, simulating the 14 pressure water reactors in the fuel cycle. They all descend ultimately from class PROCESS, meaning that they can run independently in parallel, and from class ELEMENT, from which they inherit the ability to display their status on the graphics screen. They communicate with other instances by sending messages. The PWR instances, for example, place orders for more fresh fuel at the LEU and MOX fabrication plants after refuelling, inform the relevant IAEA inspector when a plant shutdown occurs, create transport vehicle instances to ship spent fuel to the reprocessing plant or away from reactor store, and so on.

To illustrate some of the detail involved in the simulation, Figure 4 shows a screen capture plot of the feed, process and product inventory as well as inspection profile for one of the LEU fabrication plants over a five year period. The other fuel cycle facilities are simulated at a similar level of detail.

The simulator can be interrupted at any time and detailed status reports displayed for any process. An example, again for the LEU fabrication plant, is shown in Figure 5. Simply clicking with the mouse on any facility displayed on the screen will, without interrupting the

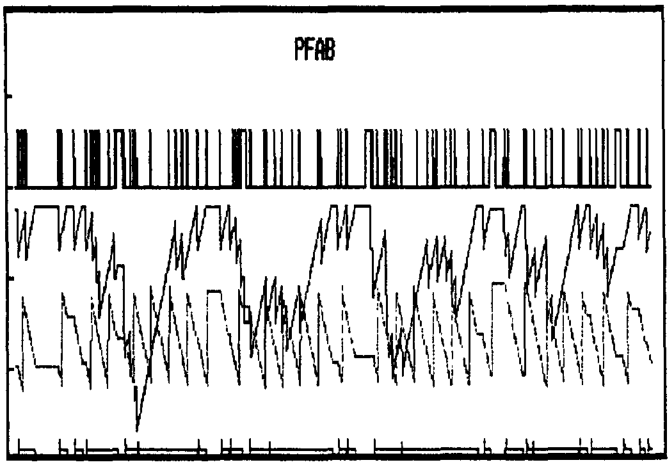


Fig. 4: Simulated key measurement inventories at a LEU fuel fabrication plant as a function of time. The histogram at the top indicated inspection visits.

simulation, open a small window showing its status (e.g. on-line, shutdown, taking inventory) and the amount of material currently present at its various key measurement points. XY-plots of key measurement point inventories (e.g. Figure 4) as a function of time can be displayed for any facility, again without interruption of the program.

Aggregate statistics on inspection effort, travel time, material under safeguards, etc. are collected by the inspector instances and written to an output file at the end of each calendar year. A typical record is shown in Figure 6. These statistics, averaged over several years of simulated fuel cycle activity, form the basic data for comparison and evaluation of alternative safeguards strategies.

3. Safeguarding a Digital Fuel Cycle

The application of the simulator will be illustrated with simulation experiments involving the following safeguards strategies:

Base Case: The reference strategy consisting of an ideal facility-oriented safeguards regime. All physical inventories are verified, all bulk facilities processing low enriched uranium are inspected for flow verification, and all item facilities containing plutonium are subjected to interim inspection for timeliness – every month if unirradiated direct use material is present and every 3 months otherwise. The sensitive facilities of the back-end, when in operation, are subject to permanent inspection.

Interim Randomization: Interim inspections at item facilities, carried out for reasons of timeliness of detection, are randomized according to a game theoretical analysis /9/. All other base case activities remain unchanged.

Reduced Frequency Unannounced Verification: As described in /4,5/ the inspectorate's physical inventory verification activities are randomized according to the saddlepoint solutions of a zero sum game. In calculating the randomization probabilities, the goal quantity for diversion/detection is taken to be independent of material location within the fuel cycle, and international standards of accountancy are assumed to be met or exceeded.

The base case strategy is the limiting case as the randomization probabilities in the other two strategies go to 100 %. It will not be discussed in more detail here.

```
STATISTICS for pfab on 30.12.1990

 Inspectors present: 0
 Inspection effort in calendar year:     62 mandays

Material Balance Report (Bulk Facility)
 Last PIT:        4081.00   Completed on 24.6.1990
 Material received: 4587.00
 Number of receipts:      4
 Material shipped:   5544.00
 Number of shipments:     7
 On Inventory:       3124.00
 MUF (numerical):      0.00
 Sigma MUF:           13.76

Next PIT scheduled for  3.6.1991
```

Fig. 5: A typical status report for a fuel cycle facility.

```
STATISTICS for IAEA on 27.12.1994

Current strategy is base case
 Inspectors in the field: 5
 Cumulative effort in this calendar year:   1493 mandays
 Cumulative travel in this calendar year:    203 mantrips
 Facility ICRs received in this calendar year: 125
 Facility MBRs received in this calendar year: 27 verified: 27
Material Under Safeguards
 LEU                            25575.00 kg U-235   341.0 SQ
 IDU in reactor cores           13305.60 kg Pu     1663.2 SQ
 IDU outside reactor cores      19334.40 kg Pu     2416.8 SQ
 DU (separated Pu)               3443.40 kg Pu      430.4 SQ
Current inspection randomization probabilities
 Facility  P_piv     P_int
 conv      1.00      1.00
 pfab      1.00      1.00
 bfab      1.00      1.00
 repr      1.00      na
 ncon      1.00      na
  mox      1.00      na
  afr      1.00      1.00
   pl      1.00      1.00
   bl      1.00      1.00
Expected number of PIVs in this calendar year =  28.00
PLARIE (Planned effort for PIVs in this calendar year) = 192.00
ARIE (Effort actually expended to date): 202.00
```

Fig. 6: A report on accumulated inspection statistics within one calendar year, generated by an inspector instance.

3.1. INTERIM RANDOMIZATION

Assume that the detection probability for an abrupt diversion from an MBA is 1 for any inspection of that MBA, that n_1 opportunities for an inspection are available within some reference period and that the inspector will take advantage of precisely k_1 of them, deciding on a random, unannounced basis whether or not to inspect at any given opportunity. Then it can be shown to be optimal to inspect at opportunity i with probability

$$\frac{k_i - 1}{n_i - 1} \tag{3.1}$$

where n_i is the number of opportunities remaining and k_i the number of inspections not yet used prior to the ith

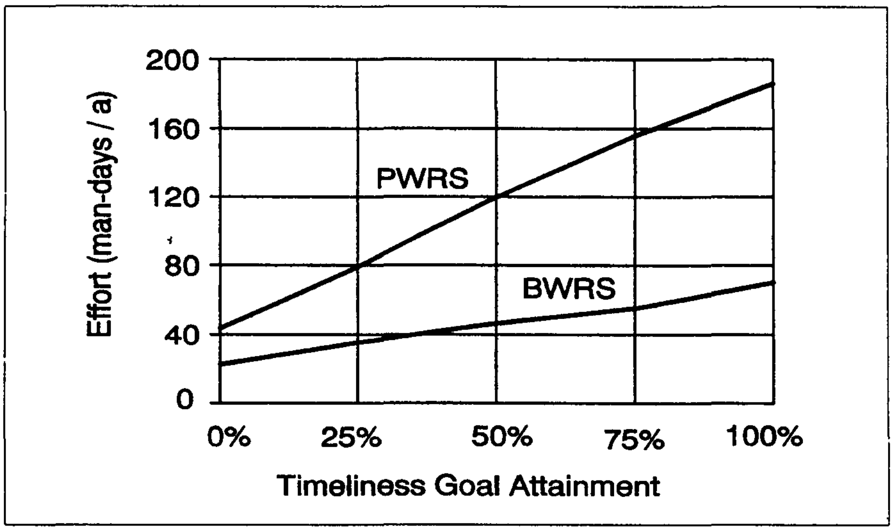


Fig. 7: Inspection effort as a function of timeliness for item facilities. See text for details.

opportunity /9/. (The ratio $\frac{0}{0}$ is understood to be 1 so that the last opportunity is always used.) The guaranteed mean time to detection, at the beginning of the reference period, can then be shown to be

$$\frac{n_l}{k_l} \qquad (3.2)$$

measured in intervals between opportunities.

This strategy was simulated for the item facilities (reactors and AFR store) of the reference fuel cycle, with the reference period taken to be one material balance period for facilities with irradiated direct use material, or, for reactors with fresh MOX fuel, the fraction of a balance period for which such material was present at the site. The number of opportunities n_l was chosen such that the current timeliness goals would be met if every opportunity were used, for example at 3 month intervals for irradiated Pu. A range of k_l values was then chosen between

a) full attainment of current timeliness goals, i.e, the base case strategy (this corresponds to $k_i = n_i$, or no randomization at all (base case), as can be seen from equation (3.1)), and

b) timeliness of detection determined by the frequency of PIV inspections only. This corresponds to $k_l = 1$, or no interim inspections at all (equation (3.1)).

A graph of inspection effort at the item facilities as a function of the degree of attainment of the timeliness goals is shown in Figure 7. A goal attainment of 0 % is understood to mean that only PIV inspections are carried out. The item facilities at which fresh MOX fuel is present for at least some of the time (the PWRs) are distinguished from those having only irradiated Pu on inventory (BWRs and AFR store).

The statistical error on the two curves in Figure 7 is about 8 %, corresponding to a simulation time of 5 calendar years. The greatest relative saving in effort by randomization is obtained for the direct use item facilities. It is almost a factor of 5 for 0 % timeliness (no interims whatsoever) as opposed to a factor of 2 for the irradiated direct use facilities.

The travel costs (measured by the simulator in inspector man-trips per year) exhibit analagous behavior, although results are far less realistic since the program does not (yet) simulate inspection itinerary optimization: each inspection generates one man-trip.

The random unannounced inspections can be calculated, using equation (3.1), by the inspectorate in advance, allowing for efficient planning. (They would have of course to be kept secret.) Regarding consistency with existing safguards agreements, randomizations of this kind present no difficulties, since the *opportunities* are in any case allowed for in the facility attachments. The present IAEA internal performance evaluation criteria set rather strict limits on the requirements for interim inspection frequency and these would have to be relaxed in any ac-

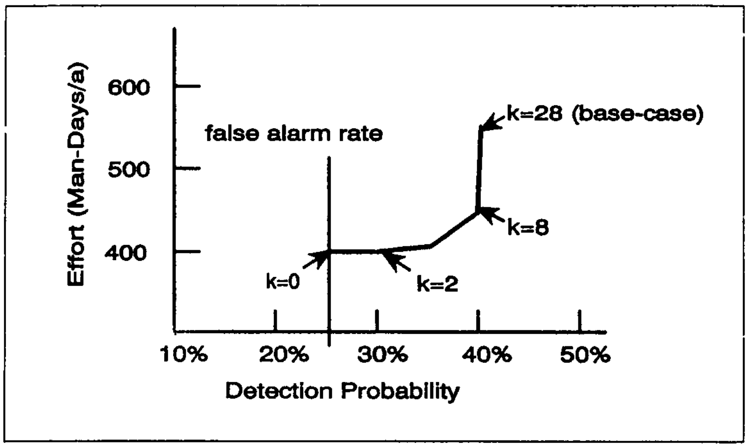


Fig. 8: Inspection effort as a function of overall detection probability for diversion from the fuel cycle. See text for details.

tual implementation of such a scheme. One might justify such a relaxation with the subjective argument that, from the operator's point of view, an inspection might occur at *any* opportunity so that a considerable deterrence effect is present. Strictly (i.e. game theoretically) speaking, of course, the objective performance of the scheme is simply that given by equation (3.2).

3.2. RANDOMIZATION OVER FACILITIES

The Reduced Frequency Unannounced Verification model /4,5/ has to do with randomization over physical inventory verifications (PIVs) at the facilities in the fuel cycle. Based on the *a priori* probabilities of detection for diversion of one significant quantity from any one facility, the optimal inspection randomization strategy for PIVs is determined as a function of the total number k of facilities to be inspected within one reference period (taken to be a calendar year), there being n facilities in all. The calculated strategies are determined game theoretically and hence automatically focuss inspection effort on the most sensitive facilities, i.e. those most attractive to the diverter. (Of course *all* facilities receive finite PIV inspection probabilities in any given strategy.) See /4,5/ for more details.

Figure 8 shows the total annual inspection effort for the fuel cycle as a function of the achievable overall detection probability for diversion of one significant quantity. Not included in the inspection effort is the effort associated with permanent inspection at the back-end bulk facilities (dynamic accountancy, verification of internal flows or whatever), although the effort for verification of facility receipts and shipments is included for all facilities.

The *a priori* detection probabilities for the bulk handling facilities were calculated by assuming the significant quantities for low enriched uranium and plutonium given in the IAEA Glossary /10/, and furthermore that the international standards of accountancy (also given in the Glossary) are bettered by a factor of two. Detection probabilities associated with PIVs at the item facilities were taken to be 100 %. The false alarm rate indicated in the figure is for the fuel cycle itself, and corresponds to 5 % per year per facility for 6 bulk handling facilities, or 26 %.

The efforts were determined by simulating the RFUV model for several different k-values over a period of ten years of fuel cycle operation and averaging. Statistical errors are about 5 %. From Figure 8 it can be seen that the inspection effort increases nonuniformly but monotonically with k, and that the overall detection probability saturates at about 40 % at $k = 8$. Thus approximately 100 man days of effort could be saved by applying the RFUV model, without sacrificing overall detection capability.

Implementation of the RFUV model would again entail no modification of facility attachments: the inspectors would visit a given facility after the operator's physical inventory taking on an unannounced basis. Unverified inventory listings would be accepted at face value. Current IAEA-internal performance criteria for PIV activities would have to be modified substantially.

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EVOLUTION DES TECHNIQUES DE VERIFICATION DANS LES USINES DE FABRICATION DE COMBUSTIBLE MOX

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Abstract

In the MOX fabrication plants the increase in throughput and the consequent changes in the conditioning of the nuclear material and in the fabrication process applied is requiring a constant tailoring of the verification methods and techniques to the operational constraints.

This paper describes the evolution of the verification scheme as a response to the changed conditions and the experience gained on the technical means used (NDA, DA, C/S). it describes as well the foreseeable evolution and the means which will be necessary to develop or to adapt.

La progressive introduction de combustibles de type MOX dans le chargement des réacteurs de puissance à eau légère et le nombre réduit d'installations ayant, à ce jour, développé la technologie nécessaire à leur fabrication, s'est traduite par une rapide augmentation des deux grandeurs conditionnant le système de vérification, leur stock physique et le flux de matières.

Cette évolution a été également qualitative dans ce sens qu'elle s'est traduite par la dispersion des matières nucléaires dans l'installation et par la diversification de leurs types et conditionnement. La figure n° 1 illustre schématiquement la structure de flux et distribution de matières d'une telle installation.

L'orientation générale des stratégies appliquées a ainsi évolué depuis la simple recherche d'une conclusion quantitative dans un délai couvrant le temps de détection ("Timeliness Verification") et visant essentiellement la détection d'une éventuelle "abrupt diversion" jusqu'à un régime d'inspection continue fondé sur le suivi des mouvements externes et internes à l'installation, leur vérification systématique et le recours au confinement/surveillance et ainsi diminuer l'effort d'inspecton au moment de l'action visant la "timeliness detection".

La raison de ce changement d'orientation réside dans le fait aue la recherche d'une conclusion quantitative à chaque période correspondant au temps de détection exigerait un effort d'inspection considérable en cas de vérification ponctuelle mais aussi, et surtout, imposerait des contraintes opérationnelles à l'exploitant pratiquement inacceptables en raison de la nécessité d'arrêter la production pendant des périodes prolongées et qui se traduiraient par un accroissement sensible des coûts de production.

Pour faire face à ces changements des équipements et instruments de mesure ont dû être ameliorés ou parfois mis au point. Ils peuvent être considérés actuellement comme pleinement opérationnels. Les problèmes techniques résolus, le système doit être optimisé au moyen d'une gestion exhaustive des informations recueillies durant les activités d'inspection et l'extension du confinement/surveillance à la plus grande fraction envisageable de l'inventaire dans l'optique d'une projection des conclusions au niveau de la période bilan matières.

L'expérience montre que la projection des résultats partiels exige en préalable une stratification aussi pousse que possible. Celle-ci doit être basée, non seulement sur les informations fournies par l'exploitant en application du Règlement 3227/76 D'Euratom, mais aussi sur une foule d'informations techniques ou opérationnelles recueillies sur le terrain.

La gestion d'une telle masse d'information impose, "de facto", le recours à l'outil informatique pour déplacer l'action des inspecteurs vers l'évaluation et la prise de décisions motivées. Nous avons développé un logiciel qui intègre l'ensemble des informations dérivées des actions de vérification pour en faire un outil de diagnostic, d'evaluation et d'assistance à la prise de décisions.

La figure n° 2 décrit les grandes fonctions de ce logiciel dans le contexte de la mise en application des schémas décisionnels propres à une action de contrôle continu et ceux visant à la détection d'un détournement abrupte dans le temps de détection. Ces schémas sont illustrés dans les figures 3a et 3b.

Les points les plus sensibles de ce logiciel résident dans la stratification et la gestion des discordances constatées au niveau d'un article, d'un stratum vu de la totalité de l'inventaire, ou respectivement dik, Dk et D (fig. 3).

Nous avons choisi une stratification "à priori" sur les mouvements vers l'installation ou à l'intérieur de celle-ci. La stratification est interactive dans ce sens qu'elle peut être revue dès qu'un nouvel élément d'information se présente aux inspecteurs, ce changement pouvant être repercuté au niveau de l'évaluation globale ou partielle.

Un des bénéfices escomptés par la mise en place d'un tel logiciel est de pouvoir discerner, à terme et par l'accumulation d'informations, les composantes aléatoires et systématiques des erreurs de mesure NDA et DA et de pouvoir engager des actions correctives.

Il deviendrait ainsi possible d'atteindre des conclusions quantitatives d'un niveau de précision comparable au σMUF, ce qui permet d'envisager une évaluation de type MUF-D sur le bilan de matières.

En tout cas, il sera possible d'effectuer une évaluation de type "D-statistic" au niveau de, successivement, les flux de matières dans l'installation, l'ensemble de l'inventaire et le bilan de matières selon la relation bien établie

DMB = DPE - DPB + DI - DO

DMB: Différénce estimée pour la période bilan matières
DPE: Différénce estimée pour l'inventaire final
DPB: " " " " initial
DI: " " " les réceptions de matières
DO: Différénce estimée pour les expéditions de matières

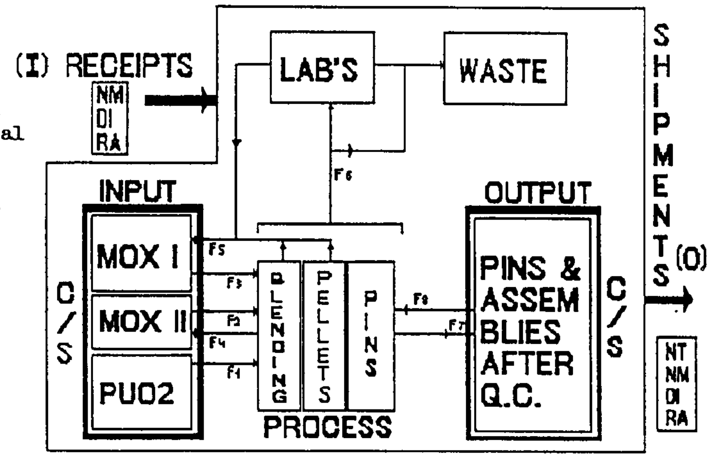


FIG. 1 INVENTORY & FLOW STRUCTURE OF A TYPICAL MOX FABRICATION PLANT

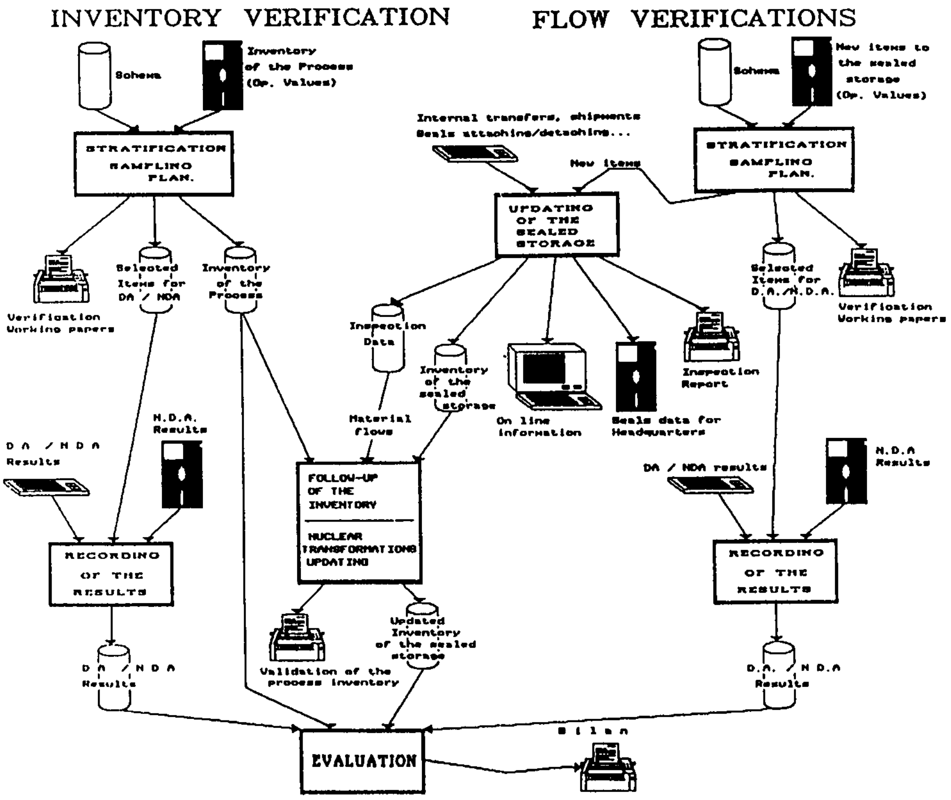
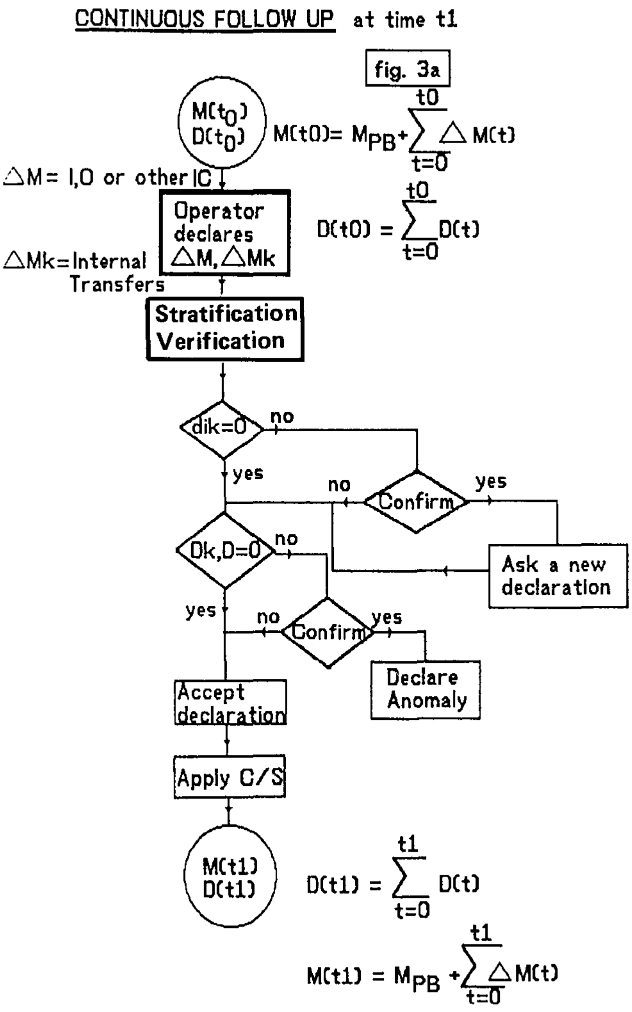
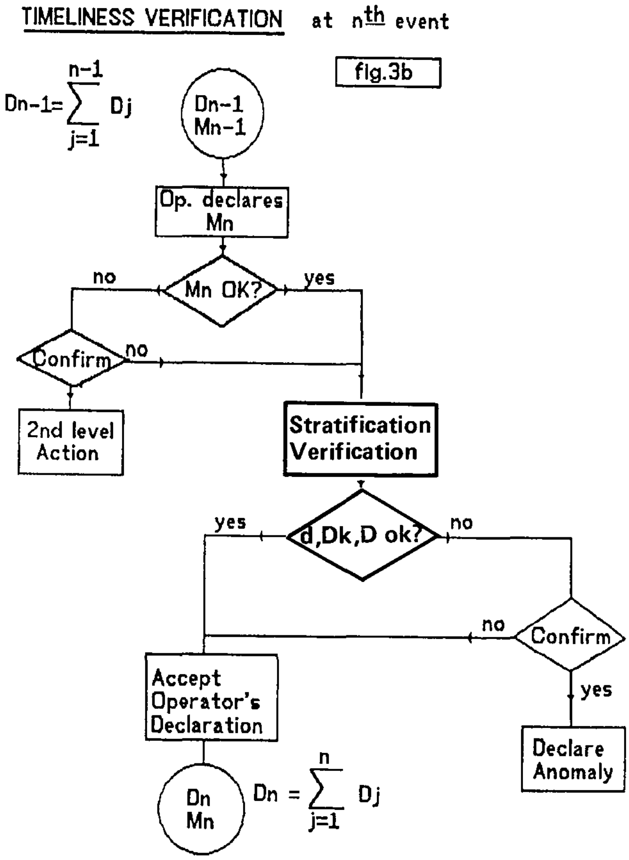


FIG. 2 GENERAL STRUCTURE OF A COMPUTER SOFTWARE FOR A FOLLOW-UP & CONTINUOUS EVALUATION OF A LARGE MOX FUEL FABRICATION PLANT.

Fig. 3



CONTINUOUS FOLLOW UP at time t1
fig. 3a
M(t0)
D(t0)
M(t0)= MPB+ Σ t=0 to t0 △ M(t)
△M= 1,0 or other IC
Operator declares △M, △Mk
△Mk=Internal Transfers
D(t0) = Σ t=0 to t0 D(t)
Stratification Verification
dik=0
no
yes
Confirm
no
yes
Dk,D=0
no
Ask a new declaration
yes
no
Confirm
yes
Accept declaration
Declare Anomaly
Apply C/S
M(t1)
D(t1)
D(t1) = Σ t=0 to t1 D(t)
M(t1) = MPB + Σ t=0 to t1 △M(t)



TIMELINESS VERIFICATION at nth event
fig.3b
Dn-1= Σ j=1 to n-1 Dj
Dn-1
Mn-1
Op. declares Mn
no
Mn OK?
yes
Confirm
no
2nd level Action
Stratification Verification
yes
d,Dk,D ok?
no
no
Confirm
yes
Accept Operator's Declaration
Declare Anomaly
Dn
Mn
Dn = Σ j=1 to n Dj

DESIGN CONCEPT FOR ANALYTICAL FACILITIES (ON-SITE LABORATORY) FOR THE EURATOM SAFEGUARDS DIRECTORATE AT BNFL SELLAFIELD WORKS

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ABSTRACT

The independent analysis of samples of special fissile material is an important part of the overall effort of the Euratom Safeguards Directorate to assure itself that the materials are not diverted from their intended uses as declared by the users and that any particular safeguarding obligations are complied with. The majority of the safeguards verification samples are transported to the European Commission's Safeguards Analytical Measurement (ECSAM) laboratories. In view of the large number of samples to be taken at the new thermal oxide reprocessing plant (THORP) in Sellafield, the Euratom Safeguards Directorate has initiated a design study for on-site analytical facilities. This paper describes the design concept resulting from this study.

1. Introduction

The new large scale industrial reprocessing plants recently started up or approaching completion present a new type of challenge to the Euratom Safeguards Directorate (DCS). The independent analysis of samples of special fissile material is an important part of the overall effort of the Euratom Safeguards Directorate to assure itself that special fissile materials are not diverted from their intended uses as declared by the users and that any particular safeguarding obligations are complied with in these plants. Up to now almost all safeguards verification samples taken at reprocessing plants are sent off-site for chemical analysis at one of the European Commission's Safeguards Analytical Measurement (ECSAM) laboratories/1/. In view of the large number of samples to be taken at the new reprocessing plants the Euratom Safeguards Directorate has initiated a design study for on-site analytical facilities. Although this study was primarily aimed at investigating the feasibility of on-site facilities at BNFL Sellafield Works it is intended to apply the concept to other large reprocessing plants under Euratom Safeguards.

2. User (Safeguards) Requirements

The Euratom approach to reprocessing plant safeguards includes sampling each input and Pu output batch for safeguards verification analysis. Additionally samples have to be taken from process tanks to verify in-process inventory, from certain waste streams, from Uranyl-nitrate tanks and from drums containing Uranium oxide powder. The periodic wash-out inventory presents a supplementary demand for sampling and verification analysis. Taking into account that both sites under consideration for the implementation of Safeguards on-site laboratories are multiple-facility sites, the annual number of samples to be analysed in an on-site laboratory (OSL) was estimated to be of the order of 1000. The different types of samples expected are shown in Figure 1. In view of the large throughput of nuclear materials in these facilities the analytical accuracy should be the best currently available for routine analysis for at least a subset of verification analyses. The laboratory must therefore routinely achieve ESARDA target values /2/ or better. Attention must be drawn, however, to the fact that the safeguards objective is the verification of the total quantity of nuclear materials in tanks or drums. This is determined by combining the results of volume or weight measurements of the material in the vessel or container with the nuclear materials concentrations obtained from chemical analysis or non-destructive analysis of samples drawn from these vessels or containers. The overall error variance of the determination of the quantity of nuclear materials is therefore given by the sum of the error variances of the chemical or non-destructive analysis and the volume or mass determination. Hence it would seem appropriate to aim for analytical accuracies equivalent to those achievable for volume or mass determination. It should also be remembered that only systematic errors usually dominate campaign evaluation.

In view of the expected operating life of the laboratories it must be ensured that full advantage can be taken of future improvements in techniques and procedures for the determination of volumes, masses and concentration of nuclear materials. Table 1 summarises the short term and medium term accuracies required for the principal types of samples.

Analytical facilities must be available to the inspectors for 45 weeks of the year with back-up and fast repair/maintenance for the case of instrument failure.

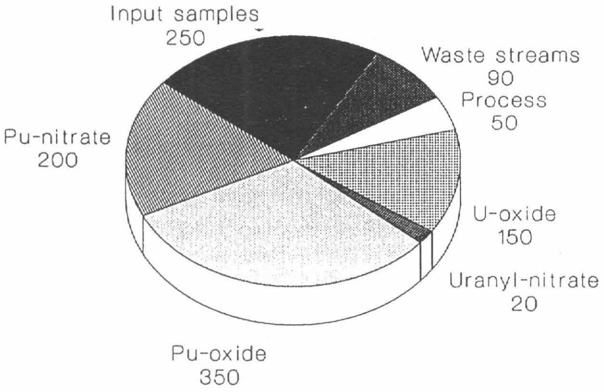


Figure 1: Expected annual sample throughput for on-site laboratories
Values estimated for multiple facility site

3. Boundary Conditions

If possible, the on-site laboratory should be integrated into existing or new operator laboratories and run under the main site licence to avoid problems with national licencing authorities and to make full use of supporting services. A green field on-site laboratory outside the perimeter fence of the facility to be safeguarded would be prohibitively expensive, would pose some of the transport problems encountered for transfers to ECSAM laboratories and would have to go through the complete approval procedures for a new nuclear facility. The space requirements must be reduced to a minimum because laboratory space is usually at a premium. The staff requirements must also be kept to the minimum figure still compatible with the throughput envisaged and with the safety regulations applicable in the facility. All analytical procedures and working practices must comply with the safety, operating and security rules which apply for the operator's laboratories. Similarly, the laboratory concept, final design, commissioning schedules and operating procedures have to pass the same formal approval procedures as those required for the operator's laboratories.

Staff for on-site laboratories will have to be trained and certified to the same standards as the operator's analytical staff. They will be subject to the operator's radiation dose monitoring service and will comply with all local rules regarding safety, security and administrative arrangements. Interfaces and services will have to be provided by the operator. Ports for sample transport (bagged and bagless) must be compatible with the systems employed by the facility. Whenever possible the analytical methods employed and the instrumentation implemented in the on-site laboratories should be the same as those used at the European Commission's Joint Research Centre Institute for Transuranium Elements since on-site laboratory staff are expected to be based there.

4. Analytical Options

Input samples: The most accurate analytical technique currently available for the determination of Plutonium and Uranium concentrations in samples of LWR fuel reprocessing input liquor is isotope dilution mass spectroscopy (IDMS). Its overall accuracy (including random, systematic and sampling errors) is believed to be better than 0.7% and some laboratories claim < 0.5% . There is scope for short to medium term improvement through the use of internal ratio standards and universally agreed spikes. A recent exercise at one reprocessing facility involving a complicated analysis tree with duplication and branching after almost every step of sample preparation and analytical procedures demonstrated that IDMS inter-laboratory differences in the order of 0.1% - 0.2% can be achieved for Uranium and Plutonium concentrations excluding sampling errors. The method is however complicated and time consuming.

The determination of Uranium and Plutonium concentrations by K-edge absorption densitometry and XRF measurements in samples of input liquor is an attractive alternative for routine analysis/3/. The accuracy for U-concentration is better than that routinely achieved with IDMS and the performance observed for Pu-concentration equals that found for the average of paired comparisons of inspector/operator results for this type of sample when both laboratories use IDMS. There is scope for some improvement in the accuracy of Pu-concentration through the use of fast pulse processing equipment and longer measurement times. The method has the enormous advantage of requiring no sample

		Method 1		Method 2		
		short term	medium t.	short term	medium t.	long term
Input	Pu-conc	0.6	<0.5	0.7	0.3	<0.1
	U-conc	0.3	0.1	0.7	0.3	<0.1
Pu-nitrate	Pu-conc	0.3	0.1	0.3	0.1	<0.1
Pu-oxide	Pu-conc	0.6	0.5	0.3	0.1	<0.1
UN-nitrate	U-conc	0.3	0.2	0.3	0.1	<0.1
U-oxide	U-conc	1.0	0.5	0.3	0.1	<0.1

Table 1: Target performance values for the on-site laboratory in percent. Method 1 is the routine (NDA) method, Method 2 the (potentially) more accurate DA method

preparation whatsoever and of being a simple, non-destructive method. Measurement results can be available as early as one to two hours after sampling.

After completion and confirmation of measurement results the sample liquor could be recycled, thereby reducing analytical waste.

The solution chosen for the OSL is the measurement of input samples by hybrid K-edge to the extent practically possible (undiluted samples must be available at the K-edge location).
A subset of approximately 20 percent will be analysed by IDMS.

Pu-nitrate samples: The analytical techniques used for this type of sample in the ECSAM laboratories is potentiometric titration and K-edge absorption densitometry. The accuracy currently achieved with titration in routine use is approximately 0.3% including the sampling error. There is scope for improvement.

The same performance is obtained with K-edge absorption densitometry. Again the K-edge has the advantage that it is a non-destructive instrument requiring no sample preparation and leaving no waste as the liquor could be recycled to the process.

Pu-isotopics in Pu-nitrate samples can be determined by mass spectrometry or by gamma spectroscopy. While mass spectroscopy offers higher accuracy for most Pu isotopes, gamma spectroscopy requires no sample preparation steps, its results are available one hour after sampling and the performance achieved with the Lawrence Livermore National Laboratory Multiple Group Analysis (MGA) code /4,5/ is better than 1% for all isotopes if the operator's values for Pu-242 are accepted.

In the OSL all Plutonium-nitrate samples will be by measured by K-edge absorption densitometry and high resolution gamma spectroscopy. Approximately 5% of the samples will be titrated and filaments prepared for mass spectroscopy. This figure will be increased if and when titration performance can be shown to reach the medium term target of 0.1%.

Pu-oxide powder samples: As for Pu-nitrate samples the method routinely used by the ECSAM laboratories is potentiometric titration. The accuracy expected from ESARDA target values for these samples is 0.3% including the sampling error. In most cases this is not quite achieved in practice because samples frequently take up water and oxygen while awaiting transport. The IAEA analytical services have introduced a procedure for the correction of these effects. Since this method has been adopted by DCS the performance values have been consistent with the ESARDA target values.

DCS have previously demonstrated that the Plutonium content in gramme size samples may be routinely determined to better than 1% accuracy with neutron coincidence counters/6/. A value of 0.5% can be achieved if detector design and calibration are optimised for small sample size. Additionally Pu isotopics can be determined to better than 1% using the MGA code. If Pu-242 is determined from isotopic correlations this may increase to 2-3% depending on burn-up. For first cycle reprocessed spent fuel however there are ways of improving the reliability of the correlation and thus for reducing the error on Pu-isotopics to approximately 1% /7/.

All Pu-oxide powder samples arriving in the OSL will be measured by combined neutron coincidence counting/high resolution gamma spectroscopy. A subset of the order of 20% will also be dissolved, titrated and filaments will be prepared fore mass spectroscopy. The proportion of samples analysed destructively in future will depend on the relative performance figures for both techniques.

Uranyl-nitrate samples: ECSAM laboratories normally use modified Davies and Gray potentiometric titration or K-edge absorption densitometry for these samples. The performance expected for titration from ESARDA target values (0.3%) is routinely achieved. Again the K-edge instrument matches the performance of chemical analysis.

The enrichment in U-235 is determined by mass spectroscopy (Thermoquad or Sector field) to 0.3-0.5%.

U-235 content may also be measured by gamma spectroscopy to approximately 1% but the counting rates observed for these samples would normally require excessive counting times to achieve this. If samples can be placed in a near 4' geometry (well detector) 1% accuracy can be achieved in 1-2 hour measurements.

In the OSL the initial method will be the use of the non-destructive techniques described, with less than 10% of samples also being titrated and prepared for mass spectroscopy.

U-oxide powder samples: As for Uranyl-nitrate samples Uranium concentration and U-235 content are determined by potentiometric titration (modified Davies and Gray) and mass spectroscopy. For samples transported off-site there is a problem with sample stability similar to the one discussed for Plutonium oxide powders. Again there exist procedures to correct for these weight changes but such changes are not expected to create problems at on-site laboratories.

There is no established easy non-destructive method to determine Uranium concentration in these samples with reasonable accuracy. They can, however, be dissolved and measured as liquid samples in a k-edge absorption densitometer. The low Uranium concentration obtained in such liquid samples (~40g/l) limits the accuracy which can be achieved to approximately 1-2%. In spite of this limitation all such samples will be measured in this way, with approximately 20% of the samples also being titrated and about 10% prepared for mass spectroscopy.

5. Laboratory Concept

In view of the safeguards requirements, the boundary conditions and the analytical options discussed above the on-site laboratory design concept had to include the following key elements:

- maximum degree of automation through the use of robots
- where possible routine analysis of samples by non-destructive methods to limit resource requirements and to reduce analytical waste
- classical chemical analysis for a subset of samples only, to check for small biases.
- shipment off-site of a small part of the samples chosen for destructive analysis for comparison of results and quality control.
- very compact design

- duplication and physical segregation of analytical processes, to meet the target throughputs while ensuring that localised failures do not disable the entire operation.
- compliance with national licencing requirements and the operator's site regulations

The solution found by the design team from NNC Ltd (formerly National Nuclear Corporation) in close collaboration with DCS and TUI for the particular case of BNFL Sellafield is shown in Figures 2 and 3. Glove boxes, instruments and other items are being designed into two laboratories made available to DCS by British Nuclear Fuels plc. One laboratory (lab 2) will contain a small office area which will also house computer equipment for data evaluation, data storage, communications, quality control, nuclear materials accountancy and expert system advice.

Under routine conditions all diluted **input samples** will be analysed in laboratory 2. It is expected that approximately 250 diluted input samples per year will arrive in the laboratory through a pneumatic transfer connection from the operator's shielded cell facilities. They will be aliquoted, spiked and separated in the IDMS robot glove box /8/. The robot also deposits small droplets on the filaments for mass spectroscopy already placed on the turret and prepares planchets for alpha spectroscopy which are subsequently automatically placed in the measurement cavity. The robot is controlled by a special unit which receives its instruction from the central laboratory computer.

All analyses of product samples will normally be carried out in laboratory 1. Samples of **Plutonium oxide and nitrate** will arrive either at a dedicated receipt station directly from the sampling stations or will be manually transferred in bags or containers for bagless transport. All Plutonium nitrate samples (~ 200 p.a.) will be measured by K-edge absorption densitometer, a subset will be analysed for Pu-isotopics in the robot box for mass spectrometry. The samples will be lowered into a special cavity protruding from the bottom of the box for K-edge measurements.

Every PuO_2 powder sample will be weighed (the samples will be taken in pre-weighed vials) and then measured by neutron coincidence counting and high resolution gamma spectroscopy in a finger protruding from the bottom of the glove box into the body of the neutron detectors. A subset will be dissolved - probably microwave enhanced - and titrated. The design shown in Figure 2 still uses a suite of glove boxes for manual sample manipulation. It is very likely that this will be replaced by a single robot glove box /8/ currently being designed by TUI, Canberra and the Karlsruhe nuclear research centre (KfK). Filaments for mass spectroscopy will be prepared in the MS robot box.

The **Uranyl-nitrate** samples and the dissolved **Uranium oxide** samples will be measured in a combined new compact K-edge/ enrichment meter instrument /9/ developed at KfK which will be attached to a small glove box not shown in Figure 2.

The use of robots for filament preparation and dissolution/titration will enhance sample throughput above that achievable by manual operation alone, by enabling activities to proceed during silent hours. It will also reduce dose uptake of OSL staff. In addition, electronic logging of analysis results will reduce the scope for data transmission errors.

6. On-site vs off-site analysis

The front end design prepared by NNC demonstrated the feasibility of on-site analytical facilities for Euratom Safeguards. It also gave DCS reliable estimates for investments and running costs for the on-site laboratory. There are seven principal reasons why DCS is following a policy of implementation of on-site laboratories as the preferred solution for safeguarding large reprocessing plants:

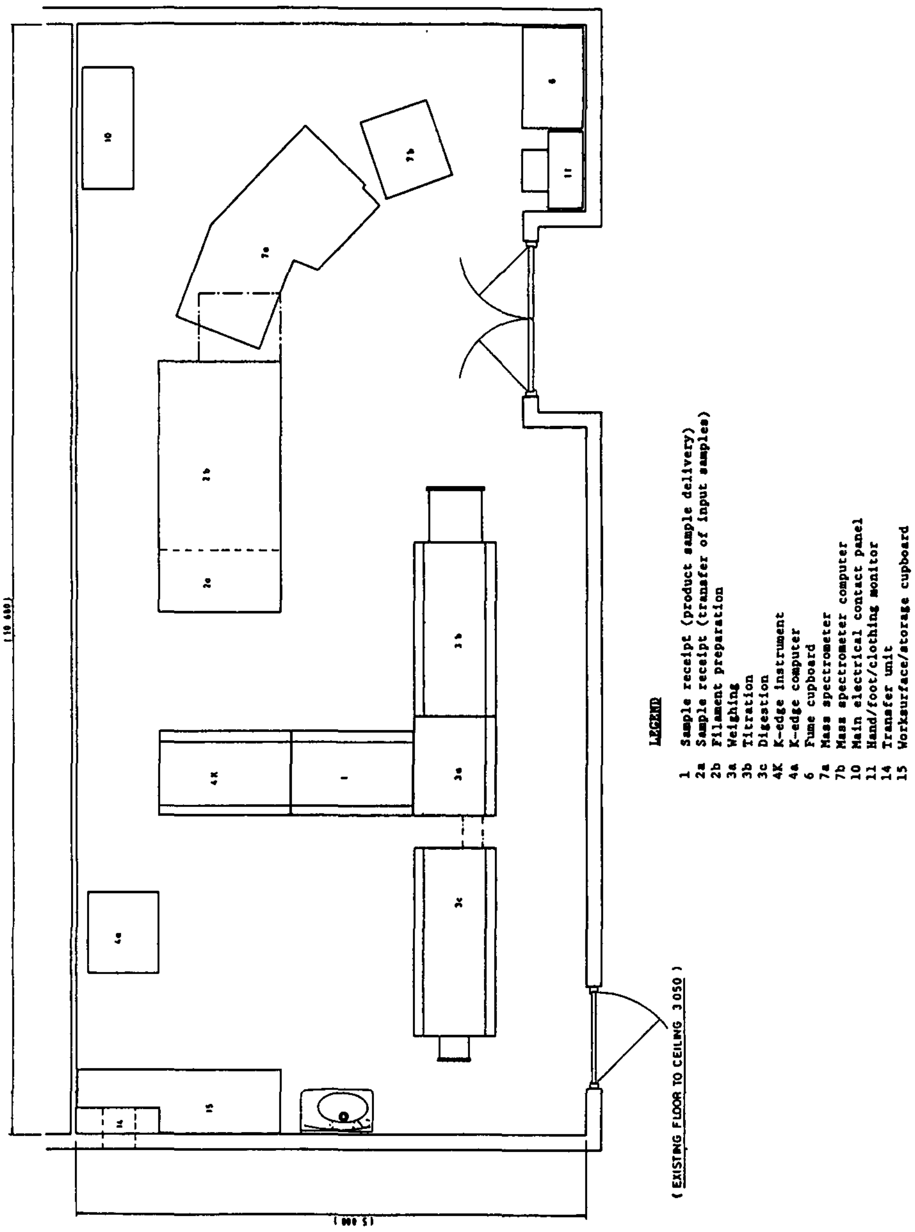
1. It is the only solution which guarantees that analytical results are available to the inspector in time for follow-up.
2. It is the only solution which guarantees timely verification of operator's declarations.
3. The analytical results will most likely be of higher accuracy than those obtained off-site because error sources related to transport preparation and transport delays are avoided.
4. It increases the inspector's confidence in the authenticity of some types of samples.
5. It significantly reduces operator and inspector human resource requirements and dose uptake.
6. It almost completely eliminates the need for transports of samples of nuclear material from the sites where on-site analytical facilities are implemented.
7. It is the cheaper solution.

These statements require some additional comments.

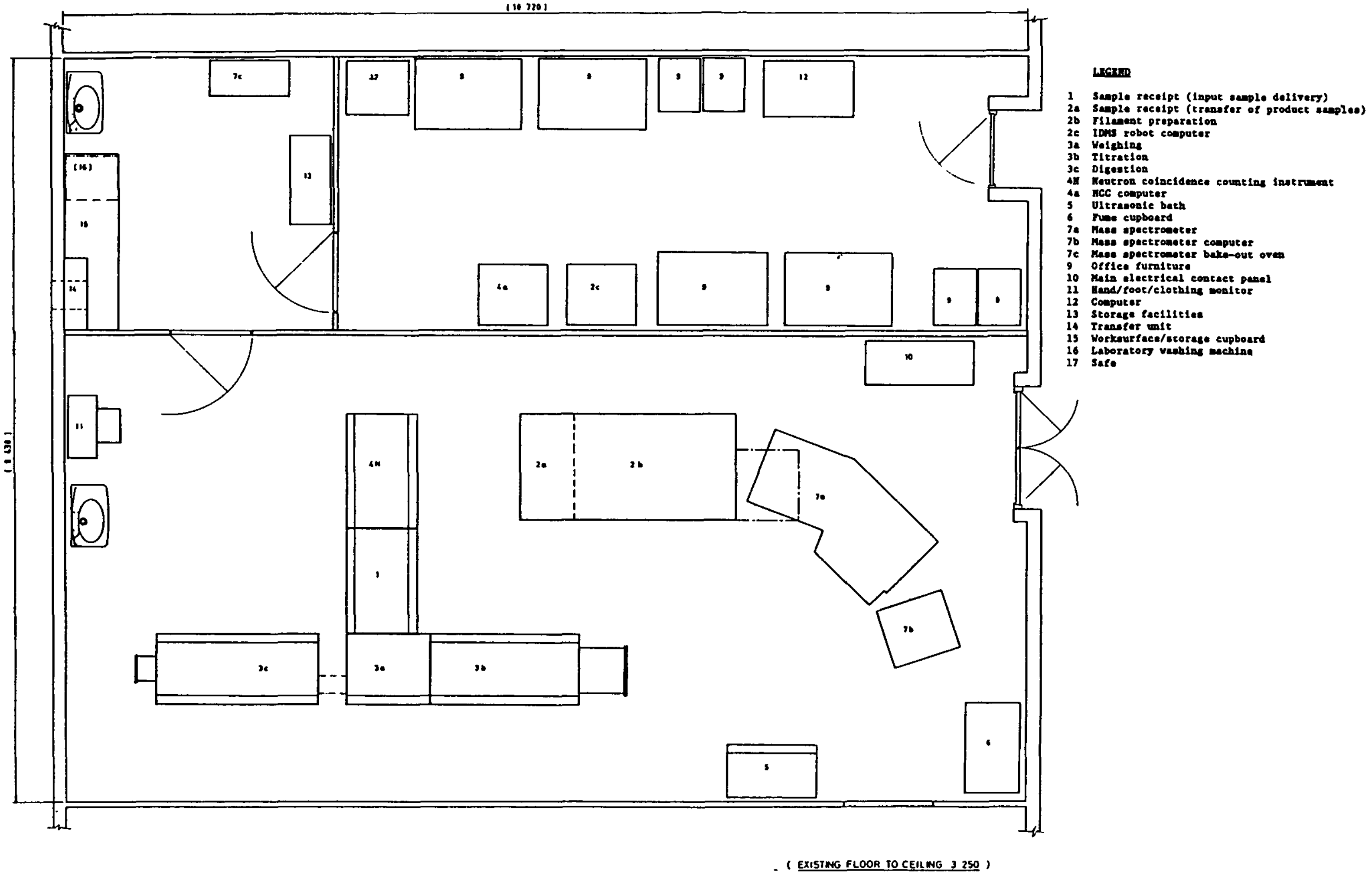
Ad 1&2) The efforts made by the IAEA and DCS to expedite transport of safeguards samples /10/ have been largely successful. Nevertheless recent developments in some Member states have shown that events beyond the control of the Safeguards Authorities can virtually block transport of safeguards samples for long periods. Strict interpretation and application of the terms of global transport licences is likely to lead to increasing delays.

Ad 3) It is believed that the expertise and experience available at ECSAM laboratories can not be matched by the small team on duty at the on-site laboratory. Therefore the basic analytical accuracy at on-site analytical facilities can at best be the same as that off-site as long as the same analytical procedures are employed. Transport of many samples, however, leads to additional error sources such as conditioning errors (input samples) and sample instability (powder samples). These errors can be eliminated at on-site laboratories. Also since any analytical problems will be detected while the material will most likely still be available for repeat analysis, it is unlikely that any such problems would influence most or all sample results for a reprocessing campaign.

Ad 4) Any sample conditioning steps (except for straight sample transfer from one vial to another) carried out by operator's staff decrease the confidence the inspector can have in the authenticity of the safeguards sample, i.e. his assurance that the sample has not been tampered with.



Laboratory 1 - product analysis Fig 2



Laboratory 2 - input sample analysis

Fig 3

Ad 5) The conditioning steps and any other activities by operator staff involving unsealed samples require direct or indirect surveillance (camera) by inspectors. This can take between 1 hour for product samples and several hours for input samples over and above the time the inspector has to be present to observe the sampling procedure itself.

Transport documentation has to be prepared by operator staff who also have to pack the samples in transport containers and to check the containers for compliance with national and international transport regulations and to measure surface dose.

Ad 6) The only transports still required are of unusual samples or of samples where some form of arbitration analysis or quality control analysis is desirable. Also spikes, reference materials and quality control samples will have to be transferred from other facilities to the on-site laboratory.

Ad 7) This is the most controversial point. It is best demonstrated using the following symmetry argument. The number of samples currently analysed by the ECSAM laboratories in one year is approximately 500, representing about 75% of the analytical capacity available. The additional analysis of 500 samples per year per reprocessing plant would require similar investments for laboratory extensions, refurbishing and new equipment as does the implementation of on-site laboratories. The main part of the running costs will also be the same since there is no reason why the cost of rent or rent-equivalent and services per square meter of laboratory space should be more or less expensive in industrial nuclear facilities than in nuclear research facilities. The only extra costs of the on-site solution are the increased staff requirements to make up for the time lost in travelling to and from the on-site laboratory (expected to be of the order of 50% i.e. 4 staff members per on-site laboratory, if it is assumed that the OSL will be operated by teams of four and therefore 8 persons would have to employed for corresponding off-site analysis at an ECSAM laboratory) and the costs incurred for travel expenses. This has to be compared to the costs for transporting samples (Currently of the order of 1000 Ecu per sample on average), purchasing and maintaining containers (estimated at 100 000 Ecu capital costs per lab and at least 20 000 Ecu per year running costs and depreciation), additional inspector hours required (of the order of 500 to 1000 Ecu per sample) and additional staff required at DCS and at the ECSAM laboratories to organise and/or control transports (Expected at least 2 posts i.e. ~130 000 Ecu per year). When these figures are added up for both options there is a clear cost advantage for the on-site laboratory ranging from 200 000 Ecu per year to 600 000 Ecu per year depending on the assumptions made for reductions in transport costs per sample and for the costs of additional inspector presence.

7. Conclusions

Efficient, effective and timely safeguards verification analysis of samples taken at large reprocessing plants can, in the opinion of DCS, best be achieved through the implementation of on-site facilities for destructive and non-destructive safeguards verification analysis. A concept for such facilities has been developed and negotiations are under way to resolve the questions of its feasibility and compliance with licences and site regulations. It is expected that the construction and. operation of such facilities will be cheaper than to send samples off-site to central safeguards laboratories. For these reasons the Euratom Safeguards Directorate intends to install on-site laboratories at the sites of the new large reprocessing plants in the European Communities.

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THE EUROPEAN COMMISSION'S SAFEGUARDS ANALYTICAL MEASUREMENT (ECSAM) LABORATORY NETWORK

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Abstract

The ECSAM activities performed in collaboration between services of the Commission of the European Communities at DG XVII-DCS and at DG XII-JRC are described. Essential part is the analytical verification of samples from the nuclear fuel cycle, taken by Safeguards inspectors and measured at ECSAM laboratories. Details about the type of samples, their origin and flow, about transport of samples and its impact on timeliness, and the measurement methods and efforts are given. Also the central data evaluation is described. The activities are supplemented by a quality control programme, by in-field measurements and by specific development work. The ECSAM system works successfully since 20 years. Future trends (on-site laboratories, more in-field measurements, tighter analytical target values, etc.) are indicated.

1. Introduction

The Euratom Safeguards Directorate (DCS) of the Commission of the European Communities has the special task and duty to verify that fissile materials are not diverted from their intended uses as declared by the users and that any safeguarding obligations are complied with. To this end the Commission deploys a corps of safeguards inspectors who carry out inspections in nuclear installations within the Community. An important element of their verification activities is the taking of samples of materials at the nuclear facilities, the analysis of these samples and the evaluation of the results. In the framework of the European Commission's Safeguards Analytical Measurement (ECSAM) laboratory network, the majority of these samples are sent for chemical analysis to laboratories at three institutes of the Commission's Joint Research Centre (JRC), namely

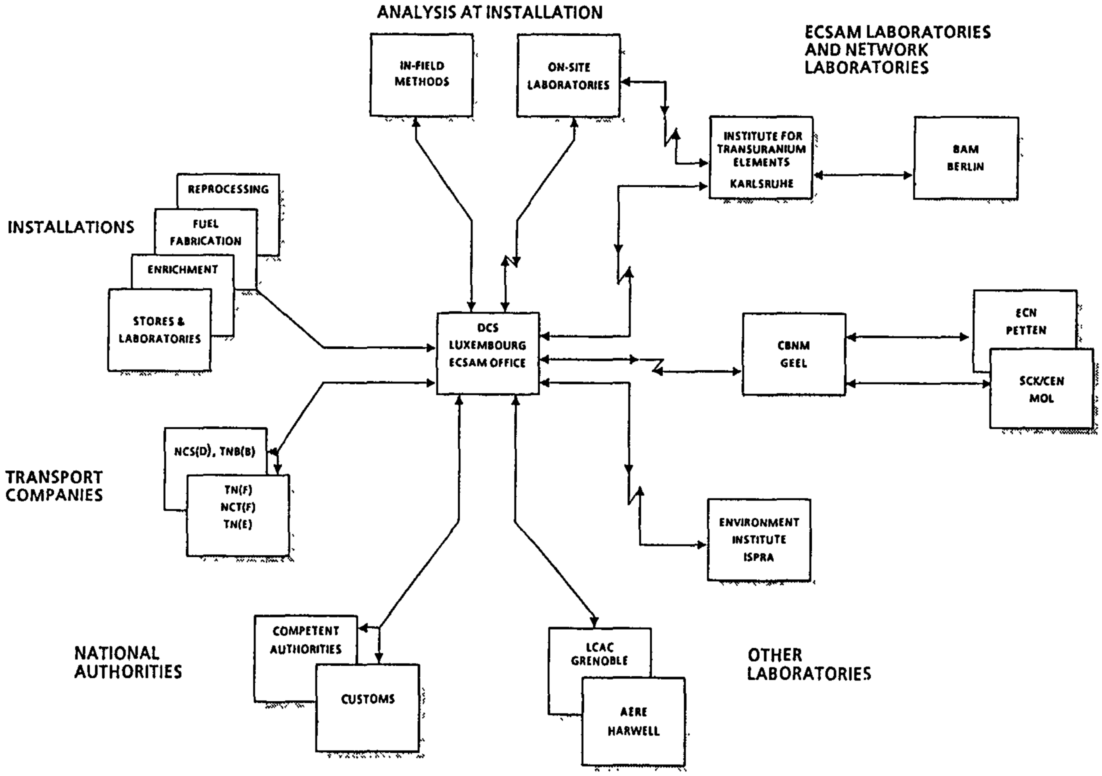


Fig. 1 - DCS VERIFICATION ANALYSIS SYSTEM

the Central Bureau for Nuclear Measurements (CBNM), Geel, the Institute for Transuranium Elements (ITU), Karlsruhe, and the Environment Institute (EI), Ispra.

After twenty years of successful activities, it appears worthwile to present ECSAM in more detail.

2. The ECSAM Scheme

A schematic view of the relations in the DCS verification analysis system is given in Fig. 1, indicating also the interactions of the ECSAM laboratories CBNM, ITU and EI.

Special ECSAM procedures have been elaborated and sanctioned subsequently by inter-DG agreements. They serve as guideline for the execution of the daily work (verification measurements, special developments and assistance), as well as to ensure that the necessary resources are available for the support given from DG XII-JRC to DG XVII-DCS. The activity details are laid down in Work Sheets which are annually reviewed and mutually agreed.

In 1991, 18 JRC staff members were involved in ECSAM activities, 14 in Karlsruhe, 3 in Geel and 1 in Ispra.

The three JRC institutes - being services of the Commission of the European Communities - have been chosen as they guarantee independence of the measurement results with regard to the analyses. The institutes may commission measurements to "satellite-laboratories" in a coded way, however they remain responsible for all results thus obtained.

The allocation of samples to the various laboratories has mainly been done on geographical grounds (minimizing transport distances and formalities). In cases, umpire settlements had to be carried out exceptions from this general trend are evident. The task of organizing a Quality Control Programme was assigned to CBNM. This gave use to the more extensive measurement evaluation programme REIMEP (Regular European Interlaboratory Measurement Evaluation Programme) for which participation of ECSAM and satellite laboratories is compulsory.

The analytical programme has also been supplemented by some development work in order to overcome specific problems or to adapt methods to appearing or future requirements in Safeguards. These activities were mainly performed by ITU and by EI.

The ECSAM responsibles from the various services of the Commission meet regularly, in general twice a year, for mutual information, discussion of problems and of future trends and developments. The 30th ECSAM meeting was held on 20 March 1991 at Geel.

3. Type of samples, origin and flow

Table 1 gives an overview of the different types of DA (destructive analysis) samples taken at facilities in the fuel cycle whereas in Fig. 2 the distribution of samples between the various facilities is given for 1990.

	Reprocessing	MOX Fabrication	LEU Fabrication	HEU Fabrication	Enrichment	Conversion	Stores / Labs
• Irradiated U/Pu-nitrate solutions	X						
• Pu-nitrate solutions	X	X					(X)
• U-nitrate solutions	X	(X)	X	X		(X)	(X)
• Pu-oxide powder	X	X					(X)
• U-oxide powder	X	(X)	X	X		X	(X)
• U-oxide pellets		(X)	X				
• MOX powder		X					
• MOX pellets		X					(X)
• U metals and alloys				X			
• UF_6			(X)		X	(X)	
• UF_4			X			(X)	
• HAW concentr.	(X)						
• Oxalate mother liquor	X						
• ADU		(X)	(X)				
• Other samples including Th, carbides, nitrides, etc.							(X)

Table 1 - TYPE OF SAMPLES FROM VARIOUS FACILITIES
(X) indicates low to very low sampling activities

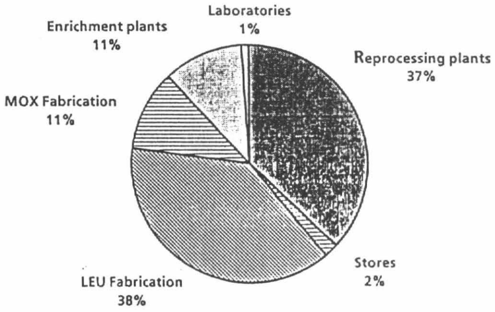


Fig. 2 - SAMPLING ACTIVITIES IN 1990
BY FACILITY TYPE

The corresponding repartition for the sample material type is shown in Fig. 3, the sample flow diagramme in Fig. 4. The number of samples taken from different facilities in the nuclear fuel cycle depends on the amounts of nuclear material present in the flow and inventory of the plant and on the form in which the material is processed or stored. The majority of samples originate from fuel fabrication facilities and reprocessing plants whereas the number of samples varies from year to year with the number and duration of reprocessing campaigns.

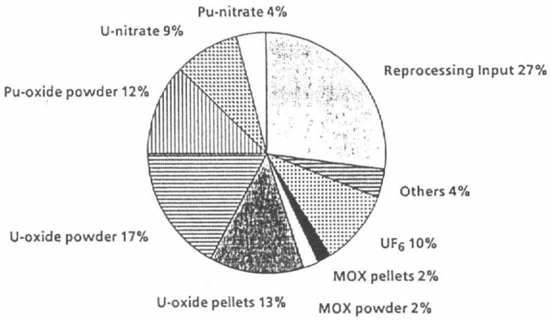


Fig. 3 - DESTRUCTIVE ANALYSIS SAMPLES 1990

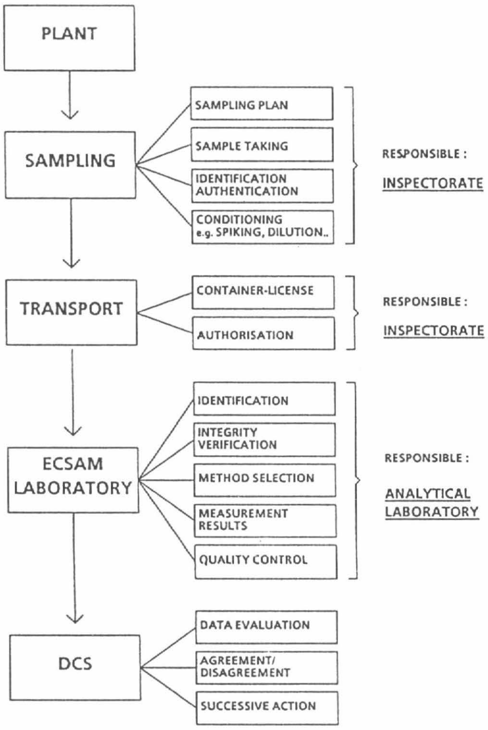


Fig. 4 - FLOW DIAGRAM OF INSPECTION SAMPLES

4. Sample taking at site of inspection

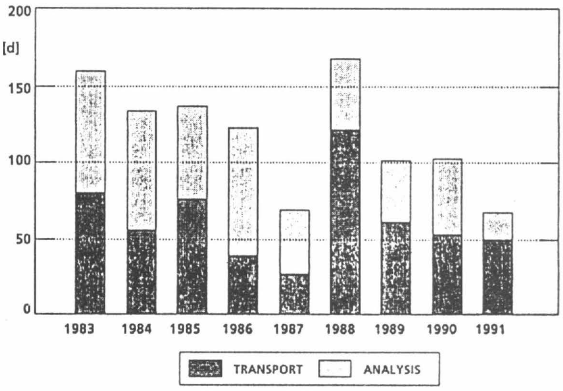
During the preparation for an inspection at Headquarters in Luxembourg or on site, the inspectors decide on the number and type of samples to be taken. The numbers are normally calculated on the basis of a stratified list of inventory items (for inventories), on the goal quantity, the detection probability, the expected accuracy of the analytical methods and the average nuclear material content per item and stratum. Samples for flow verification at reprocessing plants are usually taken after the nuclear material in the tanks has been homogeneised before the content is transfered across MBA (Material Balance Area) boundaries and as well as product powder batches are filled into cans at the hoppers. At MOX fabrication facilities they are usually taken from input material and when pellet batches pass the surface control stage after sintering and grinding. Samples are chosen at random from the population of items, tanks or silos, except for those sampling points where every transfer of nuclear material is sampled.

The samples are physically taken by operators' staff under inspector observation. Wherever possible the inspector asks for separate samples to be taken into specially marked and sometimes preweighed sample bottles supplied by the inspector. The samples are either placed under seal and/or under camera surveillance or remain under observation by the inspector until they can be sealed at a later point in time.

Samples of input liquor and some process liquors from reprocessing plants need to be diluted, aliquoted and spiked and then dried before being bagged out. Plutonium nitrate liquor of concentrations above $50 g.l^{-1}$ must be diluted. This sample conditioning process must either be directly observed by the inspector or by a suitably placed video camera connected to recording equipment. Together with information obtained during the verification of the basic technical characteristics the above ensures the authenticity of the samples, i.e. the assurance that they originate from the correct item, tank or silo and that they have not been tampered with.

5. Sample transport

The majority of the Euratom safeguards verification samples are transported to the ECSAM laboratories. Delays in transport can be detrimental to the usefulness of destructive analysis with regard to timeliness and quality, as changes may occur in the sample adversely affecting the quality of the analytical result, and since late availability of the results to the inspector may make follow-up difficult or impossible in case of differences. Figure 5 shows delays in transport and time between sample arrival in the laboratories and the availability of the results in Luxembourg.



Values for 1991 up to 11.03.1991

Fig. 5 - TIME FOR ANALYSIS AND TRANSPORT 1983 - 91

The reasons for the delays in transport have been examined in detail /1/.

They are :

- problems with import/export licenses
- shipment approvals
- certificates in lieu of end user certificates
- lack of approved or validated transport containers
- complicated interaction between several organisations.

In the same publication a new approach to transport was suggested which relies mainly on forward licensing and global export/import and transport licenses. The implementation of this concept led to marked improvement until legal problems of one carrier in a particular Member State in 1988 virtually paralyzed the transport of safeguards samples. Since then matters have again been improving slowly but the delays encountered especially for samples of Plutonium oxide, nitrate, MOX and to a lesser degree, reprocessing input solution are still too high.

In view of these problems and taking into account the high costs for transport of samples of nuclear material, the Euratom Safeguards Directorate decided to introduce mobile equipment for in-field analytical measurements of UF_6 and UO_2 samples and to install on-site laboratories at the sites of the large new reprocessing plants. For the other facilities further concerted efforts by all parties concerned are necessary to expedite the transport of safeguards samples.

6. Measurement methods, equipment and efforts

All three ECSAM laboratories have 25 to 30 years experience in the handling of radioactive material and the destructive and non-destructive determination of isotope abundances and element concentrations of samples from the nuclear fuel cycle. Analytical details are given below. Whether U and Pu element concentrations are determined by direct assay by e.g. titration, or by Isotope Dilution Mass Spectrometry (IDMS), is decided by DCS and indicated in the formal request for analysis.

CBNM, Geel

A main part of CBNM's specific research programme concerns the preparation and certification of nuclear reference materials and spikes at a European level. Already due to this task a number of methods for very accurate U and Pu analyses exist in this laboratory. From the available mass spectrometers four are used for work related to ECSAM : Finnigan-MAT 260 (for U), 261 (for Pu), 262 (for U-minor isotopes) and 511 (for UF_6).

For isotope analysis, U and Pu purification is done on all samples, including the related reference materials, by ion-exchange procedures prior to analysis. U/Pu separation is done twice to eliminate a possible influence of ^{238}U on the ^{238}Pu measurements.

The chemical preparation of samples is considered to be integral part of the measurement process and hence contributes to the total uncertainty of the end result as all other uncertainty components.

On this basis, ≤ 0.05% uncertainty at the 2s or equivalent level is delivered routinely on ^{235}U in UF_6, ≤ 0.3% on ^{235}U in Low Enriched Uranium (LEU) fuel, and ≤ 0.2% on Pu isotope ratios.

^{238}Pu abundances are exclusively determined by mass spectrometry, ^{241}Am by γ-spectrometry.

For uranium elemental assay two independent methods are available : controlled potential coulometry and automated potentiometric titration according to the well established Davies and Gray method, yielding in uncertainties of ≤ 0.05% at the 2s or equivalent level.

Plutonium is determined by controlled potential coulometry only. The overall uncertainties lie in the range of 0.08%.

ITU, Karlsruhe

Specialities of this laboratory are the determinations of U and Pu concentrations in reprocessing plant input solutions by IDMS, supplemented by α- and γ-spectrometry, and the application of K-edge densitometry for the elemental assay of U and Pu.

For the isotope ratio measurements thermoionic mass spectrometers type Finnigan MAT 261 and 262 are available, using multicollectors and double rhenium filaments. The technique used is that of total evaporation which is well suited to Pu samples of a few nanograms and to U samples of around 30 nanograms.

For α-spectrometry multichamber counters fitted with surface barrier silicon diodes are used. The samples are prepared by a modified single drop procedure. A peak resolution of better than 18 keV at 5 MeV is obtained.

^{241}Am is determined by a γ-spectrometer with germanium detectors.

The K-edge densitometer, designed and built at the KfK, Karlsruhe , consists of a X-ray generating tube, operating at 150 kV/15mA with a tungsten target, a collimating tube of lead and tungsten and a planar Ge detector (200 mm^2 x 10 mm) /2/. The sample solutions are placed in optical absorptiometry vials for the measurement.

Titrations of U and Pu are carried out by means of two Radiometer Titralab systems, each consisting of a video titrator, autoburettes and a sample changer.

Solution densities are determined by measuring the natural frequency of a vibrating reed in a Paar DMA 10 density meter. With this instrument densities up to $4 g \cdot cm^{-3}$ can be determined with an accuracy of $2.5 . 10^{-4} g \cdot cm^{-3}$.

All analytical results are transmitted to central computers e.g. PDP 11/73, for data evaluation, archiving and quality control.

EI, Ispra

The Environment Institute is taking care of uranium analyses only. For ECSAM activities one mass spectrometer Finnigan-MAT 261 with a variable multicollector system is available, and two potentiometric systems (Metrohm 682 Titroprocessor) are used for the elemental assay of uranium by the Davies and Gray method. The analytical laboratory is equipped for the adequate treatment of the samples. The uncertainty routinely achieved is ≤ 0.1% in the analysis of pure products.

A considerable effort was made over the past 20 years by the ECSAM laboratories to satisfy the DCS requirements for verification analyses, as can be seen on Fig. 6 and Fig. 7.

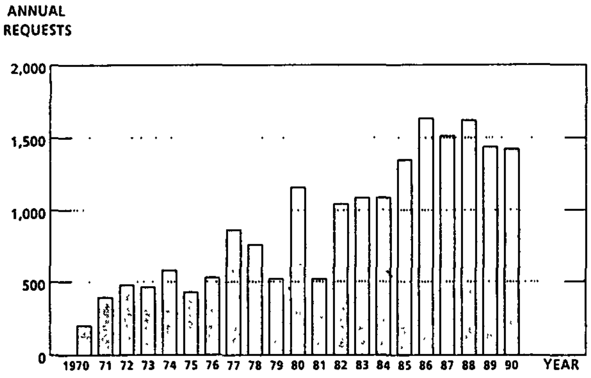
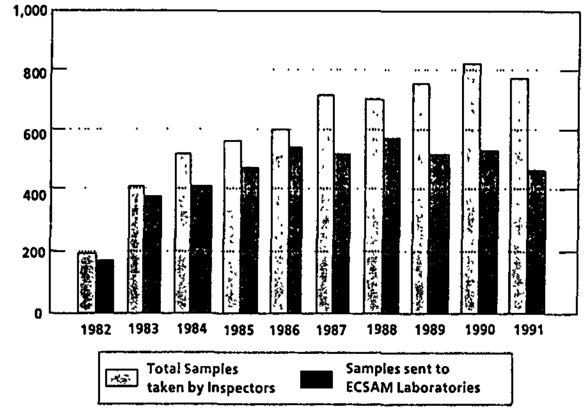


Fig. 6 - ANALYSES REQUESTED FROM ECSAM LABORATORIES



Values for 1991 estimated

Fig. 7 - NUMBER OF DA SAMPLES 1982 - 1991

7. Quality Control

Quality of all analytical measurements is ensured by the application of adequate reference materials and internal laboratory control procedures. For isotope ratio measurements at CBNM, all mass spectrometers are calibrated by synthetic isotope mixtures, prepared and certified to 0.01% or, at the other institutes, by certified reference materials of other sources.

For the elemental assay by coulometry or titration the corresponding EC certified nuclear reference materials (e.g. EC-NRM 101) or others originating from NBS/NBL are applied for calibration.

In the frame of the CBNM guided REIMEP (Regular European Interlaboratory Measurement Evaluation Programme) the participation of the ECSAM and satellite laboratories is compulsory /3/. The results are evaluated and discussed during the regular meetings of the ECSAM Committee. Some of the results are presented at this Symposium.

So called "Target Values" for uncertainties in Safeguards related analytical measurements have been conceived by CBNM, discussed in the ESARDA Working Group on Standards and Techniques for Destructive Analysis, and published /4/. They are used as criteria for minimum performance : measurements must aim to be at least as good as the Target Values indicate, but preferably better. In reality the values are reached in most cases, if not passed, but in view of the future Safeguards development more tighter values appear to be necessary.

8. The central data evaluation

After the reception of the analytical results by DCS, they are entered into a database and they are analysed, individually and in groups chosen by material type, installation, laboratory and by evaluation period. At present the modules of these procedures exist as stand-alone routines only. Work is underway to integrate these into one application. Evaluation reports are produced which flag samples for which the relative differences are larger than expected from the ESARDA Target Values and/or other thresholds. The standard deviation for operator-inspector differences is calculated for all variables and again compared to performance values (previous evaluation periods) and to values expected from the ESARDA Target Values. In case of differences or estimators larger than predefined thresholds, a follow-up procedure is initiated which may involve the analysis of reference samples and detailed discussions with the facility operator. Table 2 presents an overview of evaluation results covering approximately 1500 samples taken during the period 1986-1990. Attention is drawn to the fact that ranges are given rather than average values for the standard deviation of operator-inspector differences because it was found that these values may vary significantly with the facility-laboratory pairs.

For most types of material and analytical methods the ESARDA Target Values are achieved. Where this is not the case, there is strong evidence that the problems are partly not due to insufficient analytical accuracy but to problems with sample conditioning (e.g. filtering), sample stability (uptake of oxygen or water by powders) or insufficient material characterisation. There has been a marked improvement in the laboratory performance for IDMS over the evaluation period discussed and since the use of the Target Values. The only areas where there might still be some analytical problems are the analysis of MOX samples and that of thorium samples.

Sample Type	Number of Facilities (3)	Analytical Methods	s(Pu)(4) [%]	s(U)(4) [%]
Reprocessing Input	6	IDMS Hybrid K-edge	0.2 - 1.5 ~ 0.7	0.1 - 1.5 ~ 0.3
Plutonium Nitrate	4	Potent. Titration K-edge	0.2 - 0.6 0.2 - 0.3	-
Uranium Nitrate	6	Potent. Titration K-edge XRF	-	~ 0.1 ~ 0.2 ~ 1
Plutonium Oxide Powder	4	Potent. Titration NDA (n/gamma)	0.2 - 1 (1) 0.5 - 1.5 (2)	-
MOX Powder	4	Potent. Titration NDA (n/gamma)	0.2 - 1 (1) 0.5 - 2.5 (2)	~ 0.2 ~ 0.5
MOX Pellets	4	Potent. Titration NDA (n/gamma)	~ 0.3 0.5 - 2 (2)	~ 0.3 ~ 5 (2)
Uranium Oxide Powder	8	Potent. Titration Lab In-field	-	0.1 - 1 (1) 0.1 - 0.2
Uranium Oxide Pellets	7	Potent. Titration Lab In-field	-	0.03 - 0.3 < 0.1
Uraniumhexafluoride	5	Mass Spectroscopy	-	^{235}U : 0.2 - 0.3

(1) Upper values believed to be caused by oxygen/water uptake
(2) Depends on counting time, burn-up, Pu-242 correlation
(3) Facilities where very few samples were taken are not included
(4) Outliers excluded

Table 2 - RANGES/AVERAGES FOR RELATIVE INSPECTOR-OPERATOR DIFFERENCES (STANDARD DEVIATIONS "s") FOR MAJOR SAMPLE TYPES (1986 - 1990)

9. In-field Measurements

To overcome the transport inconveniences described above, in-field mass spectrometric and potentiometric determinations of uranium in uranium dioxide pellets and powders and in uranyl nitrate product have been tested and performed since 1987 /5/. To date, 15 experiments corresponding to about 300 samples analysed have been carried out in four different plants. The equipment (titroprocessor and mass spectrometer), the accessories and the reagents were transported to the plants by road. The results obtained can be considered as very good. They have demonstrated the feasibility of the in-field potentiometric analysis, included the isotopic determinations, and the quality and stability of the equipment. Hence, in-field measurements represent an interesting alternative or supplement to the conventional analytical scheme, especially in view of timeliness requirements.

10. Development work

The verification measurements are supplemented by some analytical development activities.

K-edge densitometry has been adapted to determine the U concentration in spent fuel solutions, and U and Pu concentration in processing product solutions. When combined with X-ray fluorescence, Pu-concentration in input solutions can be determined as well. After intensive field-testing at ITU the instruments have been applied for routine analysis /2/ and a K-edge/X-ray fluorescence spectrometer is routinely used at the reprocessing plant at La Hague, where ECSAM is giving the technical support.

The presence of Np in Pu products so far was very difficult to detect. It would, however, falsify the Pu-titration methods in use. An ICP-MS at ITU was adapted for the analysis of Pu-product samples and offers now a mean to measure Np in Pu samples. The unexpectedly high Np-content observed in Pu products caused the development of new methods for Pu determination /6/.

With increasing fuel burn-up the amount of plutonium found in residues in the dissolution tanks of reprocessing plants increases. At ITU methods were developed to determine the fissile material content in such highly active materials by IDMS, ICP-MS solutions or by laser ablation of solids.

The preparation of the OSL (on-site laboratory) concept required development work for the automatization and robotization of some analytical methods. This has been done successfully at ITU.

The EI at Ispra concentrated on the development of the in-field measurements which were tested with success under real conditions.

11. Future Trends

At present the situation for verification measurements in the EC seems to evolve in the following way :

a) stabilizing in the LEU sector
b) increasing in reprocessing because of the new - and larger - installations.
c) increasing in the MOX sector because of the successful use of Pu for recycling in thermal reactors.

With three new large reprocessing facilities starting or having recently started operation the number of reprocessing input samples and of plutonium product samples in the form of plutonium oxide powder and plutonium nitrate liquor will increase drastically. Similarly the new or modified MOX fabrication plants will lead to a considerable increase in the number of MOX powder and pellet samples to be verified.

Especially the much higher throughput of nuclear material in the facilities represents a challenge and may lead to problems. Constant relative measurements uncertainties supposed, the uncertainty, expressed in absolute material amounts on the overall material balance verification, will become unacceptably large. Much smaller relative uncertainties on the measurements (and the sampling) will have to be achieved. Hence it is necessary to tighten Target Values for uncertainties and to make further analytical progress without increasing the burden on the analysts (of both operators and inspectors). Fortunately, there are good prospects that this can be achieved.

The total number of Safeguards verification measurements will probably increase but there is a development in different directions : on-site laboratories (OSL) and in-field measurements.

The reasons are obvious, both are in relation to the considerable time and efforts required for the transport on nuclear materials, and the first one also with the planned large throughput of fuel in the new commercial reprocessing plants.

The OSL concept has been initiated by DCS and is being developed by ITU in close contact with DCS. By installing highly automatic instrumentation with computer link to the ITU central laboratory at Karlsruhe, the staff number needed can be kept at the minimum of two per laboratory (required from safety standpoint as well), giving still access to the knowledge and experts at the central laboratories /7/. A simplified cost balance proves that OSL's are cost neutral compared with the conventional off-site concepts. Instead of transporting with higher cost (and longer delay) radioactive material, the laboratory staff travels to the plant site. The costs to install and maintain a laboratory on either site are about equal; the same is valid for the equipment. The OSL staff is planned to be exchanged in a revolving system each week. When not staying at the plant site they will work at ITU.

In-field measurements of uranium have been demonstrated successfully mainly in LEU plants. This is also valid for non-destructive assay (NDA) of ^{235}U-abundances in fuel. In the future it could be of interest to extend this concept of in-field measurements also to the analysis of plutonium in MOX in fuel fabrication plants. The equipment and the methods are very similar, but the plutonium handling has to be done in 2 - 3 glove boxes installed in a controlled area of the plant and permanently reserved for Safeguards purposes. From this point of view this concept has some similarities with the OSL one. Main difficulties may be not of technical nature but possibly related to the acceptance by the plant operator.

The outlook in the future can be summarized as follows :

a) Verification measurements in ECSAM laboratories will stabilize in number or slightly decrease.
b) The OSL concept will become reality but will provide a new type of challenge to DCS and the central laboratory ITU.
c) The number of in-field and NDA measurements should and can increase.
d) Target Values for uncertainties must be tightened and the accuracy of measurements must be increased.
e) In relation to d), new reference materials are requested and should be prepared in time, aiming at uncertainties of ≤ 0.05% on the certified values (≤ 0.02% for those required for reprocessing plants).

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PERLA: FOUR YEARS OF OPERATION - A PROGRESS REPORT

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Abstract

In the field of Safeguards (and in particular of NDA) for many years there has been a growing awareness that the emphasis in the development of NDA should have been put not only on the development of new techniques, but also on the assessment of the performances of existing techniques in different plant conditions, and in filling the gap between development of instruments and their field use. As a result, more and more emphasis was given to customer-oriented research, thus improving the aspect of **support and technology transfer.**

At the Joint Research Centre (JRC) this guideline has led to building a training and PERformance LAboratory (PERLA). PERLA is a complex of laboratories of the Institute for Safety Technology of the JRC, set up in the framework of its Safeguards and Fissile Materials Management Programme. The basic aim of PERLA, right from the beginning, was to improve technology transfer from the laboratory development to the application of safeguards instruments and techniques in an industrial environment. The laboratory is oriented specifically to non-destructive assay (NDA) techniques. Current activities of PERLA are:

- training of EURATOM and IAEA inspectors;
- design and execution of NDA performance evaluation exercises (neutron, gamma and calorimetry experiments);
- integrated NDA instrument development;
- basic research.

The scope of this paper is to give a summary report on the activities carried on in PRE-PERLA (one of the facilities of PERLA), to quantify them in terms of resources, to give some scientific/technical results achieved and to present the future planning.

1. Introduction

In the field of Safeguards since some years there has been a growing concern that the emphasis in the development of NDA should have been put not only on the development of new techniques, but also on the assessment of the performances of existing techniques in different plant conditions, and in filling the gap between development of instruments and their field use /1,2/. As a result, more and more emphasis was given by laboratories to customer-oriented research, thus improving the aspect of support and technology transfer.

At the Joint Research Centre (JRC) of the Commission of the European Communities, this guideline has led to building a training and PERformance LAboratory (PERLA) /3/. PERLA is a complex of laboratories of the Institute for Safety Technology of the JRC, set up in the framework of its Safeguards and Fissile Materials Management Programme. The basic aim of PERLA was to improve **technology transfer** from the laboratory development to the application of safeguards instruments and techniques in an industrial environment. The laboratory is oriented specifically to non-destructive assay (NDA) techniques. Current activities of PERLA are:

- training of EURATOM and IAEA inspectors;
- design and execution of NDA performance evaluation exercises (neutron, gamma and calorimetry experiments);
- integrated NDA instrument development;
- basic research.

The laboratory is composed of different facilities (Fig.1), i.e.:

- PRE-PERLA;
- the Non-Destructive Assay (NDA) laboratory;
- the spent fuel pool;
- the NDA field test facility.

Some of the facilities are used only specifically for Safeguards purposes (e.g. PRE-PERLA), and others (e.g. the pool) are shared with other programmes.

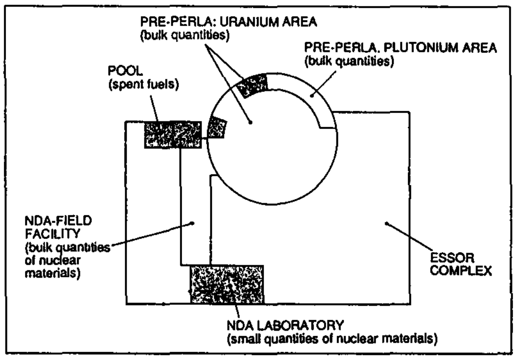

Fig.1: PERLA laboratory - general view.

PRE-PERLA Facility

In 1985 the JRC Directorate decided to build the PERLA laboratory, restructuring some existing facilities. To anticipate the beginning of PERLA-type experiments, four laboratories, storage rooms and handling rooms were immediately made available for the Safeguards programme in a protected zone where bulk quantities of fissile material in sealed samples could become operative in 1987.

NDA Laboratory

' It is a complex of seven laboratory rooms where small radioactive samples can be measured. Operative since 1986, it is dedicated to the research and development of NDA techniques.

NDA Field Facility

The NDA test field facility will be realized by modifying a large existing laboratory. It will have the same characteristics as PRE-PERLA, i.e. it will allow the measurement of bulk quantities of U and Pu. The facility will be operational in 1991.

The Spent Fuel Storage Pool

The ESSOR reactor spent fuel pond can now become very important in the global PERLA picture, because it allows the enlargement of the portion of the present fuel cycle represented in PERLA. When PETRA /4/ (a hot cell facility for treating high and very high burn-up LWR fuel in batches of 6 kg by the typical PUREX reprocessing steps) and the TAME-LAB (a tank calibration facility) become operative (1992), the JRC will have facilities representing (on small scale) spent fuel storage, reprocessing and fabrication plants, available for calibration, training and R&D.

The scope of this paper is to give a progress report of the activities carried on in PERLA so far and to report on the future planning. The paper reports in detail on the activities and results obtained in the different "functions" of PERLA, namely:
- performance evaluation function;
- preparation and characterization of large standards for NDA, with a methodology and prenormative function in the field of NDA safeguards;
- NDA system development and technology transfer functions.

2. Performance Evaluation Function

Non-Destructive Assay (NDA) techniques have, in the last few years, become more and more important and are being used to a large extent in nuclear material accountancy and control by both operators and control authorities. This is essentially due to two reasons:
1) the various improvements in most NDA techniques led some of them (gamma-spectrometry, neutron interrogation) to have performances closer to destructive analytical techniques (DA);
2) the parallel improvement of statistical and procedural inspection approaches led to abandoning the traditional scheme in which NDA is used only for semi-quantitative or consistency checks and DA is employed for quantitative measurements. In fact, NDA is now more and more used as a quantitative tool and suitable statistics procedures are developed with that aim /5/.

As a consequence the performance evaluation of NDA techniques is of particular relevance to safeguards authorities; not having the best information about the performances of NDA measurement methods means losing the potential to optimise safeguards operations and reduce costs.

One definition of "performance" has been given /6/ in the ESARDA framework:

> Whereas the knowledge of the overall uncertainty and error sources associated with measurement systems is the basic question, other important parameters must be considered in evaluating performance. These parameters are, for example:
> - reliability-ease of implementation;
> - representativeness and authenticity;
> - intrusiveness to plant operation;
> - time to obtain a result;
> - cost.

As far as uncertainty, error sources and propagation are concerned, the error structure of NDA techniques is complex but generally well known /7/ and controllable at the level of the single measurement; most of the NDA equipment today give at each determination an evaluation of the uncertainty of that measurement. Generally speaking a given NDA method has:
- random error components linked to statistical counting: they are generally well controllable through the sample count time. Their standard deviation can usually be derived from error propagation;
- systematic error components coming from calibration curves (including normalisation and nuclear constants) which cover variations in the calibration measurements, plus bias variations among the calibration samples due to some dissimilarities in matrix, geometry, composition, etc. If these sampling differences are negligible, the calibration curve systematic error can be derived from the counting statistics behaviour using error propagation;
- if the calibration "sample set" is not representative of the target population, the calibration curve can produce a bias which could vary from item to item in a "random" way (it will appear as random sampling of the population) or, alternatively, the bias will behave in a predictable way, i.e. it is constant in magnitude or it is a well defined function of the mass.

From what has been said so far it is evident that statistical modelling is a fundamental function of any "performance evaluation".

At present, PERLA staff are, in collaboration with other safeguards laboratories and in the ESARDA frame, engaged in designing and carrying out specific experimental programmes integrated by suitable statistical procedures with the scope of evaluating NDA performance values in field and laboratory conditions, following the principles mentioned above. Such values are intended to be used:
1) in planning inspections by safeguards authorities;
2) in safeguards verification and accountancy to analyse operator-inspector differences;

3) in defining the accuracy level of NDA standards.

Two examples of specific activities oriented to the evaluation of NDA performances are summarized here, concerning HRGS (High Resolution Gamma Spectrometry) on PuO_2 and MOX and calorimetry on PuO_2 bulk samples.

2.1 High Resolution Gamma Spectrometry (HRGS) on Pu-bearing material

In collaboration with other research centres an evaluation exercise of gamma spectrometry codes currently in use for Pu isotopics in international safeguards has been carried out. The results obtained by the codes, based on over 600 spectra collected on 17 Pu-bearing samples, were analyzed. The codes were experimentally compared using them to measure in the PERLA laboratory a variety of PuO_2 and MOX reference samples (PuO_2 of different burn-ups and MOX of 5 different "families"). In the framework of the CEC programme of support to IAEA Safeguards, the experiment was carried out in collaboration with Lawrence Livermore National Laboratory, where two of the codes had been developed (MGA /8/ and Blue Box), and the Ente Nazionale per le Energie Alternative (Casaccia, Italy). The experiment was carried out in the PERLA laboratory and a code developed at the Institute also participated (PUJRC). The effectiveness of each code was judged by comparing the measured gamma values with the known reference values of PERLA samples. The PERLA samples in question were a series of PuO_2 and MOX samples with well characterised values. The reference values had been established by JRC-Ispra on the basis of mass, alpha and spectrometry measurements carried out by ALKEM GmbH, IAEA Seibersdorf and CEN-SCK Mol. The procedures of preparation, measurement and characterization of these reference samples are described in /9/.

An integrated data management system /10/, with a variety of tailor-made menus allowing the user to select data sets, to apply statistical analyses anbd to create reports from the resulting analysis, was created. The system operates with a library of gamma spectrometry codes or versions of the same code. The system is implemented on a PS/2 under DOS using the INFORMIX data base management system. The software is written in C and INFORMIX 4GL. The system can also be implemented under UNIX.

Results, discussion and conclusions are extensively reported in /11/. It is worth mentioning here the outcome of that exercise - not yet published - concerning two sensitivity studies conducted on Pu-240 equivalent and the sample Effective Power.

Pu-240 equivalent. Apart from other tasks, the exercise had the aim to assess the sensitivity of $Pu\text{-}240_{eq}$ uncertainty as a function of Pu-242 uncertainty and abundance. As is well known, the Pu-242 abundance cannot be directly determined by HRGS; therefore, isotopic correlations are generally used by codes to evaluate it. Uncertainties on Pu-242 - particularly on badly known or recycled and blended materials - can then range from 5 to 20%. The results of the study are given in Fig.2. If the MGA /8/ code is used to determine all abundances but Pu-242, for which DA values are used, quite high accuracies of $Pu\text{-}240_{eq}$ are reached, better than 0.5%. With Pu-242 determined by correlation with the uncertainties as mentioned above, the total uncertainty in $Pu\text{-}240_{eq}$ can increase to 2% for "bad" materials.

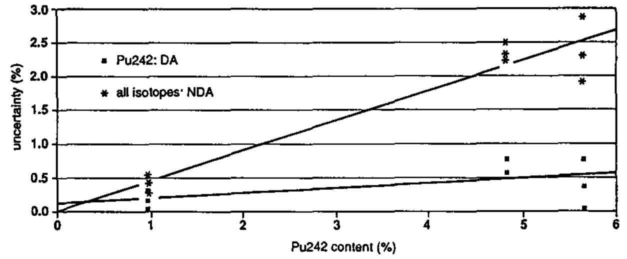

Fig.2: $Pu\text{-}240_{eq}$ uncertainty vs Pu-242 abundance: PuO_2.

Effective Power. An analogous evaluation of the error propagation from gamma isotopics to the Effective Power of the sample was carried out giving the results shown in Fig.3. The uncertainty on Effective Power from pure HRGS (MGA) increases with increasing Pu-238 content and also increases the uncertainty when "expected uncertainties from DA laboratories in 1987" are used /12/. The uncertainty behaviour on PERLA declared values is generally much lower, reflecting the low uncertainty value of these samples /9/, which corresponds to the "state of the art" (1987), rather than the state of the practice. Other samples, characterized also by DA but with higher uncertainties, might have rather higher error propagation as far as the Effective Power is concerned.

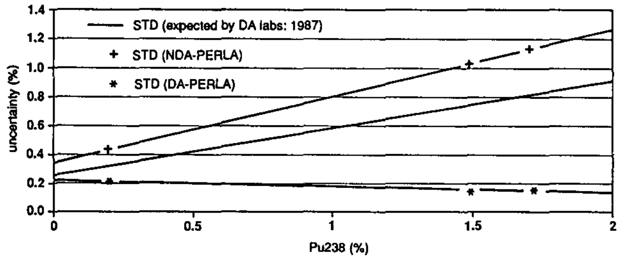

Fig.3: Effective power uncertainty evaluation from DA-NDA: PuO_2.

2.2 Calorimetry

In the last few years some attention has been paid to the determination of Pu weight by means of calorimetry /13,14/. Potentially, calorimetry is a powerful tool because of its insensitivity to perturbations like geometry, matrix, humidity, etc. Calorimetry can be used to determine or verify the Pu content of a sample under the condition that its isotopic composition is known. The Pu mass (M) is calculated following the formula:

$$M = \frac{W}{P_{eff}} \qquad (1)$$

where W is the power output of the sample (measured by the calorimeter) and P_{eff} the Effective Power (Watts per gram of plutonium); the latter can be obtained by calculating the contribution of the different isotopes according to the formula:

$$P_{eff} = \sum_{i} R_i P_i \qquad (2)$$

R_i being the isotopic fractions (measured by DA or HRGS); this carries a component of the uncertainty discussed in the previous section. P_i being the corresponding specific powers (Watts per gram of isotope).

Another component of uncertainty is carried by power measurement through the calorimeter. This measurement cannot be assumed to have a negligible error contribution, particularly when very accurate values of P_i (i.e. of isotopic ratios) are achieved. In PERLA, an error sensitivity study was carried out and tailored experiments were done. A number of measurements were collected with **air** and **water** type calorimeters employing both PERLA PuO_2 standards /9/ and MOUND Pu-238 standards. With both kinds of samples the power output ranged from roughly 2 Watts up to 40 Watts. In Fig.4 the results for different calorimeters (both air and water) are summarized.

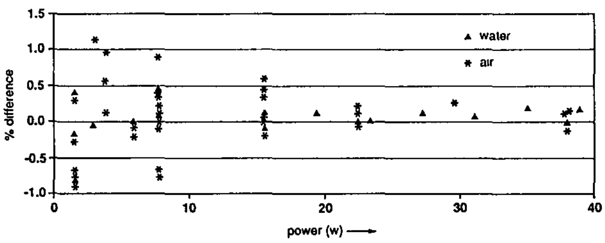

Fig.4: Comparison between declared and measured power values. Air and water calorimeters on PERLA and MOUND standards.

The overall conclusion is that the power measurement accuracy increases with increasing sample power. The water calorimeter is more accurate in the low power range. From roughly 5 Watts upwards the accuracy of power measurements becomes such that, coupled to accurate isotopics, it can guarantee overall accuracies in Pu mass better than 1%. Measurement times are in the order of 2/3 hours for air calorimeters and 4-8 hours for water calorimeters.

3. Preparation and Characterization of Standards for NDA

The increased use of NDA in nuclear material accountancy and control and the rapid improvement of NDA techniques determine that one of the major issues in the application of NDA is the availability of adequate Reference Materials or Standards.

The uncertainty with which an NDA standard or a calibration curve is known is frequently a not negligible component in the overall NDA measurement uncertainty. Since the NDA measurement uncertainty, in turn, one of the major components of the inventory verification uncertainty, one can see that only when providing suitable standards having uncertainties which are negligible with respect to other sources of error, one can appreciably improve the safeguards measurement performances in certain types of plants.

But apart from performing quantitative and accurate material accountancy, well characterized NDA standards play a primary role in:

- the normalisation and calibration of measurements;
- bias corrections;
- resolving discrepancies;
- authentication of instruments;
- NDA performance assessment.

On the other hand, the nature of NDA techniques is such taht the standards should almost always be very similar to the items verified in the field, which would represent quite a costly exercise. From that stems the need to reduce the number of standards with ad hoc techniques (Monte-Carlo calculations for instance) or with appropriate management of the system instruments/standards.

The need for each NDA inspection measurement to be traceable back to a primary standard can be fulfilled following different paths, composed of different steps; for instance, a measurement may be traced back directly to a primary standard when it exists. In other cases, the path may be much longer, implying a comparison with a secondary standard and a calculation of correction factors. Alternatively, few specialized laboratories and facilities connected amongst them in a sort of link might keep "few" standards which are representative of the major nuclear fuel productions, and then instruments are to be circulated and (inter)calibrated against those standards.

This section of the paper presents a number of considerations based on the experience gained at JRC during the operation of PERLA, and the preparation of some sets of standards /9/.

3.1 Requirements for NDA Standards

There are a number of requirements that have to be fulfilled by the NDA standards if they are to be used in the calibration and performance evaluation of NDA techniques for Safeguards:

a) They must be representative of plant samples (e.g. PuO_2 cans, MOX industrial pins, etc.).
b) They should be prepared and characterized for specific NDA methods with defined "performance values". Therefore, overall random and systematic uncertainty must be planned "a priori" so that calibrations with these standards do not introduce an appreciable uncertainty component in the NDA measurements.

c) Most of them must belong to the same family, i.e. come from the same original production batch and thus have the same chemical and isotopic characteristics. In this way, for instance the gamma spectrometrist can measure the same sample in powder, pellet and pin form, excluding any influence from the above-mentioned chemical and physical parameters.
d) Their characterization must be traceable back to primary standards through a certificate that has to fulfil precise requisites, e.g. ISO or ASTM norms.
e) A quality control procedure has to be predefined and then followed in the preparation and certification of the standards in order to ensure that the required levels of accuracy are met.
f) All possible error sources have to be defined, investigated and evaluated, bearing in mind the future application (NDA technique and predefined uncertainty).

In the following we give a list of uncertainty sources.

A. Homogeneity definition

Precision and accuracy requirements for the knowledge of weight and composition of the original batch are so stringent that the homogeneity of the batch itself is one of the major concerns in the characterization of standards. Homogeneity is requested at any stage of the preparation process. To define the homogeneity qualitatively and quantitatively, the following points can be made:

1) At the sampling level the original batch must be homogeneous with respect to the single sample size, i.e. a few grams. Therefore, DA tests at the level of approximately 1 g and collimated high resolution gamma spectrometry (HRGS) on different positions of the batch have to be planned and carried out.

2) At the single standard sample level the homogeneity is generally conditioned by the gamma spectrometry technique uncertainty.

B. Weighing

Careful sample weighing protocols have to be established for both DA and NDA samples. The characterizing laboratories have to be required to establish carefully documented balance controls based upon reference weights.

C. Humidity

One of the most important parameters which might affect NDA measurements (particularly neutrons) is the humidity content. General H_2O content limits must be imposed so as to ensure, on the one hand, a representative sampling and DA analysis and, on the other, that no further H_2O pick-up occurs after sampling.

D. Impurities

According to the specifications, impurities must be kept at a minimum level, especially those which might affect the characteristics of the chosen materials.

E. DA analytical measurements

Very detailed procedures must be adopted and imposed to DA laboratories, ranging from the number of repetitions, to the determination of the DA expected uncertainties for the laboratories and techniques used, to the types of containers to be used, to the transport of the analytical samples, to the chemical treatments and to the nuclear data set to be used in decay calculations.

Quality control measurements must be made during the analyses, employing certified reference materials in order to evaluate and correct systematic errors, if any, in the analytical techniques.

Finally, a predetermined data analysis (ANOVA or similar), evaluating averages and SDs, must be employed which is tailored to the data structure and protocols as "imposed" to the laboratories.

3.2 Standards Available at PERLA

In Table 1 a summary of the standards available or under procurement at PERLA is given. In the table the 'level of certification', i.e. the number of DA laboratories participating in the characterization and the organization issuing the certificates, is also given.

TABLE 1: Nuclear materials for PERLA

	Material type		Certific. level*
HEU	MTR platelets, plates	3 enrichments	4
	MTR assemblies (18)		4
	UO_2 powders, pellets (g.kg)	6 enrichments	3
	THTR Particles, pebbles		3
	Metal buttons (kg)		4
LEU	UO_2 powder, pellets (g.kg)		
	UO_2 pins	not yet procured	
	Short assemblies		
	U_3O_8 CBNM/NBS	5 enrichments	1
PuO_2	Small cans (g)	4 burn-ups	2
	Large cans (kg)	3 burn-ups	2
	CBNM	4 samples	1
	PIDIE	7 samples	5
	MOUND "Pu-238"	8 samples	5
MOX	Pins	fast thermal	2
	Pellets	recycle	2
	Powders		2

*The certification levels are as follows:
1) International reference material or many laboratories
2) PERLA certificate (3 laboratories)
3) PERLA certificate (2 laboratories)
4) PERLA certificate (1 laboratory)
5) Others

4. NDA System Development and Technology Transfer Function

Experience over the years has shown that the main problem in transfering safeguards techniques from the development laboratory to the field is that of producing reliable **safeguards** instruments for in-field use and of which the measurement capabilities under field conditions are well understood. It was becoming increasingly evident that most of the techniques when applied in-field had a lower performance than when used in the laboratory. The fundamental reasons were:

1) the fact that physical and statistical models were frequently borrowed from applications where error behaviour was different from that of nuclear safeguards measurements;
2) lack of careful attention of the developers who frequently developed "good instruments" instead of "good safeguards instruments", specifically customer-dedicated;
3) lack of definition of needs and functional specification from the side of the customers;
4) lack of suitable laboratories where the instruments could be tested in field conditions;
5) weakness of "after sales services" like field support, manuals, training, etc.

These deficiencies were carefully identified and discussed by ESARDA in the Como meeting of 1990 /15/.

It is the scope of this section of the paper to show general lines of instrument development in PERLA, giving a few examples, as well as to report on the progress made in training services.

4.1 Development of Integrated Instruments and Software Products

Modern NDA safeguards field instruments are quite complex tools. They must have tailored **software** that integrates different disciplines such as:

- physics;
- statistics;
- accounting techniques;
- safeguards procedures;
- software science.

Much effort must be placed in designing the architecture of the systems such that the product be really customer (inspector) oriented. Instruments must then link to a Head Quarters (HQ) data evaluation system, where historical data are stored in a structured fashion, to evaluate their long-term performances. HQ software must fulfil safeguards authorities' needs of retrieval and analysis functions, and must be able to identify the long-term behaviour of the random and systematic uncertainties of their devices in terms of parameters such as:

- characteristics of the samples (containers, material type, etc.);
- characteristics of the strata (e.g. importance of one stratum on the inventory);
- characteristics of the plant;
- measurement set-up;
- calibration curves;
- measurement procedures used.

The HQ system must be fully compatible with the field instrument in the sense that the field system must provide all the necessary information with the proper structure, must be tailored to it, but must provide wider statistical tools to elaborate very large amounts of data of the same or of different nature coming from different instruments, e.g.:

- gamma or neutron data;
- gamma **and** neutron data;
- DA or NDA data;
- DA **and** NDA data;
- NDA or C/S data;
- NDA **and** C/S data.

In fact, future (and even some present) developments consider to operate NDA in surveillance mode, or to activate NDA monitoring by surveillance, or to record NDA data in permanence but analyse only those that are "earmarked".

The role of statistics software tools and, in general, of system modelling in the next ten years in developing field and HQ integrated systems will be determinant, perhaps encompassing (in resources) that of other disciplines. Some examples of integrated systems developed at PERLA in the last few years are given in the following.

The PHONID Systems: **PHONID** (Photo Neutron Interrogation Device) is an instrument designed by the JRC for the monitoring of U and Pu-bearing samples. The latest development in the PHONID line of instruments led, on the one hand, to the construction of four exemplaires of the PHONID 3b series (three of them to be delivered to EURATOM). On the other hand, an intense measurement campaign, integrated by **Monte-Carlo** studies /16,17/ was launched to improve the electronics and to assess plutonium measurement. The entire U and Pu PERLA standard inventory was measured and the results are shown in /16/.

Field and Head Quarters **PHONID** software have been designed and produced and will be delivered together with the instruments /18/.

A lighter version of PHONID (version 4) is in the prototype phase and is developed particularly for Pu monitoring.

As far as **Neutron Coincidence Counting** (NCC) is concerned, some integrated field softwares for inspectors have been developed under EURATOM request and specifications, or are being written (**HLNCC** and **AWCC**) /19,20/. The next product will concern **NCC** for fuel bundles in active mode.

In the field of **Gamma Monitoring** the latest products were:

- the **Pu Meter** /21/: a field device for analysis of Pu spectra with MGA /8/, currently in use at EURATOM. A detailed inspector-oriented user interface has been realized around the Lawrence Livermore tool;
- the **Enrichment Meter** /22/: the old tool to derive U-235 abundance is undergoing a process of updating using the same hardware/software environment as the Pu meter;
- the **MTR Gamma Scanner** /22/: the device was designed to monitor MTR fuel elements in emission/transmission mode. Both hardware

TABLE 2: PERLA software-hardware products in support to EURATOM

Project	Field instrument			HQ software
	Hardware		Software	
	mechanics	electronics		
PHONID 3b	4 exemplaires	4 exemplaires	completed	completed
PHONID 4	1 prototype	1 prototype	partially completed	partially completed
HLNCC			completed	see n + gamma
AWCC			completed	
NCC (bundles)			under development	
MTR scanner	1 exemplaire	1 exemplaire	under completion	
Gamma plutonium		5 exemplaires (EUR)	completed	under def. specif.
Enrichment meter (U)			under completion	under def. specif.
DA-NDA db				under def. specif.
Neutron + gamma automated systems			specifications	

(scanning bench electronics) and software have been completely reviewed;
- the HQ data base for **isotopic abundances**: in the line of the software developed and used for the exercise described in 2.1 on Pu isotopics /10/, a Head Quarter data base for data coming from NDA-DA devices is under development on demand of EURATOM;
- a contribution in design specifications for a software for **Gamma-Neutron** automated systems has been given for EURATOM field devices /24,25,26/.

Table 2 gives a summary of the latest achievements and of products under development. Such products were realized by JRC mainly for EURATOM. Design specifications of most of them have been prepared by EURATOM. The realization itself was predominantly carried out with EURATOM collaboration.

4.2 Training

In recent years the training of safeguards inspectors has been seen with more and more attention, on the one hand, as a tool of technology transfer from the instrument developer to the customer and, on the other, it is undergoing a process of evolution. In the past, the major effort was put on ensuring that inspectors were able to make good measurements. Much attention is now paid to training courses which, apart from teaching the correct use of instruments, also provide a more integrated view of verification activities. Typically, the Physical Inventory Verification (PIV) type courses on plutonium and uranium, where inspectors are taught to plan an inspection, perform measurements and draw conclusions on a statistical basis, are being designed and given. Typical examples of the new kind of training in PERLA are Pu and U PIV courses, given in PRE-PERLA to EURATOM and IAEA inspectors. More than 50 training courses have been given in the last 12 years by JRC to EURATOM and IAEA inspectors on neutron and gamma techniques. The present training menu of PERLA consists of:
- basic disciplinary courses (neutron and gamma detection, calorimetry, statistics);
- integrated courses employing combined techniques (e.g. neutron and gamma);
- training in full-scale Physical Inventory Verifications in U and Pu fabrication plants, where several disciplines are involved (physics, chemistry, accounting, auditing, etc.).

5. Conclusions

The scope of this paper was to give a summary report on PERLA activities after four years of cooperation and to outline future trends. The previous chapters outlined some achievements and milestones of PERLA such as:
- development and production of NDA integrated instruments for inspectors;
- field and HQ software architectures, specifications and production;
- training and technology transfer functions;
- preparation, characterization and production of general guidelines of bulk U-Pu standards for NDA;
- NDA performance evaluation experiments and related statistical modelling.

A staff of about 25 people (roughly 50% R&D, 50% support to the Commission of the European Communities) is currently engaged in the above activities. The breakdown figures are approximately:
- 40% NDA instrument development, modelling and oriented research;
- 25% NDA performance assessment and characterization of standards;
- 20% training;
- 15% basic research.

Trends for the future are:
- more involvement in training;
- development of automated "intelligent" **safeguards instruments**;
- steep increase in modelling and software activities for field and HQ data evaluation packages;
- more careful attention to supplier/customer approach; therefore privileging:
  . oriented research
  . integrated systems at different levels:
    * neutron and gamma, calorimetry and gamma

systems
- active/passive
- DA/NDA
- NDA/C/S

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ON THE USE OF TIME-RESOLVED LASER-INDUCED SPECTROFLUOROMETRY IN THE NUCLEAR FUEL CYCLE

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Abstract

Time Resolved Laser-Induced Spectrofluorometry (TRLIS) has been used for actinides trace analysis and complexation analysis in the nuclear fuel cycle. Results obtained in the different fields such as in geology, in the Purex process, in the environment, in the medical and in waste storage assessment are presented.

Introduction

Fast and sensitive methods are more and more required in the nuclear fuel cycle for ultratrace determinations of actinides and lanthanides. Time-Resolved Laser-Induced Spectrofluorometry (TRLIS) is a method of choice for such determinations and has been used for now more than 10 years. The principle of this technique consists in pulsed laser excitation followed by temporal resolution of the fluorescence signal which leads to the elimination of short lifetime fluorescence. The main advantage of TRLIS aside rapidity and sensitivity is its triple selectivity. Excitation selectivity by the proper choice of the laser excitation wavelength, emission selectivity since each fluorescent element gives a characteristic fluorescence spectrum and finally time resolution selectivity which characterizes the fluorescent element environment. Among the actinides and lanthanides, the ones that are fluorescent in solution are uranium, curium, americium and europium, terbium, dysprosium, samarium, gadolinium, cerium, thulium. These elements have been studied in different complexing media (nitric, phosphoric, sulphuric, carbonate, micellar) and can be analysed from ppt and sub ppt level for uranium and curium to ppb level for lanthanides and americium /1-3/.

The different fields of interest in the nuclear fuel cycle where TRLIS has been used are in :

- Geology, for ultratrace determination of uranium (ng/l level) in waters from different localities in order to obtain a better understanding of uranium mobilization under natural reducing conditions and so to validate thermodynamic data /4/;
- All the different steps of the Purex process, in plutonium matrices for quality control, in organic matrices (TBP, TPH) where uranium used as tracer allows to calculate decontamination factors of new installations and hence to ameliorate performances of the process ;
- Medical surveillance for direct and fast determination of uranium in human urines. Hence, based on the uranium concentration in urine, metabolic models are established to estimate the body burden of uranium from persons exposed to uranium ambience /5/ ;
- Environment control for uranium determination in water and soil samples in order to have a "cartography" of specific areas ;
- Waste storage assessment, for the determination of complexing constants at very low level between actinides and organic matters. TRLIS allows to work directly at trace level and has been applied to curium /6/.

Moreover, in Purex process control, remote measurements are performed in hot cell with the adaptation of fibre-optics and optode and would be used for in-line control in the nuclear fuel process. This feature allows real time analysis.

Material and Methods

Apparatus

The experimental set-up is schematically shown in Fig. 1.

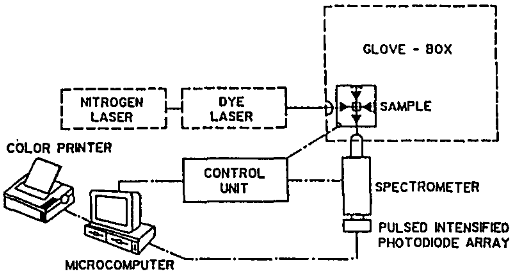

Figure 1 : TRLIS experimental set-up.

A nitrogen laser (Model 804, SOPRA) operating at 337 nm and delivering about 1.5 mJ of energy in a 5 ns pulse with a repetition rate of 25 Hz is used as excitation source. The laser beam is focused into the cell of the spectrofluorometer "FLUO 2001" (DILOR, Lille, France) by a quartz lens. In order to increase the sensitivity, two concave mirrors placed in the cell holder double the distance travelled by excitation and fluorescence emission. The radiation coming from the cell is focused on the entrance slit of the monochromator. Taking into account dispersion of the holographic grating used in the monochromator, measurement range extends to approximately 200 nm into the visible spectrum. The detection is performed by an intensified photodiodes (512) array cooled by Peltier effect and positioned at the monochromator exit. Recording of spectra is performed by integration of the pulsed light signal given by the intensifier. The integration time adjustable from 0.1 to 30 s allows for variation in detection sensitivity. Time-resolution is obtained by the control unit that assures pulsed running of the intensifier and the photodiodes array. A photodiode located in the vicinity of the cell produces with each laser pulse, a reference signal from which command signals for the high voltage are obtained. The logic circuit used to generate the signals allows measurements with a delay adjustable from 1 to 999 μs during a time of 1 to 99 μs. The signal acquisition is performed via DMA (direct memory access). All functions of the apparatus (temporal delay, gate width, integration time, reading of spectra, ...) are programmable by the use of an IEEE 488 interface. The whole system is controlled by a PC-AT microcomputer. Analytical software automatically calculates concentrations by the standard addition or the initial fluorescence methods.

Procedure

Samples are directly diluted in phosphoric acid (0.75 M), sulphuric acid (4.5 M) or Fluran (10 %) depending on the matrix composition. Then one millilitre of this solution is introduced into the quartz cell. The standard addition method is used for uranium concentration determination. Small quantities of a known uranium solution are successively added in the cell, fluorescence intensity is measured at 494 . nm (phosphoric and sulphuric acid) or 499 nm (Fluran) and then background (450 nm) subtracted. For each solution, uranium concentration in the sample is obtained by plotting fluorescence intensity versus concentration and extrapolating back to the origin.

Results

In this paper, only three examples of the previously quoted fields of application of TRLIS will be given. *In geology*, the acquisition of thermodynamic data for uranium in alkaline solution is hampered by problems such as the ionic strength of the experimental solution, the stability of U (IV) complexes and uraninite and the very low level uranium concentrations. As a result, published data are often conflicting. A set of waters from French Pyrennes at temperature ranging from 60 to 70°C were chosen to check on the accuracy of solubility data. The two possible equilibria between U(VI) and uraninite are considered.

$$U(OH)_4^0 \rightleftharpoons UO_2 + 2\,H_2O \quad (1)$$
$$\text{Log K} = 9.47 \pm 0.3 \quad (100\text{-}300°C)$$

$$U(OH)_5^- + H^+ = UO_2 + 3\,H_2O \quad (2)$$
$$\text{Log K} = -19.86 \pm 0.3 \quad (25°C)$$

From the concentrations determinations of these different sample, figure 2, where the activity of uranium in solution is plotted versus pH was obtained.

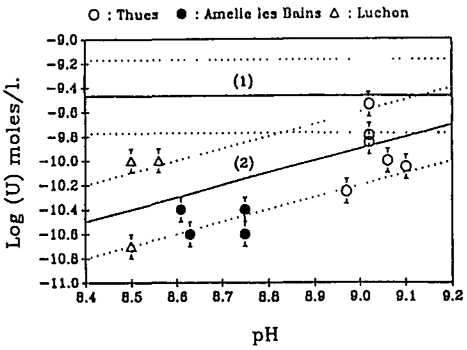

Figure 2 : Activity of uranium versus pH of groundwter samples. Solid lines represent the relation given by Eqns. 1 and 2, respectively; dotted lines give the possible limits with respect to the data accuracy.

From this figure, it seems that with equation 2, the uranium-pH correlation agrees better with the observed trends. This trend seems to confirm that uranium is controlled in solution by a mineral phase.

In the Purex process and recently in the TEO (Organic Waste Treatment) project where the main goal was to determine the decontamination factor (DF) of an installation by using uranium as tracer in the organic phase (TBP, TPH) to simulate radioactive elements behaviour. This installation is briefly shown on figure 3.

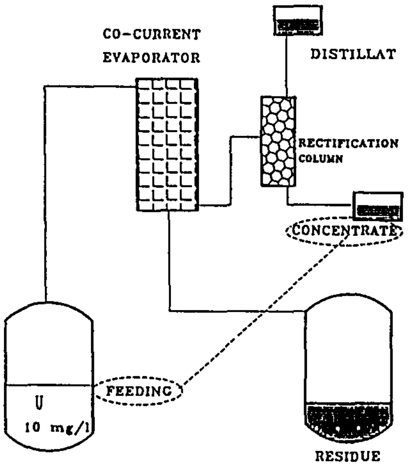

Figure 3 : Oversimplified scheme of the TEO project.

The DF factor simulated by calculation was expected to be in the 10^3 to 10^4 range. The uranium initial concentration in the feeding was set to 10 mg/l and practical measurements by TRLIS in the μg/l to sub μg/l have confirmed previously quoted figures. These numerous data (more than 1000 determinations) have allowed to visualize effects of eventual modifications of the installation on the decontamination factor.

In waste storage assessment, the knowledge of radioelement behaviour is of great importance. In particular, complexation reactions of radioelements with natural organic ligands such as humic substances (humic/fulvic acids) present in aquifer systems are important, because of the strength of complexes that humic substances form with cations, in particular trivalent elements like americium and curium. TRLIS is the only technique that allow to work *directly* at trace levels.

Typical titration curve (fluorescence signal versus humic substance concentration) of curium (III) by humic acids is presented on Figure 4 at fixed pH and ionic strength.

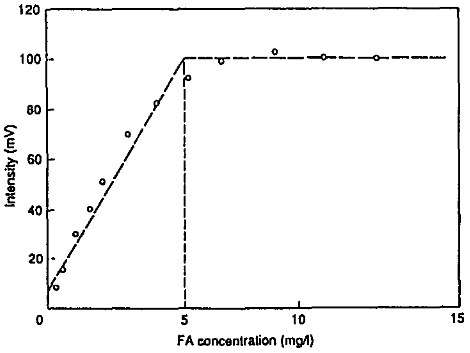

Figure 4 : Curium fluorescence intensity as a function of fulvic acids concentration. [Cm] 2.10^{-7} M,pH 5, I=0.1

The complexation of curium by humic substances leads to an increase of the curium fluorescence signal until saturation of the complexing sites occurs. From similar titration curve, different binding parameters are obtained such as the complexing capacity W (expressed in mmoles/g) and the conditional stability constant β. Several factors such as the concentration, pH and ionic strength have been investigated.

Conclusion

Time-Resolved Laser- Induced Spectrofluorometry is a fast, sensitive and selective technique for uranium determinations in the nuclear fuel cycle. Other elements such as curium, americium and several lanthanides can also be analysed. One great advantage of this technique is the possibility to perform in-line measurements by the use of fibber optics and optodes for real time analysis.

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COMPONENTS FOR AN ON-SITE SAFEGUARDS LABORATORY

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1. Abstract

The components under development for an on-site Safeguards laboratory are described. These include the instrumentation necessary for the bulk analysis of Pu and MOX samples, and for the IDMS analysis of U and Pu in input solutions.

2. Introduction

The concept of on-site laboratories has been introduced previously /1/. It includes automatic analytical instruments which could analyse nuclear fuel samples taken under routine nuclear material safeguards inspections. To reduce the labour costs in such an on-site laboratory, expert systems and computer links to a central laboratory would be employed.

The present status is described here, including:

- the bulk analysis of Pu and MOX samples, which employs robot techniques extensively,
- the components needed for alpha- and mass-spectrometry, again employing robot techniques,
- an expert system which delivers the necessary information to the laboratory robots for the appropriate conditioning procedures for the samples.

Under the present time schedule, the development and construction, followed by field-testing is planned for 1992.

3. Methods

Bulk analysis of Pu and MOX samples

For the bulk analysis of Pu and U in reprocessing output samples (nitrate solutions or oxide powders) and from fuel production (mainly MOX pellets), three options are foreseen:

- titration for quantitative element analysis and thermal ionisation mass-spectrometry (TIMS) for isotopic analysis ,
- K-edge densitometry for quantitative elemental analysis, γ-spectrometry for isotope analysis of Pu and TIMS for the isotope distribution of U (for the MOX samples only),
- passive neutron counting for Pu-containing powders plus γ-spectrometry for the isotope ratios.

The layout of a glovebox (Fig. 1) which houses instrumentation for bulk analyses is given here.

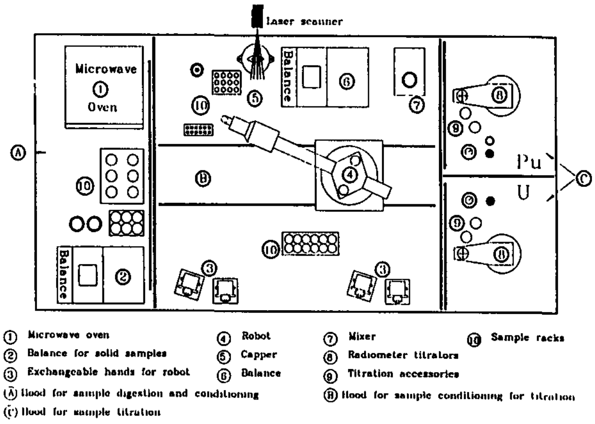

Fig. 1 Layout of glovebox for bulk analysis of Pu and MOX samples

Titration of U and Pu

For the titration of U and Pu, a commercial titrator (type RADIOMETER) has been adapted for glove-box operation and is now in routine use. All manipulations are carried out by a laboratory robot (type ZYMARK) which posses exchangeable hands for gripping (3) and for dispensing solutions. The samples are weighed (2) and dissolved. Weighed aliquots of the dissolved samples are conditioned (decapper (5), mixer (7), dispenser (9)) before titration. To limit any possible spread of contamination and to contain corrosive fumes, the glovebox is divided into three compartments with sliding connecting doors which can be opened and closed by the robot (A,B,C).

The setup has been coupled into the laboratory data system so that the expert system, described below, designates which samples have to be analysed. Barcode labels are printed which are attached to the flasks and checked at weighing (Fig. 2).

The procedure is given in Fig. 3. After dissolution and weighing of the samples aliquots the titration is then carried out automatically. All necessary data are sent to the main laboratory computer (Fig. 2). After the titration is finished the concentration is calculated so that the analyst is in the position to judge if the analysis is acceptable or not.

Blank measurements and standards are measured routinely. The results from these are archived. At the end of each day or on instigation of the operator all measured samples in the period are recalculated using the latest standards and blank values as a basis.

The results can be checked by the operator who is in the position to recall each titration curve onto the computer screen. All measurements are archived for further statistical control and also for internal fissile material control purposes.

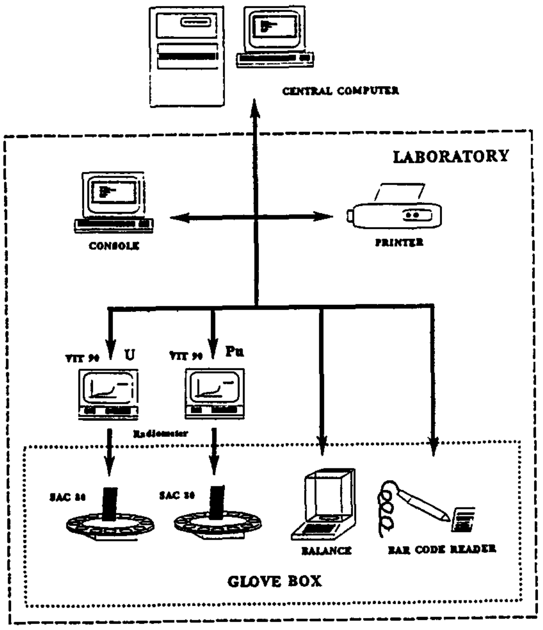

Fig. 2 Automatic titration and data flow

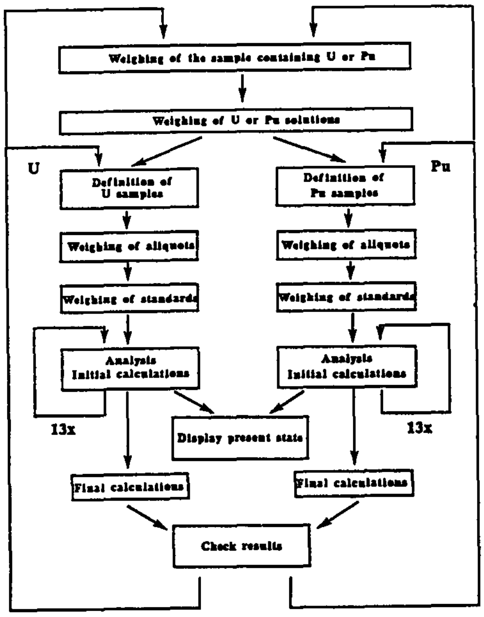

Fig. 3 Titration procedures

Enhanced robotised conditioning of solutions for isotope dilution

A system based on a laboratory robot installed in a glovebox for the spiking, conditioning and separation of reprocessing input samples has been successfully implemented in the Safeguards laboratory of the Institute and has been fully operational since 1987. This first version (Fig. 4) has been improved and extended with the intention of implementing it in the on-site Safeguards laboratories.

Two pieces of apparatus have been developed which extend the function of the robot and are described here. The first is a sample changer for alpha counting (10) to determine the ^{238}Pu / $^{(239+240)}$Pu ratios. The robot, after preparing the samples for alpha spectrometry, loads the sample changer and signalises that the first batch can be counted. It has positions for 18 samples and can measure 6 samples simultaneously. The spectra are sent from the multichannel analyser to a computer which calculates the peak ratios for the subsequent determination of ^{238}Pu and also an approximate value of the concentration of the plutonium in the solution as well as that of uranium from the concentration of ^{233}U. These concentrations are used to calculate the optimal amounts of solution to be loaded on the filaments for mass-spectrometry.

The second apparatus is for the preparation of mass-spectrometer filaments (8,13). An Eppindorf pipette is used by the robot to deposit a volume of between 1 and 10 µl of the sample on the filament. The filaments are mounted on a magazine which is rotated under robot control to expose the next filament. After all filaments have been prepared, they are heated by an electrical current to dry the samples and are then ready to be taken out for mass-spectrometry.

Extension of the expert system for the automatic safeguards laboratory

The expert system as described previously /2/ has been extended and can now choose between 22 different analytical procedures. All types of Safeguards samples can be dealt with. The program works out the following necessary parameters for the preparation of the sample:

- determination of the analysis code from the sample data with a simultaneous check of the completeness and coherence of the data,
- assignment of internal numbers which are used to check the flow of the sample through the measurement steps,
- calculation of the analytical parameters: dilution factor (should a dilution be necessary), amounts of the redox reagents in the chemical conditioning stage and weights of spikes and sample,
- determination of the number of alpha planchettes and mass-spectrometry filaments to be prepared. Later on the robot (after having identified the sample by its bar-code) will employ this information together with the parameters calculated as above.

The samples are measured and the results evaluated. Measurement repeats where necessary are then reinitialised by the expert system. Different types of repeats are possible:

- a new dilution of the original sample, assuming enough material is available, followed by a complete new measurement process,
- the chemical conditioning only is repeated,
- further measurements are needed: the robot then prepares fresh filaments and/or alpha planchettes.

The expert system at present covers the isotope dilution mass-spectrometry. It is being extended to include the bulk analysis methods for Pu and MOX samples.

Data aquisition and evaluation for bulk and isotope analysis of U and Pu

A system of automatic data collection, reduction and evaluation has been developed for all the analytical techniques at present being employed. The instruments are presently situated in different laboratories where dedicated processors collect and partially reduce the data before sending them to a PDP 11/73 computer. The full evaluation takes place later in a Micro VAX II computer.

The information from the expert system (see above) for the robot is filed in the PDP 11/73. At the same time data files specific for the sample are opened into which the reduced data are deposited after each measurement. For the moment some instruments have to be programmed by the operators according to instructions given by the expert system. In the future this step will be eliminated as far as possible.

The status of the analysis of each sample can be followed, to see for instance if the results are already available from mass-spectrometry.

For each of the instruments and robots, individual software has been written. The software is at present either being tested or in routine use. For the future an extension is planned to include an expert system for the quality assurance of the measurements.

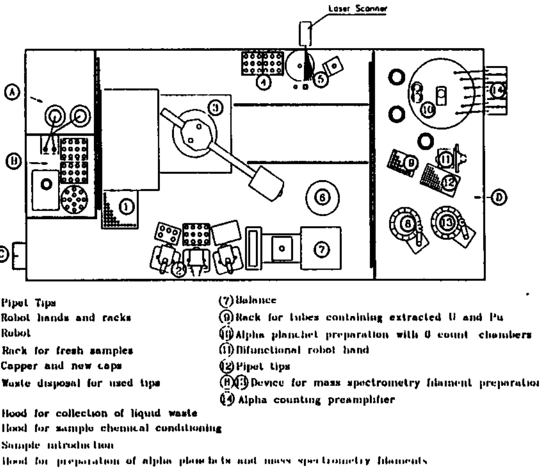

Fig. 4 Robotised conditioning for isotope dilution analysis

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APPLICATION OF THE BUBBLING TECHNIQUE FOR THE CONTROL OF NUCLEAR MATERIALS
- EVOLUTION OF THE DEVICE -

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Abstract

The paper describes an evolution of the device presented at Luxembourg in May 89 for direct measurement of liquid densities in tanks and determination of filling levels.

It appears that these two parameters can be measured with a reproductibility of 5E-5 and an accuracy of 5E-4 in relative values.

Initial device gave a result in 15 minutes, the new one provides the same result in less than 4 minutes without accuracy damaging when using a specific computer programm.

Nowadays this system is used in a reprocessing plant :

- to measure on line the density of an organic solvent in order to determine when recycling is necessary,
- to check the homogeneity of uranium nitride in large tanks.

1. Introduction

Le dispositif baptisé "monocanne" (figure 1) permet la mesure in situ de la masse volumique et du niveau de remplissage de liquides contenus dans des cuves. En particulier, il s'applique aux liquides agressifs, ionisants, dangereux ou créant un milieu hostile soit pour le matériel soit pour l'opérateur.

Le système n'utilise aucune source électrique au niveau de la prise d'information de pression au point de mesure ; seul l'air ou un gaz inerte (azote) est utilisé comme transmetteur jusqu'au capteur de pression. Ce dispositif permet en outre, par son déplacement vertical dans la cuve, un contrôle d'homogénéité en concentration et en température de la solution contenue (seuil de quelques 1E-5).

2. Rappels théoriques

2.1. Méthode générale (cf. figure 2)

Le principe de la monocanne est exposé dans la référence [1] : soit une monocanne immergée dans une solution de masse volumique W_{sol} et dans laquelle on insuffle un gaz de masse volumique W_{gaz}. Les bulles se forment successivement dans les chambres de bullage A et B et à l'extrémité C de la canne. Lorsque la bulle se forme en C, par exemple, la pression du gaz dans la bulle est :

$$P_c = P_{surf} + h_c W_{sol} g + \Delta P$$

où P_{surf} est la pression atmosphérique à la surface libre du liquide, h_c la distance du point C à cette surface et ΔP l'accroissement de pression dû à la tension superficielle.

Le manomètre est placé au point H à un niveau h_o en-dessous de la surface libre. En ce point la pression du gaz est :

$$(P_H)_c = P_c + (h_o - h_c) W_{gaz} g$$

La pression atmosphérique en H est :

$$P_o = P_{surf} + h_o W_{air} g$$

Le manomètre indique la différence de pression :

$$(P_H - P_o)_c = h_c(W_{sol} - W_{gaz})g + \Delta P + h_o(W_{gaz} - W_{air})g \quad (1)$$

Des formules analogues donnent les indications du manomètre lorsque la bulle se forme en A et en B.

Lorsque la bulle grossit dans des conditions voisines de l'équilibre (bullage lent, cf. référence [2]), la pression ΔP passe par un maximum dont la valeur est indépendante de la profondeur et pratiquement indépendante de la vitesse de bullage. Si on prend les points de pression maximum en A et B on peut écrire :

$$(P_H - P_o)_B - (P_H - P_o)_A = (W_{sol} - W_{gaz})\ g\ \Delta H \quad (2)$$

où ΔH est la différence de profondeur des points A et B. Cette distance est constante par construction.

Pour déterminer la valeur de W_{sol}, la quantité $(g.\Delta H)$ est déterminée à l'avance par étalonnage, c'est-à-dire en utilisant un liquide de masse volumique connue, l'eau en général.

La masse volumique du gaz est calculée à partir de sa valeur W_o dans les conditions normales (pression P_o et température absolue T_o), de la pression P du gaz et de sa température absolue T :

$$W_{gaz} = W_o \frac{P}{P_o} \frac{T_o}{T}$$

Il faut donc mesurer la température T au moment du bullage. La pression P est donnée par la mesure au point C :

$$P = P_o + (P_H - P_o)_c$$

2.2. Mesures relatives

La masse volumique mesurée W_{sol} est celle de la solution au point C. Une application pratique de la monocanne est donc la vérification de l'homogénéité d'une solution contenue dans une cuve. Il suffit en effet de faire plusieurs mesures en des points différents de la solution. Si l'on néglige les variations de W_{gaz} avec la pression (donc avec la profondeur), la mesure directe de la différence de pression entre A et B suffit, sans étalonnage préalable, pour déceler les variations de W_{sol}, qu'elles soient dues à des variations de concentration ou de température.

3. Dispositif utilisé (figure 3)

Le système actuel est un ensemble industriel totalement autonome et mobile : il peut être disposé dans le coffre d'une voiture. Outre la monocanne immergée dans la solution à contrôler, le système de mesure est composé d'un coffret électronique dont les éléments sont les suivants :

Le capteur de pression utilisé est un manomètre de marque CROUZET, de type 44/3. Celui-ci est relié à un multimètre HP 3456 A, utilisé en voltmètre. Un calculateur HP 9816 est connecté à l'ensemble de mesures ; il enregistre et visualise, en temps quasi-réel, la pression existant dans le circuit pneumatique associé à la monocanne. Les variations nécessaires pour obtenir le bullage de gaz sont obtenues avec deux débimètres massiques BROOKS de type 5850 E. Les consignes de ces débimètres ainsi que les électrovannes utilisées sont commandées directement par l'ordinateur, pilote du dispositif, via une interface. Un clapet anti-retour, monté directement sur la canne, évite toute remontée imprévue de la solution dans le circuit pneumatique.

De façon générale, la première mesure n'est pas prise en compte pour les calculs, cette mesure sert uniquement au rinçage de la canne et à sa mise en équilibre thermique avec la solution à mesurer.

Un programme informatique de traitement automatique enregistre en continu les variations obtenues pendant la remontée et la descente du ménisque dans la monocanne (figure 4). La détection du passage du ménisque aux points particuliers PS_A et PS_B du ménisque provoque des changements de consigne des débimètres pour permettre un bullage lent en A et B. Une optimisation de l'usinage de la monocanne en PS_A et PS_B et une étude spécifique du programme de dépouillement informatique ont permis de réduire le temps nécessaire à une mesure de 15 minutes à moins de 4 minutes.

Un écran visualise le profil des pressions pendant toute la durée de la mesure.

4. Mode opératoire

On descend tout d'abord la monocanne en fond de cuve, tout en insufflant un gaz (azote) à une pression estimée supérieure à la pression régnant en ce point. Après réduction du débit de gaz, on provoque la formation de bulles en C. Puis on fait remonter le ménisque au-dessus du point A. On enregistre pendant ce temps les variations de pression de manière à déterminer l'échelle de visualisation lors de la mesure effectuée (pendant la descente du ménisque dans la canne).

On effectue alors 5 cycles de mesures entre les points A, B et C en faisant varier la pression dans le circuit pneumatique.

Cette série de mesures étant terminée, on remonte la monocanne à une nouvelle hauteur. On observe ainsi un bullage libre dû à la diminution de pression dans le liquide. On peut alors effectuer une nouvelle série de mesures en respectant le protocole décrit précédemment.

5. Mesures

5.1. Vérification de l'homogénéité d'une cuve remplie de nitrate d'uranyle

La campagne d'essais, réalisée sur une cuve contenant du nitrate d'uranyle recyclé (présence de produits de fission), a pour objet un contrôle d'homogénéité de la solution. Une mesure absolue de la masse volumique n'étant pas demandée, l'emploi de la méthode de mesure simplifiée est suffisante : on n'effectue donc pas d'étalonnage en eau, ni de mesure de température de la solution analysée.

Les mesures présentées dans le tableau ci-après sont faites à 3 hauteurs différentes. Les différences de pression observées entre les points A et B sont exprimées en mV ; elles devraient être exprimées en unité de pression (coefficient de conversion lié au capteur CROUZET) si l'on désirait obtenir la masse volumique.

	Fond de cuve	Milieu de cuve	Sommet de cuve
	121,571	121,694	121,719
	121,520	121,863	121,764
	121,611	121,789	121,746
	121,656	121,794	
	121,550	121,693	
Moyenne :	121,582	121,767	121,743
écart-type relatif :	4,4 E-4	6,0 E-4	1,9 E-4

soit une estimation moyenne de l'écart-type relatif de 0,04 %.

Ces résultats montrent que la reproductibilité des mesures étant meilleure que 5 E-4, elle permet de déceler des variations de densité entre le fond et le sommet de la cuve.

5.2. Vérification industrielle sur du solvant recyclé (figure 5)

Une application particulière du bullage lent permet, en exploitation industrielle, la mesure en continu et en temps quasi-réel de la masse volumique du solvant organique recyclé dans l'installation. Ceci est réalisé en utilisant une canne de bullage fixe, immergée dans un pot spécifique monté en dérivation sur le flux de circulation de solvant, la solution ayant un niveau de remplissage maintenu constant par déversement. Dans ces conditions, en supposant la température constante, toute variation de masse volumique se traduit par une variation de pression.

6. Conclusion

L'expérience d'utilisation de la monocanne en milieu industriel, avec un niveau de reproductibilité de 5 E-4, a montré qu'il était possible d'utiliser un tel dispositif même en milieu contaminant. En cas de mise en oeuvre par un inspecteur chargé du contrôle des matières nucléaires, les manipulations de la canne, sur la cuve, doivent toutefois être prises en charge par un inspecteur aguerri aux problèmes de contamination. En fin d'expérience, il faut prévoir un certain temps pour effectuer les opérations de décontamination et de contrôle de radioprotection.

Un avantage de la monocanne pour les mesures in situ est la possibilité de son remplacement, en cas de détérioration, par une autre monocanne étalonnée hors cuve.

Moyennant un appareillage spécifique, une autre utilisation de ce système pourrait être la mesure sur échantillons en laboratoire, sans avoir pour cela à utiliser toute une série d'étalons, un seul étant suffisant (eau par exemple).

La mise en oeuvre d'un système de bullage à niveau constant a montré, que les techniques de bullage lent permettent à l'exploitant d'effectuer des mesures rapides, simples et fiables des paramètres nécessaires à la conduite d'un procédé (densité du TBP par exemple).

7. Références

[1] R. PLATZER, M. NEUILLY, M. ORLOFF
11 th Annual Symposium ESARDA, LUXEMBOURG 1989.

[2] R. PLATZER, M. NEUILLY, M. CARRIER, P. DEDALDECHAMP
7 th Annual Symposium ESARDA, LIEGE 1985 CR p 193/199.

FIGURE : 1

PLAN SCHEMATIQUE DU PROTOTYPE INDUSTRIEL
version portable et mobile en cuve

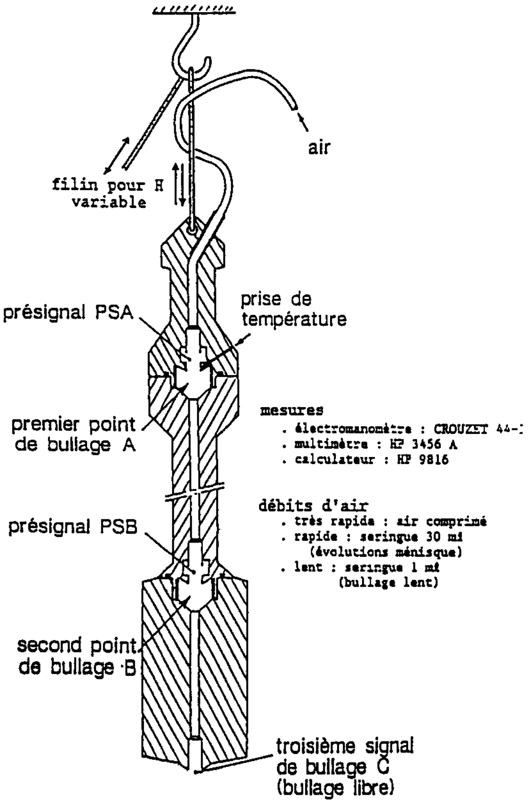
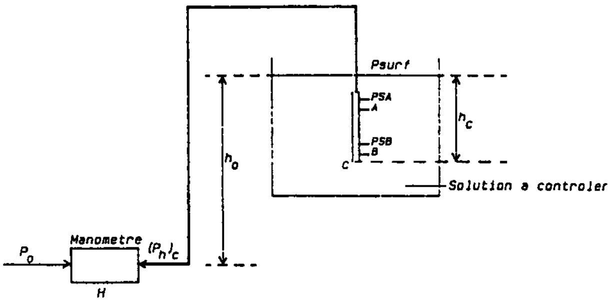


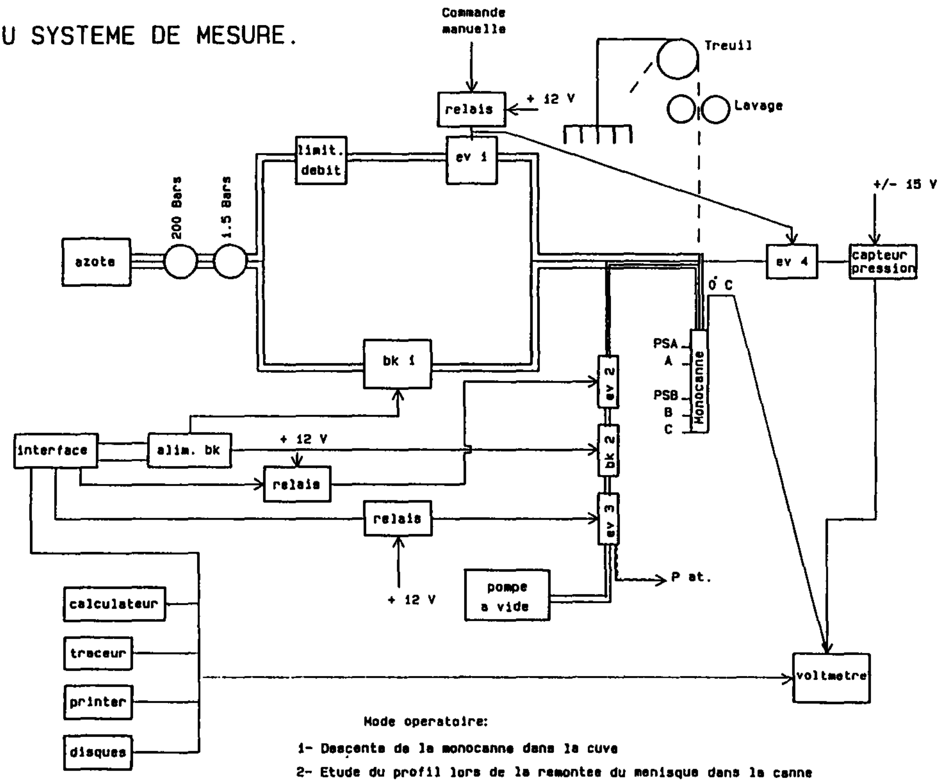
FIGURE : 2



Emplacement typique du manometre par rapport a la canne de bullage

FIGURE : 3

AUTOMATISATION DU SYSTEME DE MESURE.



Mode operatoire:

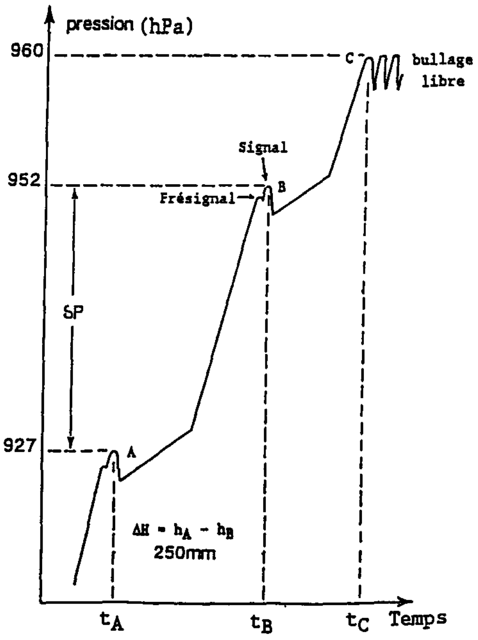
1- Descente de la monocanne dans la cuve

2- Etude du profil lors de la remontee du menisque dans la canne

3- Mesure - Descente du menisque dans la canne.

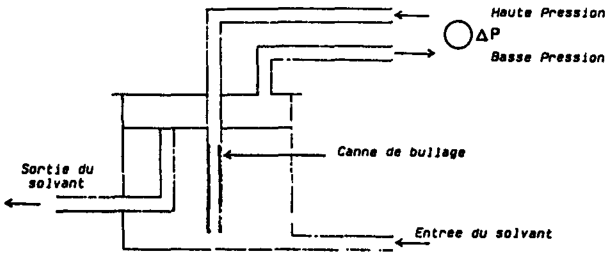
FIGURE : 4

EVOLUTION TYPIQUE P_{gaz} = f(temps)
descente du ménisque
de haut en bas de la monocanne
(à débit d'air constant)



le dessin du signal a été amplifié
pour faciliter l'observation
.en réel : quelques 10 Pa

FIGURE : 5



Verification industrielle sur du solvant recycle

FIBRE OPTIC NETWORKS FOR SAFEGUARDS APPLICATIONS

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Abstract

The Euratom Safeguards Directorate has recently installed a fibre optic network in a new large scale nuclear facility in the European Communities.

The selection, installation, and commissioning of the fibre optic network is discussed from the viewpoint of network topology, physical testing, trouble shooting and authentication.

The future use of fibre optic networks for safeguards applications is discussed.

1. Introduction

The Euratom Safeguards Directorate has over the last 6-7 years acquired a considerable pool of safeguards equipment, e.g. NDA instrumentation, video recorder systems, sealing hardware, etc. for use throughout the nuclear facilities of the European Communities. Whilst this equipment is essentially stand-alone in nature nevertheless there is an increasing demand for customized installation of equipment as a function of specific nuclear facility characteristics. In addition the Euratom Safeguards Directorate has over the past 2-3 years been increasingly requested to introduce in to new nuclear facilities specially designed equipment as an integral part of the facilities handling routines and general layout. This approach has involved the following:

- equipment included in to the routine handling arrangement of the facility
- automation of equipment to the point where direct inspector presence is no longer necessary or even no longer possible
- signal and data transmission network linking the safeguards equipment distributed throughout the nuclear facility to recording and computer systems in a central office.

With respect to this last point the Euratom Safeguards Directorate has installed in one particular large scale nuclear facility in the European Communities a fibre-optic network. This network is presently being used for the transmission of video and sensor signals and some non-destructive assay (NDA) measurement data. The network installation was started in mid-1989 and commissioned more than 1 year ago. In Fig. 1 is shown a general schematic of the network topology were it can be clearly seen that the objective is to provide a single integrate approach to data communication between local information acquisition points and a central hub for data recording and analysis. The fibre-optic network has been used to for video signals, sensor information, and some limited data communications. There have been no fibre-optic network component failures detected to date. The potential of the fibre-optic network for the transmission of all safeguards relevant signals and data is also being evaluated.

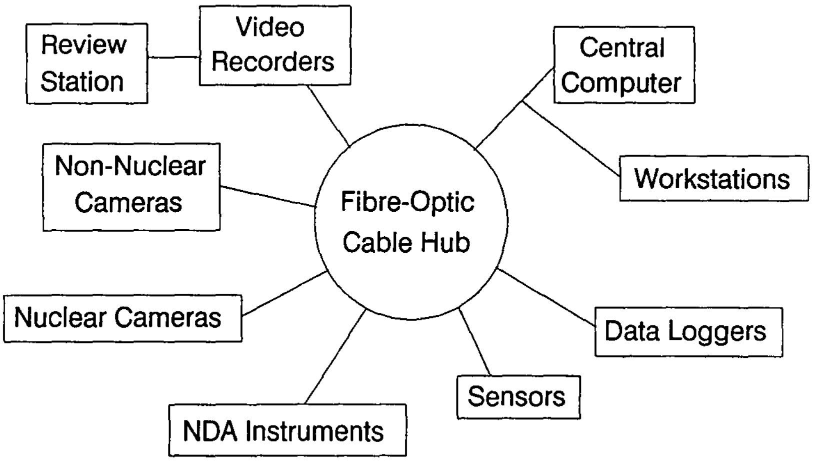
The principle justifications for deciding to install a fibre-optic cable network were that it was a proven technique, physically possible, and reasonably priced. The technical justifications for the selection of a fibre-optic cable as opposed to a metallic conductor were:

- significantly lower signal attenuation
- greater transmission bandwidth capabilities for baseband transmission
- galvanic separation between components
- immunity to electrical interference and cross-talk
- safety in hazardous or volatile environments
- smaller weight and size for the same ruggedness and flexibility as metallic conductors
- communication security, ie. more difficult to regenerate signal and introduce passive taps or beam splitters.

Here it is worthwhile to note that whilst fibre-optic cables are of course preferred for situations requiring high bandwidth nevertheless they are also extremely useful for the transmission of low bit rate signals through extremely noisy electrical environments such as those potentially found in the new automated nuclear facilities. The use of fibre-optic cables is not new in safeguards although the implementation described below appears to be the first large scale use specifically for safeguards purposes. Fibre-optic cables are used in the fabrication of several new types of sealing systems, for example, the Cobra or Vacoss, and for specific types of measurement instruments such as on-line spectrophotometry. Fibre-optic data transmission has also been implemented in the Consulha safeguards project/1/, and video signals are transmitted on fibre-optic cables for safeguards purposes in PFPF/2/.

One of the principle difficulties encountered in establishing a communications network, based upon metallic conductors or fibre-optic cables, is the lack of standard set of recommendations concerning the type of cables and network topology to be used in any particular situation. This problem was one of the reasons why the Euratom Safeguards Directorate used

Fig. 1 Schematic of Network Topology



both a main contractor with a proven experience in installing a fibre-optic network and specialised sub-contractors. In addition the Euratom Safeguards Directorate participated in the implementation of a set of detailed hardware test protocols and imposed a complete document trace for future reference. The Euratom Safeguards Directorate also participated in factory and on-site reception tests with contractors and performed separate functional tests on installed systems.

2. Topology of the Network

The first phase in the project was the selection of type of network and planning of the network topology. Given that there existed at that time no consistent set of recommendations that would have helped in the decision making process several types of networks, e.g. star, bus, ring, etc. were considered/3/. One of the main decision criteria was to provide an initial network which would reduce or remove the need for future installation work in what then would be active areas of the nuclear facility. This implied the initial installation of fibre-optic cable runs having a high level of redundancy and a built-in possibility of future development and expansion. Both the bus and ring topology were felt to have little overall resistance to failure since a single optic-fibre break would affect the complete network unless cable runs were duplicated and expensive additional electronics components added.

The star topology was felt to offer more advantages than disadvantages, it was considered as being more resistant to single fibre-optic cable breaks although it was expected to require more cable than other layouts. It was decided to adopt a point-to-point network for the video signals and to design it in such a way as to facility a physical star topology for the transmission of measurement data, etc.. The star is considered one of the most flexible and manageable types of network. The star can include logical buses and logical rings, so it supports Ethernet, token ring, and Fibre Distributed Data Interface (an ANSI LAN standard) networks. The physical star is preferred and even endorsed in the USA by Commercial Building Telecommunication Wiring Standards-made by Telecommunication Industry Association and Electronic Industries Association/4/.

The planning of the topology of the network, i.e. establishing the cable runs and paths, was dependent upon many factors, including:

- number and location of cameras, instrumentation, sensors, etc.
- location of hub
- nature of terrain, i.e. inside, outside, physical obstacles, etc.
- presence of existing cable runs
- funnelling and refilling of wall penetrations
- security and redundancy requirements
- safety standards and applicable codes of practice.

It was also felt necessary to establish some rules concerning the laying and pulling of the fibre-optic cables which would reduce the overall installation costs, permit easy functional testing and technical reception, and facility future authentication activities. These requirements were:

- use existing outside cable paths
- use existing cable paths, trays, etc. as those used by the operator
- cable paths, trays, etc. should provide for visual inspection of cables
- all distribution cabinets and all termination boxes, etc. to be sealable
- all cables etc. to be adequately labelled.

The network topology, once finalised, was drawn on to a set of site plans. Fig. 2 shows a more detailed view of the fibre-optic network topology. The network installed included:-

Fibre-Optic Cable Requirements

A total of 43,430m of fibre-optic cable was required, including 1,600m of multi-strand fibre-optic cable (24 single strands) and 2,515m of dual fibre-optic strand cable. The multi-strand fibre optic cable was required in three separate portions, two 700m portions, and a 200m portion. The multi-stranded fibre-optic cable functions as a type of "backbone" with pairs of individual fibre-optic strands connected locally to dual stranded fibre-optic cables. The dual stranded fibre-optic cable was used for both video signals and information transmission, e.g. measurement data, sensor signals, etc. A total of 1,935m of dual stranded

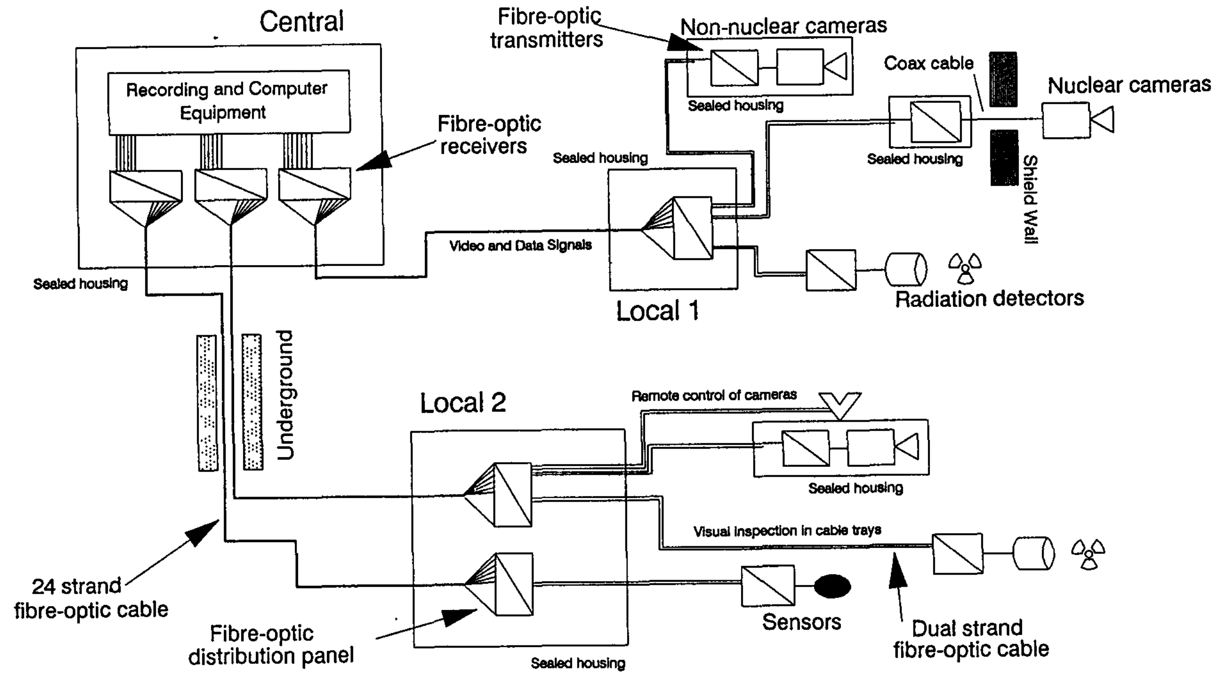


Fig. 2 Detailed Schematic of Cable Layout

fibre-optic cable was used for the video signals with local cable runs of between 25m to 110m. Information transmission is carried over 580m of dual stranded fibre-optic cable with local cable runs of 35m to 100m.

It can thus be seen that most video or information signals are carried over 750m to 850m between the point of local acquisition and the point of central recording.

Infrastructure

A total of 615m of additional cable trays were installed with open cable trays widths ranging between 50mm and 200mm. The general rule was to use where possible existing cable trays of the facility operator. The installation of additional cable trays was conditioned either by the absence of cable trays in a given part of the facility or by the need to conserve a 25% reserve occupancy in the facility operators cable trays. It should be noted that more than 60 wall penetrations were performed including load bearing walls and walls being part of the physical and biological protection.

One obstacle to the use of fibre-optic cables for the transmission of video signals is that the camera power supply must be provided using an independent set of electrical cables. In this particular case this was not see as being disadvantageous given that an elaborate uninterruptable power supply had to be installed for other reasons, but in other applications the cost of installing a separate set of power cables would need to be factored in to the overall cost/benefit analysis. A total of 4685 m of power cables were laid and pulled at the same time as the fibre-optic cables.

3. Selection of Fibre-Optic Cable and Associated Hardware

In the previous section the decision to select a physical star topology was described along with the quantities of fibre-optic cable and cable trays needed. The next phase was the selection of the hardware components, i.e. type of fibre-optic cable, terminations, transmitters, receivers, etc. Elements in the hardware decision procedure were:-

transmission requirements, bandpass and optical power budget
- power coupled to the fibre-optic strands by optical sources
- fibre-optic strand attenuation
- any fibre-fibre coupling losses, e.g. splices, etc.
- fibre-receiver coupling loss
- receiver sensitivity
- system power budget safety margin

mechanical planning
- cable runs/paths
- nature of terrain
- type of soil, etc.
- mode of laying.

Three very important points had been established during the preparation of the topology. The first was a requirement for a certain level of physical redundancy, i.e. redundant fibre-optic able strands in active areas of the facility were costs would be very high if additional cables were to be laid or pulled after active trials. The second was the decision not to mix cable types, i.e to not install a mixture of cable types such a fibre-optic, coax, twisted-pairs, etc. A single cable type reduces the management burden. The third was the need for secure links, i.e. fibre-optic links containing no connectors, splices, or passive/active components.

In addition details concerning test and reception procedures were established as were the standards to be respected in documenting the installation. It was decided that the main contractor would perform the factory reception of the hardware but that the Euratom Safeguards Directorate would observe these tests.

Selection of the Fibre-Optic Cable

The selection criteria for the fibre-optic cable was based upon both the need to transfer both video and data signals. There appeared to be at the time of installation no set of standards which would have facilitated the decision making process. It was decided to try to conform to a physical specification for a fibre-optic cable recommended at that time for IEEE 802.8 approval, see Annex I.

Additional criteria for the selection of the fibre-optic cables were:
- the requirements conditioned by the selected topology
- linking all local transmitters to a pair of fibre-optic strands for redundancy
- the need to lay an outside underground set of fibre-optic cables
- the availability of armored multi-stranded fibre-optic cables
- local codes and practices concerning fire safety, etc.

Several different suppliers were consulted and the final choice was based first upon technical specifications and secondly upon the overall cost after installation, test, and reception. The specification of the separate fibre-optic cables were:-

Outside Underground Multi-Strand Fibre-Optic Cable
- *Manufacturer:* Techmation
- *Model:* B 24 A4 FB CST/FR
- *Type:* 24 strand multi-fibre breakout (type B) cable, tight fibre with PVC sheath
- *Fibre Size:* A4 "graded index" 50/125 microns
- *Fibre Wavelength and Attenuation:* attenuation 5db/km max. at 850nm
- *Fibre Bandwidth:* FB 400Mhz/km at 850nm
- *Operating Temperature:* -40 °C to +85 °C
- *Jacket/Coating/Protection:*
    FR flame retardent (type C1)
    CST protected cable
    anti-rodent for direct burial
    armor/overjacket steel tape armor 5 mil.
corrugated copolymer coated
    lighting protection for aerial installation
    polyethylene outer jacket
    fungus resistant
    colour coded strands
    tight buffer isolates fibre coating from moisture
    fibre coated strands.

Inside Dual-Strand Fibre-Optic Cable
- *Manufacturer:* Techmation
- *Model:* B 02 A4 FB FR
- *Type:* 2 strand multi-fibre breakout (type B) cable, tight fibre with PVC sheath
- *Fibre Size:* A4 "graded index" 50/125 microns
- *Fibre Wavelength and Attenuation:* attenuation 5db/km max. at 850nm
- *Fibre Bandwidth:* FB 400Mhz/km at 850nm
- *Fibre Coating Diameter:* 250 micron
- *Operating Temperature:* -40 °C to +85 °C
- *Jacket/Coating/Protection:*
    FR flame retardent (type C1)
    colour coded strands.

Termination of Fibre-Optic Cable

A total of 284 stainless steel SMA 906 were used. It should be stressed that no splices were used and all connectors were housed in sealable distribution cabinets.

Distribution panels were placed in three sealed distribution cabinets, one placed centrally, at the hub, and two distributed at the ends of the multi-stranded fibre-optic cables. The distribution panels were supplied by Techmation and were specially designed for interconnection of outside fibre-optic cables to transmission or terminal equipment for point-to-point connection. The panels were front locking 19" rack mounted.

Fibre-Optic Emitters and Receivers

In order to provide for the signal transmission over the fibre-optic strand a special transmitter/receiver pair must be installed. The following are the specifications of these transmitters and receivers:-

Fibre-Optic Video Emitter/Receiver Modules
The FX-1000 is a analog transmitter that converts a standard composite video signal to modulated light. The FR-1000 is an analog receiver that converts modulated light to a standard composite video signal. The specification is:
- *Manufacturer:* Math Associates Inc. (USA)
- *Model:* Fibervision FX-1000, FR-1000
- *System Bandwidth (+0, -3dB):* 10Hz - 8Mhz
- *Signal to Noise Ratio:* 67dB typical
- *Linearity:* 1% or better
- *Emitter/Receiver:* LED and photo diode
- *Operating Wavelength:* 820nm
- *Optical Connection:* SMA type
- *Transmission Distance:* up to 3km.

RS-232C Transceiver Modules
The XR-1100 is a pure DC coupled digital transmission module that can convert electrical input in to light, or light in to electrical output. A pair of the modules require a pair of fibre-optic stands, can operate with any duty cycle and code format, and are suitable for simplex, or full duplex asynchronous transmissions. The modules are fully compatible with EIA standard RS-232C at data rates up to 100kbits/s. The specification is:
- *Manufacturer:* Math Associates Inc. (USA)
- *Model:* Fibervision XR-1100
- *System Bandwidth:* DC to 100kbits/s
- *System Rise/Fall Time:* 200ns typical
- *Emitter/Receiver:* LED and photo diode
- *Operating Wavelength:* 820nm
- *Optical Connection:* SMA type.

4. The Installation of the Network

Topology and Cable Runs

In order to ensure a satisfactory project management for the installation, testing, and functional acceptance of the fibre-optic network a single main contractor was selected to oversee the whole project. It was necessary to select a main contractor that was not only technically valid for the installation of fibre-optic cables but also one that had both a detailed knowledge of national and local codes of practice and a proven experience in the nuclear facility construction field.

The company selected as main contractor had the additional advantage in that they had already been involved in laying a similar fibre-optic network on the same site and thus were particularly familiar with sub-contractors having experienced and qualified staff with rights of access to the site. It is useful to note that more than 15 different sub-contractors were employed on this project.

It was decided from the outset that there would be agreed meeting notes and that the main contractor would prepared acceptance documentation for both the Euratom Safeguards Directorate and the facility operator. In addition the main contractor would also prepare documentation for submission at end of project.

Outside

The layout of the network required that the two 700m 24 strand fibre-optic cables be laid outside. Direct burial, aerial installation and installation in trenches and conduit, existing or otherwise, was considered. The most cost effective solution was to use existing sets of specially prepared trenches. The justifications for this were:
- for long outside runs a separate trench or conduit would have been too expensive
- aerial installation was not considered acceptable or reasonable (it is an unusual approach)
- trench and conduit provides best physical protection (normally used approach)
- would simplify future cable placement, replacement, and retrieval
- space was available in the existing trenches
- pulling was straight forward
- no problems with codes of practice
- existing trenches were below frost line
- the existing trenches conditioned the cable bends, etc. and thus limited eventual cable damage during laying and pulling.

The cable laying and pulling conditions are very similar for metallic and fibre-optic cables although it was expected that greater care would needed to be taken with bending the fibre-optic cable. The cables were pulled by hand (max. 600lb pulling tension) and left under limited tension (max. 135lb) to favour long term reliability.

Inside

The remaining 200m 24 stranded fibre-optic cable and the 2.5km of dual stranded fibre-optic cable were laid inside existing buildings. The connection between the outside and inside fibre-optic cables was performed in a sealable distribution cabinet. The termination of the outside fibre-optic cable in a central cabinet was the most cost effective way despite the fact that connectors and not splices were employed.

Important considerations in pulling and laying fibre-optic cables inside building were:
- the topology inside building was more complex due to the severe physical limitations imposed, e.g. horizontal and vertical runs, more short runs, more terminations, more bends, etc.
- often new cable trays had to be installed
- new wall penetrations had to made
- the laying of extra fibre-optic strands in difficult or complex areas for redundancy or future needs was nevertheless considered less expensive than to pull new cables later
- cable runs did not have to be placed under false floors or drop ceilings since in nuclear facilities everything is normally open for ease of access
- the way the installation of fibre-optic cables was combined with other installation operations, e.g. laying of power cables, etc..

The fibre-optic cables were laid in either existing or newly installed cable trays. The use of cable trays was obligatory, but nevertheless besides providing physical support they do assure greater easy of management. The installation of new cable trays, or the use of existing cable trays of the facility operators, requires that local codes for cable arrangement and tray inter-distances be respected. Specifically cables carrying electrical power, lighting, low current, control signals and measurement signals must be placed on separate cable trays.

It is useful here to also note that the facility operators cable annotation and labelling standards had also to be adopted, i.e. language, colour codes, sizes, etc. when installing cables for safeguards purposes. All cabinets etc. had to be labelled with descriptive text and warning messages against impromptu access by operator staff or sub-contractors. Naturally all cabinets had to sealable.

Cable laying and pulling conditions inside buildings and outside were relatively similar. However pulling tensions were often lower inside buildings given the shorter distances and smaller cables used.

Termination

In the distribution cabinets the individual strands in the multi-strand fibre-optic cable were directly connected to the continuation strand of the dual fibre-optic cables (2 strands) using 2 SMA 906 male connectors and a female-to-female connector.

Given that the exact length of the fibre-optic cables to be pulled were often not exactly known it was not possible to use pre-fabricated lengths of fibre-optic cable. Polishing and terminations were thus performed by hand.

The termination of the fibre-optic cables for the nuclear cameras took place in a termination box outside the cell. The nuclear camera was connected through the cell biological shield with a coax cable. The connector to the nuclear camera was selected so as to permit the connection to be made and broken using the cell manipulators. The connection of the nuclear camera directly to the fibre-optic cable inside the cell would not have permitted the removal and exchange of the nuclear camera using the cell manipulators.

Independent cell biological shield cable penetrations were included as were independent mobile camera supports on separate camera support rails fixed to the cell walls.

All redundant fibre-optic strands were terminated in the same manner as for those is use, thus providing a immediate reserve connection in case of failure of the principle strand.

5. Testing and Installation Procedures

Factory Testing

A protocol was established by the main contractor for the factory testing and reception of components at the sub-contractors. Representatives from the main contractor and the Euratom Safeguards Directorate were present during the factory acceptance at sub-contractors.

The sub-contractor performed detailed tests on each component, e.g. attenuation on the reels of fibre-optic cable to be supplied, etc.. The attenuation tests were then documented and compared with the required specification, i.e. max. 5db/km which is relatively relaxed as compared to the typical 1-3 db/km used for multimode fibre-optic cables.

One other important technical requirements was that no splices would be used throughout the network so no additional attenuation losses were expected due to splices. This requirement is considered of fundamental importance for safeguards purposes since it provides the assurance that any splice detected on the fibre-optic cable would automatically compromise the authenticity of the data being transmitted.

A continuity test was performed by the main contractor. This test consisted of the transmission of a video image over each fibre-optic strand of the different cables and connection to each and every fibre-optic/video emitter and receiver. Other tests involved the conformity of the components used, technical verification of earth, electrical isolation, transmitted image characterisation, and video signal quality.

The fibre-optic/RS-232 connectors used for data transmission and remote control functions were also tested.

On-Site Testing

On-site tests involved also the preparation of test protocols and the presence of the main contractor and the Euratom Safeguards Directorate during the tests.

The first requirement was to adjust the fibre-optic emitters and receivers. These tests involved using a sinusoidal signal and provided a test of signal amplitude and bandwidth.

The principle test for those fibre-optic cables intended to carry video picture involved the recording of a video test card. The test card image was used to assess the focus of the camera and to correct any eventual distortion in the image and excessive loss in resolution. These tests were performed with a "standard" camera, monitor, and video recorder. Additional tests, outside the scope of this review, were performed once each individual fibre-optic link was established with the definitive video camera and recording system. Separate checks and verifications were performed by staff of the Euratom Safeguards Directorate upon commissioning of the different systems. It is possible that additional tests would be required if the fibre-optic cable were to be used for highly specialised data transmission, e.g. it has been mentioned that more stringent tests would be required if a tamper resistant link were to be installed on the video signal.

The data transmission and remote control fibre-optic connections were also functionally tested.

Fibre-optic strands that did not meet the required specifications were examined using an optical time domain reflectrometer (OTDR) which can measure individual connector splice losses, locates cable damage, and document using an internal printer. The availability of an (OTDR) is essential for commissioning trials, technical reception, and fault finding, and is of great use in providing a means for authenticating the physical layout of the fibre-optic network.

Installation Protocols

In order to ensure that correct procedures were applied and that a sufficiently clear trace of the installation work was made available each and every step of the installation was defined by a series of working protocols. These protocols were issued by the main contractor and a complete set of documentation was provided to the Euratom Safeguards Directorate.

The first protocol established the conformity of the drawings of the fibre-optic cable runs with the reality and the availability of all necessary equipment and material. The conditions of each fibre-optic cable run was checked as were the locations where the cable was to be physically supported and terminated. Particular attention was given to places where the fibre-optic cable pulling would be delicate or critical. All locations where wall penetrations were envisaged were also inspected.

After the introduction of the necessary equipment and materials additional protection was applied to those locations considered delicate or critical for fibre-optic cable pulling. Additional lighting was introduced in certain specific areas, i.e. fibre-optic cables were not pulled in obscurity.

A detailed protocol was issued covering the operation of fibre-optic cable pulling. An important feature of this protocol was than pulling was performed manually using lubrification for pulling distances of greater than 20m.

A separate very detailed protocol was issued for the termination of the fibre-optic cable. It required that the personnel be trained and holding certificates of qualification. The connectors were fixed using an epoxy resin and the ends of the fibres were machine polished. The quality of the polishing was controlled using a portable microscope.

The complete fibre-optic link was then controlled using optical time domain reflectrometry to check for faults, to measure the length of the fibre-optic cable, and to verify the overall attenuation. A protocol of the tests was then issued by the sub-contractors and the existence of the fibre-optic cables was introduced in to the cable log-book. The cable log-book describes all the cables used, their types, identification codes, locations, external diameters, lengths, and type of connectors.

In addition to the installation protocols all the factory reception tests were also documented according to a separate set of test procedures and protocols. These factory reception tests covered the examination of the material to be supplied, technical control of components, performance checks, and a type examination to ensure that component dimensions and operating requirements corresponded to that envisages on-site.

Installation protocols for the nuclear cameras was particularly detailed given that once commissioned any defect or deficiency in camera installation or performance would be very difficult to correct after the cells went active, i.e. irradiated nuclear material introduced in to the cells.

Installation Diary

The following installation diary covers the entire project schedule including, where appropriate, the installation of power supplies, purchase and installation of cameras, etc.

Day 1
Order placed, project started, and team assembled.
Day 12
Shield wall inserts tender issued (9 units).
Fibre-optic cables and general hardware (supports, etc.) tender issued.
Day 19
Detailed contract study started.
Tender issued for fibre-optic transmitters and receivers.

Day 33
Civil engineering started for wall penetrations and cable trays.
Day 51
Fibre-optic cable purchased.
Day 60
Study completed involving verification of main cable routes, cable distribution points identified, and termination points verified.
Shield wall inserts purchased.
Shield wall penetrations started.
Day 71
Factory reception of fibre-optic transmitters and receivers.
Day 87
Civil engineering finished for wall penetrations and cable trays.
Shield wall penetrations finished.
Pulling of electrical cables started.
Day 105
Factory reception of fibre-optic cables finished.
Factory reception of non-nuclear cameras and sensors finished.
Day 107
Pulling of electrical cables finished.
Day 108
Fibre-optic transmitters and receivers on-site.
Day 109
Shield wall inserts delivered on-site and installation started.
Day 119
Installation of fibre-optic cables started.
Day 144
Installation of shield wall inserts finished.
Day 160
Installation of fibre-optic cables finished.
Installation of fibre-optic distribution cabinets and cable terminations started.
Installation and connection of non-nuclear and sensors started.
Day 174
Installation of fibre-optic distribution cabinets and cable terminations finished.
Installation and connection of non-nuclear and sensors finished.
Day 190
Functional tests completed on non-nuclear cameras, sensors, and associated fibre-optic cables.
Day 195
Factory reception of nuclear cameras finished.
Day 225
On-site tests of nuclear cameras finished.
Day 255
Functional tests completed on nuclear cameras and associated fibre-optic cables.

The total effort for this project was estimated at >1000 man-hours (6 people) for the project management, i.e. study (approx. 400 man-hours), purchases, contract follow-up, on-site supervision, etc., and including >150 man-hours for the testing of the fibre-optic cables and terminations as compared with 154 man-hours for the testing of all the camera and sensors installations.

In addition the installation effort was estimated at approximately 3000 man-hours (5 people). This involved >600 man-hours for the fibre-optic cable laying and pulling, and >110 man-hours for the fibre-optic cable termination and installation of the distribution cabinets.

Generally fibre-optic cables can cost up to 3 times as much as coax cables although it is probable that more coax cable would be needed for any given network topology. It is equally true that installation costs depend upon the network topology but nevertheless manpower costs for laying fibre-optic or metallic cables are more or less the same. However the total manpower requirement is greater for laying fibre-optic cables than for metallic cable.

As a percentage of the installation costs for fibre-optic cables the largest portion was for the cable laying ranging between 72% and 92%, the termination costs ranged between 3% and 12%, with testing overheads ranging between 5% and 16%.

The use of a multi-stranded fibre-optic cable over a series of single or dual strand cables probably saved up to 45% on the cost of the cable and almost certainly influenced also the installation costs.

Cable trays providing the possibility of visual inspection are between 50-90% cheaper, depending on type and width of tray, than laying stainless steel pipes which would also require some for of occasional inspecting.

6. Discussion

Security and Authentication

There are several factors to be considered in the area of security and authentication of fibre-optic cables. These can be divided in to two separate sub-areas:
- transmission security and authentication
- physical security and authentication.

Transmission security and authentication meaning the confirmation of the genuineness of the transmitted information in not considered here.

The most important aspect in authenticating the physical layout of the fibre-optic network is to detect any unacceptable physical intervention, i.e. splice, connection, etc. in the network. As has been mentioned in a previous section physical security of the link implies a physical link having no connectors, splices, or active/passive components, i.e. make it easier to detect regenerated signals and the introduction of passive taps or beam splitters. When the optical equivalence of a high impedance tap is introduced it produces a loss in the transmitted signal power of about 0.2dB/km (850nm step index multimode fibre). A splice or joint introduces between 0.1 to 3 dB attenuation and a passive tap between 0.1 to 1dB attenuation. A determined effort to minimise losses could produce splices with losses less that 0.01 dB, these splices can also be performed in less that 1 minute using an automatic high-speed fusion splicer. It is with these possibilities in mind that optical time domain reflectometry (OTDR) is introduced as a practical means of physical authentication, i.e. a practical means of detecting the illicit introduction of splices, connectors, etc. on the fibre-optic cable. For an OTDR there are four features of great importance, the ability to detect all splices, connections, etc., a minimum event dead zone, the precise determination of the fibre-optic strand length, and the physical representation of the fibre-optic strand "signature" for future reference and comparison. The required specifications for these features are:
- minimum loss readout resolution +/-0.001dB
- minimum event dead zone 0.2m, for a pulse width of 10cm and with a multimode fibre-optic strand at 850nm
- distance measurement accuracy +/-0.001%, or 1cm in 1km

- ability to display, print, record and perform comparisons of fibre-optic strand "signatures".

Topology mapping using OTDR should be possible at the maximum sensitivity so as to record every imperfection in the link. Retesting of the link should be performed systematically in order to detect the smallest deviation from the mapped data.

The use of OTDR is not the only means that should be used to physically authenticate an fibre-optic link. The physical accessibility of the cable for inspection should not be overlooked or underestimated as a authentication technique. The fibre-optic cable should follow routes which permit inspection, i.e. where possible using accessible cable runs, few wall penetrations, and open cable trays. The cable itself should be easy to identify and all cable distribution boxes, patch panels, termination cabinets, etc. should be identified and sealable. The physical layout should be well documented at the moment of delivery and all eventual modification should be equally well documented.

Future Directions

The principle justifications for selecting to install a fibre-optic cable network were that it was a proven technique, physically possible, and reasonably priced. In addition a fibre-optic cable network provides the greatest level of long term flexibility to the Euratom Safeguards Directorate since the expected lifetime of the installations could well be in excess of 25 years. It can be clearly understood that cameras, physical sensors, measurement systems, etc. may change over the next few years and that it is important that the physical network provide a stable, flexible platform for future data communication needs.

The topology chosen, i.e. point-to-point physical star, and the specification of the fibre-optic cables, "graded-index" multimode, would permit the network to be upgraded or expanded to provide both modulated transmission of video and sensors signals and the transmission of measurement data.

In practical terms the fibre-optic cable network provides:

- excess bandwidth permitting future redundant path configurations, increased overheads on information transmission, retransmission protocols, etc.
- greater efficiency in the use of future electronics systems since the network permits them to be installed centrally and to have common network circuity
- increased capability to install additional processing and storage capacity
- high reliability, maintainability, physical configuration flexibility, and lower hardware and software costs.

Looking to the near future this particular network will be developed to include the direct transfer of measurement data from the detector electronics to a central computer system for archival and analysis. Naturally this will also involve the installation on the network of intelligent terminals and workstations accessing common disk drives and peripherals. In addition a number of presently autonomous simple input-output devices will also be connected.

Another project under development by the Euratom Safeguards Directorate and its partners is the installation of a broadband network for the transfer of video, sensor, and measurement signals on a single physical carrier. This project relies on the development of industrial data communications techniques and is expected to commissioned in the next 1-2 years.

7. Conclusions

A fibre-optic cable network consisting of more than 45 km of fibre-optic strands has been installed for the transmission of camera, sensor signals, and measurement data, for safeguards purposes. After more than 1 year of operation there have been no hardware component failures in the fibre-optic network.

The topology is based upon point-to-point connection following a physical star layout. The specification of the fibre optic cable, the selection of connector types and associated ancillary hardware has been discussed. Particular emphasis was placed on the physical redundancy and availability of the network through the use of redundant fibre optic strands, etc.. Attention has been paid to the problems of physical security and authentication of the network.

The following two points are worth repeating:-

This type of project requires a project management approach that is best implemented through a main contractor and a series of sub-contractors, with the main contractor handling all the project management, purchase routines, factory and on-site reception tests, etc.

An optical time domain reflectometer is an essential tool in the process of authentication of a fibre-optic cable used for safeguards purposes.

A fibre-optic cable network of the type described is a mature approach to the central recording of both camera and sensors signals, and measurement data. The fibre-optic cable network installed is being evaluated by the Euratom Safeguards Directorate for use as a local area network (LAN) for the communication of measurement data, i.e. Ethernet. Other transmission media are also under evaluation by the Euratom Safeguards Directorate for the networking of both camera and sensor signals and measurement data on one physical carrier, i.e. broadband technology.

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Annex I - Selection of the Fibre-Optic Cable

The physical specification for a fibre-optic cable was that recommended at that time for IEEE 802.8 approval:

- *Fibre Size:* 62.5/125 microns
- *Fibre Core Size Tolerance:* +/-3.0 microns
- *Fibre Cladding Size Tolerance:* +/-3.0 microns
- *Fibre Nominal Aperture (NA):* 0.275
- *Fibre NA Tolerance:* +/-0.025
- *Fibre Coating Diameter:* minimum 250+/-15 microns
- *Fibre Wavelength and Attenuation:* 3.0 - 5.0 dB/km at 850 nm
- *Transmit Device Wavelength:* 790-860 nm
- *Fibre Bandwidth:* 100-500 Mhz/km at 850 nm
- *Connectors:* no recommendation
- *Optical Power Budget:* proposal dependent.

SPECIFIED REFERENCE MATERIALS FOR URANIUM ISOTOPIC MEASUREMENTS BY GAMMA RAY SPECTROMETRY

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Abstract

As a result of collaboration between the IAEA and the Research Institute of Atomic Reactors Uranium Enrichment Reference Materials fo gamma- and X-ray spectrometric measurements have been manufactured and characterized. The main features of the UERM: it is "quasi-infinitely thick" for photons up to 186 keV, and metallic uranium was used in it as reference material. The set of eight samples covers the full range of U-235/U abundances from 0.47% to 90%. Each sample is sealed in a rigid capsule. Abundances of the uranium reference material samples were characterized by three laboratories and UERMs characteristics were investigated by the NDA technique. In addition, a set of metallic Uranium Gamma Sources (UGS) was fabricated to be used as normalization sources.

1. Introduction

Two types of new standards for the NDA gamma-spectrometric measurements were produced in the Research Institute of Atomic Reactors (RIAR), USSR, as the result of close collaboration with the IAEA. The first one is a set of eight Uranium Enrichment Reference Materials (UERM) for an apparatus calibration and the second - Uranium Gamma Source (UGS), as normalization sample. Main characteristic features of both standards: metallic uranium as reference material and a rigid, hermetic stainless steel (SS) capsule. Each sample of the UERM is made of a 100 gram metallic uranium disk (2.6 mm thick), sealed in SS capsule (68.5 mm in diameter and 8 mm in height) with 0.48 mm SS window for photon passage. The set of eight UERMs covers the range from 0.47 to 90 at % of U-235, the U-235/U abundances have been measured by three laboratories by means of a samples mass spectrometry assay. The NDA technique (gamma spectrometry and gamma transmission) was used for homogeneity tests of every UERM. The aim of the paper is to describe UERMs and UGS preparation and performances and discuss the analytical results obtained.

2. General Requirements for the Standard

The nondestructive gamma spectrometric assay of the U-235 enrichment has reached a high level of accuracy, somewhere comparable /1/ with the destructive one. But without reliable standards NDA measurement is a relative measurement.

In /2/ it is shown that a characterization level better than 0.1 % is required for standards of the U-235 abundance determination.

This target performance level was in mind to the moment when the decision was formulated: in frame of the USSR Support Program to design and create standards of metallic uranium in wide range of U-235/U abundances. Previously it was proposed to manufacture secondary standards, which are less expensive and relatively quick to prepare. Such reference materials could be still very useful for qualifying the field measurements, improving NDA performances and create gradually an improved traceability of NDA measurement to primary standards or International Reference Materials.

Principal requirements to Reference Materials (RM) were worked out rather long ago /3/ as follows:

- uncertainties of the main characteristic (in our case-it is enrichment U-235/U) have to be small and well known;
- predictability in the response produced in the measurement process. (Ideally, the measurement process will respond to the reference material in the same way as to the unknown to be measured. If there is a difference in measurement response to the measured parameter arising from other measure-affecting factors, these effects must be known and quantifiable);
- adequate stability with respect to all measurement-affecting characteristics of the Reference Material. (It is necessary to avoid systematic errors due to changes in such properties as density, concentration, shape, and distribution);
- availability in quantities adequate for the intended application.

The analysis of factors, which influenced the enrichment measurement by gamma spectrometry /3/ makes us confident, that metallic uranium is the best material for standards: uranium - a very stable material, and is not so difficult to fabricate any form specimen from it, with very good measurable geometry. Because of high density and nuclear charge of uranium Reference Material become very compact (small in size). Being sealed hermetically into the proper capsule the uranium specimen keeps its characteristics for a very long time.

Specific requirements for enrichment reference material have been formulated by Agency employee concerning the to following features:

- physical form of the RM - uranium metal disk;
- dimensions of the RM: about 52 mm in diameter and "quasi infinite" thick for 185.7 keV gamma-rays;

. diameter of the capsule - 67.5 mm.

. quantity of standards - not less than seven standards, covered enrichment range up to 90% U-235/U;

. exact value of the enrichment must be known with uncertainty about 0.5% (relative).

Later on, when procurement scheme was worked out and manufacturing process was initiated, the characterization possibility by mass spectrometry assay of uranium samples in several laboratories of Agency net was coordinated .

3. Short Description of the UERMs

Practically all the range of enrichments is covered by means of two sets of standards (named Uranium Enrichment Reference Material, UERM). Set N1 of four low enriched uranium (LEU) UERMs includes 0.7; 1.9; 4.9; 12% enrichments. Set N2 - includes three high enriched uranium (HEU) standards (90; 60; 26%% enrichments) and additional to - 0.47% standard. The complete set contains also an empty capsule of UERM, a window material disk and stainless steel (SS) transport container (fig 1). The last one has nearly the same dimensions (88 mm in diameter and 92 mm height), as one of well known Certified Reference Material EC-171/NBS-SRM-969 (80 mm in diameter and 90 mm high).

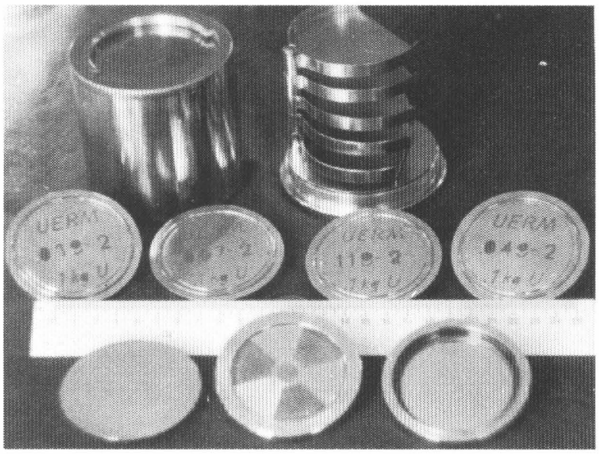

Fig.1. Set of four UERMs

The standard design (fig 2) provides perfect mechanical property and seals the 0.1 kg uranium disk (51.86+-0.16 mm in diameter and 2.60+-0.05 mm thick) . The capsule has the window (0.483 +-0.002 mm thick) for gamma rays passage slightly recessed in capsule body so that uranium disk situated at 1.20+-0.08 mm from window site surface of the UERM. The 2.6 mm uranium disk is quasi infinitely thick for 185.7 keV gamma rays of U-235.

Most of identifications deposited on the capsule surface in a form of TiN 3 mkm coating:

.on the top cover there is sign "UERM", three figures after point mean enrichment in parts (like ".119-3") and through dash the number of set, mass of uranium in the form of ".1 kg - U".

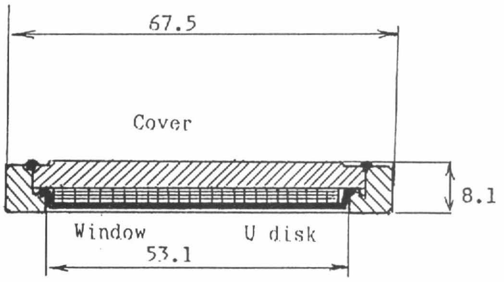

Fig.2. Cross section of the UERM

.on the window there is the sign "Radioactivity".

4. Procurement and Quality Control

Only five LEU and five HEU sets of all were manufactured. Making of the standards can be divided conventionally into several stages:

.define preparation and characterization schemes to attain specific requirements to all elements of the standard;

.preparation of uranium disks with enrichment required;

.capsule details fabrication;

.elements quality control and sets compiling;

.standards assembling and characterization test.

For each of these problems the requirements were defined and uncertainty of element parameters were assumed, securing minimum of the uncertainty when applying the set of standards.

At the stage of uranium disk preparation these parameters characterized the quality of the details and manufacturing process:

.enrichment U-235/U value and uncertainty;

.inhomogeneity of enrichment;

.local arial uranium density;

.uranium disk dimensions and mass.

Uranium disk fabrication was conducted as follows:

Initial materials proper to composite set of enrichments 1.9; 4.9; 12 %% were uranium metal 0.47% and 26.1%, and 60% enrichment standard was composed with 90% and 26.1% uranium.

Proper amount of initial materials (0.8 kg in all) was melted together in an inductive vacuum smelting furnace in MgO cap. The uranium ingot was crashed into small peaces and a part of the amount was smelt again to prepare a 0.16 kg ingot in succession.

Each ingot was tested for enrichment (by gamma spectrometry) and density (by hydro-weighting in CCl_4). Then the ingot was subjected to hot pressing in the vacuum furnace to get the ingot in form of a mushroom convenient to lathe processing.

Uranium disks were passed through complex tests allowing to determine all the parameters required.

There is some peculiarity in the capsule design. The first one is connected with the window material-calibrated and polished stainless steel sheet, that provides window thickness uncertainty less than 0.005 mm (last one value of thickness uncertainty may result in 0.1% gamma-ray 186 keV emission change /3/).

The second one: recession of the window into capsule body that prevents the window from accidental mechanical failure. This parameter was included in the set certificate.

The window, the cap and capsule body were connected by argon arc welding hermetically. Before standard assembling all details were sorted carefully in such a manner, that one set of standards had characteristics and parameter values with minimum discrepancies.

5. Standards Characterization

The standards characterization scheme foresees the use of NDA and DA at different stages of fabrication.

The most important analysis for enrichment standards - isotopic distribution was made by means of the mass spectrometry (M.S.) assay. The impurities in metal - by the emission spectroscopic analysis and chromatography measurements.

Different gamma count rate measurements have been performed with the help of the high resolution Ge counting system:

.the uniformity of the gamma count rate of U-235 at 185.7 keV as a measure of the enrichment uniformity of the uranium disk;

.the transmission tests were done using a collimated gamma-beam of Cs-137 for determination of the count rate of the transmitted gamma-rays at 661.6 keV. They are a measure of the uniformity of the disk local areal density;

. the occurrence of spurious gamma-lines uranium minor isotopes and daughter products.

5.1. Mass Spectrometry Measurements

The mass spectrometric characterizations analysis for enrichment reference materials were performed using the uranium samples in form of metallic uranium turnings.

The detailed isotopic abundance data measured in RIAR (further marked as H) are shown in table 1

Table 1. Isotopic abundances (mass %) of the UERMs

Sample	Isotopic abundances, mass %			
id.	U-235 (RSD,%)	U-234	U-236	U-238
.005	0.458 (0.65)	--	--	99.542
.007	0.707+-0.57	--	--	99.293
.019	1.839+-0.24	0.018	0.004	98.102
.0194	1.931+-0.26	0.013	0.004	48.028
.049	4.863+-0.05	0.034	0.023	95.021
.119	11.894+-0.06	0.081	0.034	87.858
.261	26.086+-0.04	0.188	0.094	73.413
.600	60.067+-0.03	0.556	0.078	38.946
.900	89.916+-0.03	0.890	0.270	8.85

Isotopic measurements were performed also in analytical laboratories of Seibersdorf (S) and Petten (P) using the following special scheme:

The sample (1 g) was split into two equal parts and they dissolved independently.

Two aliquotes from each dissolution were taken for analysis of uranium concentration. The results of four measurements - laboratory mean enrichment and RSD, which is really only a measure of scatter of the values.

The comparison of uranium materials characterization results is shown in table 2 in the form of the relative difference "laboratory mean" for each couple of the laboratories, all measurements "grand mean" and RSD of the "grand mean".

Table 2. Differences between "laboratory means" H,P,S laboratories , "grand mean" enrichment value and its RSD.

Sample	Relative difference, %			Grand mean	RSD
id.	H-P	H-S	P-S	at%	%
.005	-8.66	0.46	9.19	0.477	5.33
				0.463*	0.32
.007	0.98	0.10	- 0.88	0.713	0.54
				0.716*	0.07
.018	3.13	-0.19	1.47	1.881	1.90
				1.861*	0.14
.019	-0.36	-4.16	3.80	1.978	2.02
				1.958*	0.18
.049	-0.63	-0.06	-0.57	4.933	0.34
				4.924*	0.04
.119	-0.06	-0.19	0.25	12.032	0.13
.261	0.07	0.08	0.15	26.301	0.08
.600	0.05	0.254	0.30	60.385	0.15
				60.341*	0.04
.900	0.03	-0.16	-0.11	90.006	0.06
				89.977*	0.02

Relatively great standard errors of some "grand mean" allow to propose that some of the measurements results are biased. Suppose that is .005; .007; .019; .049 (for lab P) and the second dilution sample measurement of .0194 and - .600 (for lab S). When removing these results we can get the "best" standard error, which value seems to be more realistic and nearer to the declared laboratory uncertainty (about 0.1% for uranium samples with more than 2% enrichments.

5.2. Comparison with Gamma-Ray Measurements

All uranium "bare" disks were carefully counted by means of high resolution gamma-spectrometry (G.S.) system. The experiments had several goals: to characterize a consistent wide-range set of the enrichment reference materials, to clarify some discrepancies of the mass spectrometry measurements, and display a consistency between the certified (by M.S.) parameters and measured (by G.S.) quantities.

The count rates of 185.7 keV line were measured with precision from 0.04% (for 90%) to about 0.1% for 0.47% uranium. The pile-up correction by Am-241 reference source method obtained not exceed 1% for 90% uranium. Averaged 5 disk count rates was used for determination of the ratio n/Ai (where Ai - enrichment U-235/U in at%). Which, in principle, should be a constant for every UERM in-a wide range of enrichments. The parameter n/A analysis showed, that for all disks with enrichments 4.9% and more , and for all M.S. laboratory means the average value was n/A=71.61 l/(c*%) with RSD=0.15%. In table 3 there is relative deviation of individual laboratory n/A from this average value.

Table 3. The relative deviation (%) of laboratories n/A from averaged value for UERMs with enrichment >4%

Sample	Relative Deviation, %			RSD n/A,	Adjusted
id.	H	P	S	%	A, at%
.005	-0.44	-8.84**	2.2	2.8	0.462
.007	0.91	1.9 **	1.01	1.5	0.722
.019	0.72	-2.41**	0.91	0.44	1.875
.0194	-0.28	-0.63	-0.45*	0.43	1.950
.049	0.16	-0.47**	0.10	0.16	4.930
.119	0.18	0.24	-0.01	0.12	12.048
.261	-0.05	-0.12	0.04	0.05	26.290
.600	-0.11	-0.06	-0.04*	0.07	60.297
.900	-0.05	-0.02	-0.13	0.06	89.946

* Excluded: 2.038 and 60.519 at% of the second dissolutions assay.

** Suspected.

A first general comments of Table 3 concerns the rather good consistency between the laboratory M.S. values that are tied together by normalizing to the average n/A for five reference materials: 4.9; 12; 26; 60; 90%% enrichments. Only one of the lab P value (.049 concerned) occurs outside of +-0.3% limits value.

But it does not desirable to suspect this result, because of the "grand mean" of all .049 measurements is within +-0.3% certified value, (see fig 3). The RSD of the "grand mean" is a scatter therein.

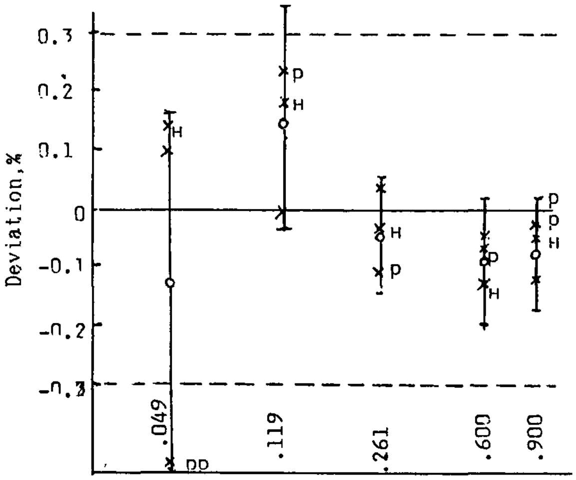

Fig.3. Plot of the n/A "grand mean" deviations for five RM : .049; .119; .261; .600; .900; from the average n/A for these RM

For these 5 lots of uranium, there is no reason to doubt that the average value of the mass spectrometry results is the best value of the average enrichment, and they can be designated as the primary working uranium enrichment reference materials.

Secondly. As for materials with enrichments lower than 4.9 % - most of the M.S. and G.S. measurements had unsatisfactory the consistency difficult to explain now. That is why the enrichments of the UERM with lower than 4.9% enrichment have to be adjusted to make the gamma counting measurements on them consistent with the 5 "primary" standards, as it can be seen in column "Adjusted A, at%" table 3. Then, four reference materials: .005; .007; .019; .0194 can be designated as secondary standard because of greater error for gamma-ray counting statistic, mass spectrometer determinations and sample representativeness problems.

5.3. Some Additional Uncertainty Sources

The gamma spectrometric measurements showed that enrichment inhomogeneity for the 5 "primary" reference materials does not exceed 0.15 % for .119 RM and is less than 0.1% for others.

The results of the gamma transmission measurements by 661 keV gamma-rays transmission gaves average areal density value 4.84 g/cm^2 (RSD=0.4%) for most (but two) of the 45 uranium disks. Minimum value of local areal density was still higher than 4.7 g/cm^2 for quasi-infinite thick uranium and did not influence the enrichment determination by gamma spectrometry.

Impurity analysis by emission spectrometry and chromatography (for carbon) assured that contents of the impurities with Z<30 did not exceed 0.5 wt%, with Z>30 - not more than 0.02 wt%. These amounts of impurities decrease 185 keV gamma-rays emission by not more than 0.07 % compared with pure uranium metal.

The UERM window thickness uncertainty q0.002 mm is less than the tolerance steel wall thickness and will generate about 0.04 % uncertainty of the measured counting rate.

We have not found any unforeseen interfering to 185.7 keV line gamma-rays sources except the trace of the Pb-212 (U-232 descendant) for .600 and .900 reference materials.

It should be mentioned that most of the individual physical properties of the UERM are well specified and it is possible to correct most of the systematic errors. An estimation of the overall uncertainty has given 0.15 % for HEU sets of UERMs, and about 0.35 % - for LEU sets.

6. Uranium Gamma Source

One more Reference Material prepared in RIAR is the Uranium X-ray and Gamma ray Source (UGS) , which can be used as a normalization source to:

- . check instrument reproducibility;
- . revalidate physical standards integrity;
- . normalize data at a later date.

A complete set of the UGS consists of the source capsule and aluminum support ring (67.5 mm in diameter and 8 mm in height). The sealed SS capsule (40 mm in diameter , 4 mm in height) contains SS substrate (20 mm in diameter, 1 mm thickness) one side of which is covered with metal uranium (90% enrichment). Uranium layer thickness ranges from 0.1 to 0.15 g/cm² and has an areal density irregularity not more than 15%. The mass of the uranium defined with the standard error less than 0.1%. Gamma rays go out of source through the 0.47 mm SS window . UGS is an easy to use tool because of all necessary signs and data deposited ed on the surface of capsule and the ring.

7. Conclusion

The experience of the Agency and RIAR (USSR) collaboration in new standards procurement and characterization was successful. NDA and DA investigations of the Uranium Enrichment Reference Materials confirmed that the set of UERMs possessed some useful features:

. two sets LEU and HEU UERMs cover a wide range of enrichments up to 90%;

. characterization uncertainty for UERMs with enrichments ranged from 4.9% to 90% do not exceed 0.3%, and are confirmed by inter-laboratory measurements of the uranium samples and careful NDA of the UERMs;

. set of UERMs - rigid, compact and transportable, convenient to use in laboratory and field.

It is worth to emphasize, that the results of the mass spectrometry measurements were obtained recently and analysis of them has to be qualified as a preliminary. It would be very useful to confirm declared UERMs performances by means of testing them in one of the internationally recognized NDA laboratory, like PERLA.

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ANALYSIS OF Th-U, U-Pu AND Pu SOLUTIONS WITH A HYBRID K-EDGE / XRF ANALYZER

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Abstract

The Safeguards Analytical Laboratory (SAL) of the IAEA has been equipped with a hybrid K-edge/XRF analyzer for the measurement of nuclear materials in liquors. The purpose of the instrument is to establish at SAL the technical capability to provide IAEA inspectors with experimental support and training in the use of such instrumentation for in-field measurements. Typical performance data for different applications are presented and discussed. The work presented is done in the frame of the Joint Programme of the IAEA and the Federal Republic of Germany for the Development of Safeguards.

1. Introduction

The Safeguards Analytical Laboratory (SAL) of the IAEA has been equipped with a hybrid K-edge/XRF analyzer similar to the instrument recently installed by the Euratom Safeguards Inspectorate at the UP3 spent fuel reprocessing plant at La Hague /1/. The decision for both installations has been made upon the successful demonstration of a prototype hybrid K-edge/XRF system designed for the uranium and plutonium assay in dissolver solutions from a reprocessing plant. The previous performance tests with this prototype, conducted between 1986 and 1988 in the frame of Joint Programme of the IAEA and the Federal Republic of Germany for the Development of Safeguards, have demonstrated the potential of such instrumentation for on-site safeguards verification measurements /2/.

The instrument installation at SAL, carried out in the frame of a task within the German Support Programme to the IAEA, represents a follow-up action to the aforementioned work. The main purpose of this project is to establish at SAL the in-house experience and the technical capability to provide IAEA inspectors with experimental support and training in the use of such instrumentation for in-field measurements. In addition, the analyzer is also designated for providing support to some of the routine analytical work on safeguards verification samples at SAL.

The present paper reviews the joint work of IAEA-SAL and KfK so far done under this project. A significant portion of this work has been devoted to careful performance studies for a range of applications. The results from these studies are presented and discussed along with a description of the principal features of the analyzer.

2. Instrument Design

General Layout

The principal design of the analyzer is based on the proven 'Hybrid Instrument' configuration, which combines the techniques of K-edge densitometry (KED) and X-ray fluorescence analysis (XRF) of K-series X-rays /2, 3/.

The analyzer design for SAL had to take into consideration the following criteria :

- Simple adaptation to one of the standard gloveboxes at SAL with the least possible modifications;
- Incorporation of a sample changer for unattended automated measurements on a larger set of samples;
- Optimization of the measurement precision for KED at element concentrations of about 100 mg/ml;
- Layout of the counting geometry for XRF to allow precise measurements on mg-size quantities.

The system assembled to meet the above criteria is shown on the photograph in Fig. 1. The mechanical assembly together with the shielded X-ray tube and the detectors for KED and XRF are mounted on a wheel table located beside the glovebox. A linear sample changer, driven by a stepping motor, extends into the box and moves the samples to the measurement position in the analyzer. The set-up is coupled to the box by means of a sealed flange in its side wall.

Counting Geometry

The sectional drawing in Fig. 2 details the counting geometry of the analyzer. The transmission measurements for KED are made on high-precision glass cells, 4 cm long and 0.6 cm wide, requiring a minimum sample volume of about 2.5 ml. The straight-through beam is collimated by means of a 10 cm long tungsten collimator with a beam hole diameter of 0.1 cm. A micrometer allows to adjust the horizontal position of the collimator relative to the beam axis for maximum counting rate.

The XRF measurements are made on thin-walled PE vials with an inner diameter of 0.5 cm at the beam level. The effective sample volume seen by the XRF detector is about 0.2 ml, which represents the minimum sample volume required for an XRF measurement. This sample geometry was chosen to concentrate mg-size quantities into the smallest possible volume. The XRF vial is 7 cm distant from the focal spot of the X-ray tube. The solid angle for the detection of the fluoresced X-rays can be varied by means of exchangeable apertures, which are mounted into a tungsten sleeve in front of the XRF detector.

Different sample magazines are available with positions for glass cuvettes or PE vials only, or with alternating positions for both types of vials as shown in Fig. 2. The center-to-center distance between adjacent sample positions on a magazine is 1.4 cm. Two sample magazines, each having positions for 14 samples, can be put in series into an adapter mounted on the sledge of the linear drive for automated measurements on up to 28 samples.

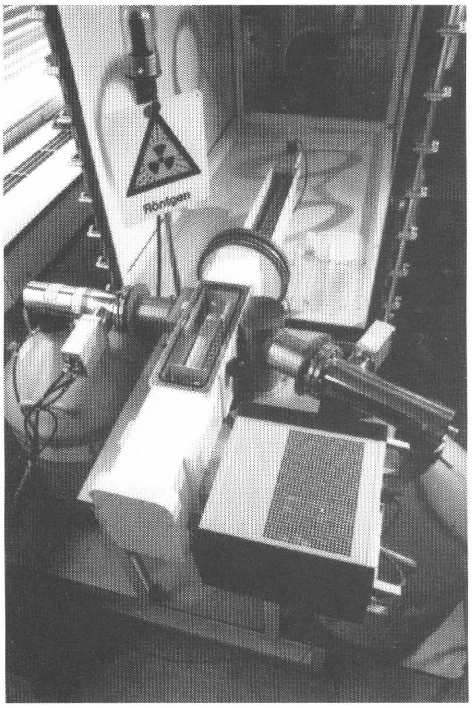

Fig. 1 : View of the KED / XRF analyzer.

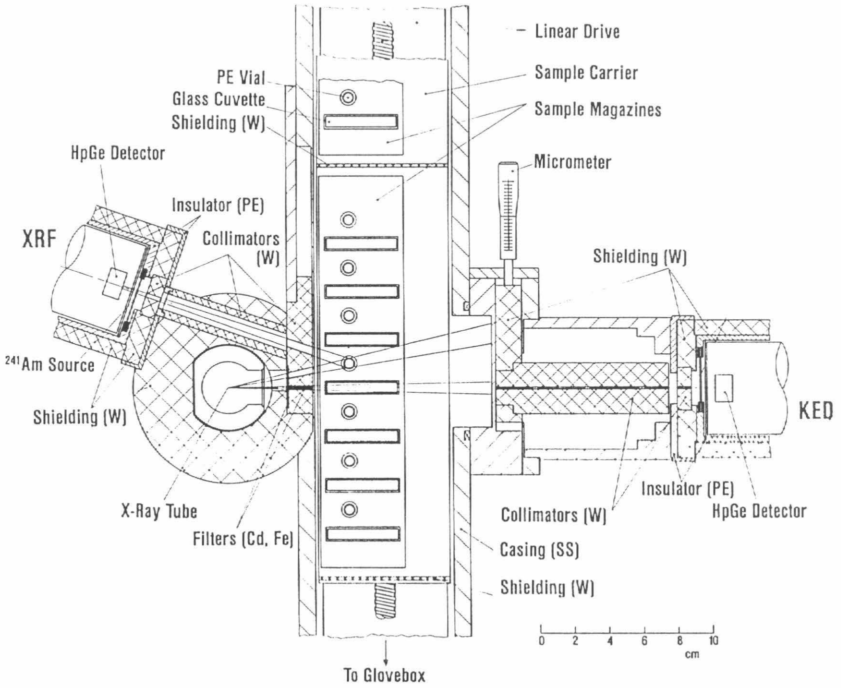

Fig. 2 : Cross-sectional plan of the source - sample - detector configuration.

Equipment

The basic equipment components include : the automated sample changer, an X-ray unit, two HpGe detectors with associated electronics for pulse processing, and a data acquisition and analysis system.

A compact 0.6 kW X-ray unit (Seifert ISOVOLT 160 H1) was chosen for the present analyzer. It provides highly-stabilized dc-voltages up to 160 kV at a fixed current of 4 mA. The standard operating voltage is 145 kV. The two HpGe detectors are of the LEGE type ($200 mm^2$ x 13 mm). Fast pulse processors based on the time-variant filtering technique /4/, which have previously been evaluated for KED measurements /5/, are used for signal processing. Data acquisition and analysis is accomplished with a VAX-based multi-user system (Genie ND 9900), which serves as central acquisition system for the different alpha and gamma counting equipments at SAL.

Modes of Operation

The analyzer provides options for different modes of operation. Depending on the type of analysis, the KED and XRF channels can be utilized for individual or for combined measurements. The desired mode of operation, and the type of analysis, is selected by the operator on a menu basis.

The existing software for automated analysis allows to run separate KED measurements for single-element samples (Th, U, Pu), and for dual-element samples (Th-U, U-Pu). The latter can also be measured in the combined KED/XRF mode. In this mode the XRF channel determines the ratio of both elements, whereas the KED measurement provides the reference concentration for the main element. Another option for separate XRF measurements on spiked Pu samples will shortly be implemented.

3. Methods of Data Evaluation

Concentration from KED

The software for KED has to evaluate the ratio of photon transmission across the K-edge from the measured spectra. The logarithm of this quantity is directly proportional to the volume concentration of the respective element. Linear least-squares fitting procedures are utilized for the evaluation of the transmission jump /6/. The fitting regions adopted for the analysis of single-element samples, and for dual elements differing by two units in atomic number, are indicated in Fig. 3. The displacements of the window limits E_- and E_+ from the edge energy, 2.36 keV below and 1.65 keV above the edge, were determined empirically to exclude those spectral regions from the fitting intervals, where distortions of the measured photon transmission due to tailing effects may occur.

The software for data analysis includes two fitting modes : 'nonextrapolated' and 'extrapolated'. In the nonextrapolated mode the ratio of photon transmission across the edge is determined at the window limits E_- and E_+ of the fitting regions, whereas in the extrapolated mode the transmission jump is evaluated directly at the edge energy E_K from extrapolations on both sides.

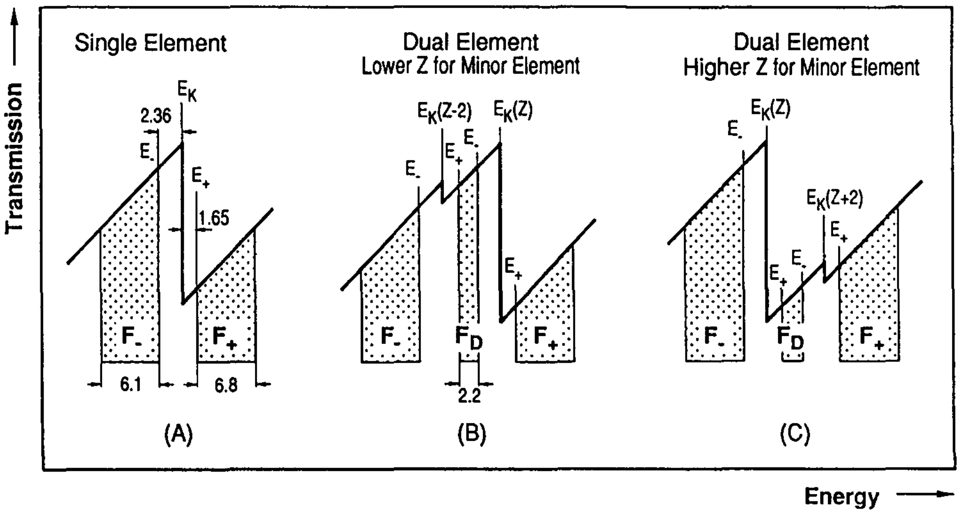

Fig. 3 : Adopted fitting regions for the evaluation of KED spectra from single-element and dual-element ($\Delta Z = 2$) samples. Energy intervals given in units of keV.

The reported result for the concentration is normally based on the nonextrapolated evaluation. This mode provides significantly better measurement precision (by a factor of 2 to 3) compared to the evaluation from extrapolations. On the other hand, the evaluation of the transmission jump at energies E_- and E_+ displaced from the edge may be slightly biased, if the sample contains high atomic-number elements other than the analyte. The extrapolated analysis, in principle, should eliminate such matrix effects. The availability of the results from both evaluations, and their comparison, provides a helpful and sensitive diagnostics tool to flag possible measurement biases associated with the composition of the sample.

Magnitude of Matrix Effects

For an assessment of the KED performance it may be useful to quantify for some practical examples the expected magnitude of systematic error introduced by matrix effects into the nonextrapolated analysis. The effects are normally negligible for matrix elements of low and medium atomic number, but should be taken into account if other actinide elements are present.

The (negative) bias for the concentration of the analyte A can be calculated from physical constants, if the concentrations ρ_i [g/cm³] of the interfering elements i are known :

$$\mathrm{Bias}[\mathrm{g/cm^3}] = \frac{1}{\Delta\mu_A} \sum_i \Delta\mu_i \cdot \rho_i \ ,$$

taking the differences of the total photon mass attenuation coefficients μ of the respective elements to

$$\Delta\mu_A = \mu_A(E_+) - \mu_A(E_-) \quad \text{and}$$

$$\Delta\mu_i = \mu_i(E_-) - \mu_i(E_+) \ .$$

The diagram in Fig. 4 indicates for some binary actinide samples the expected bias for the major element caused by a minor actinide element at different weight percentages. The respective values of $\Delta\mu_i$ lower the effect by about a factor of 3, if the atomic number of the minor element is greater than the atomic number of the analyte.

For minor element concentrations up to 1% the bias remains below 0.2% in any case. The minor elements may easily be detected and quantified for a bias correction by the XRF channel of the analyzer. An example for an XRF spectrum, taken from a Pu sample with traces of U, Np and Am at about 1%, is shown in Fig. 5. With increasing concentration of the minor element the KED measurement itself will be capable to provide an estimate for its concentration. The existing software for dual-element samples automatically corrects for the mutual bias effects. Results for binary samples will be given below.

If the atomic number of the minor element differs by only one unit from the atomic number of the analyte, the situation becomes less predictable for the existing procedure for data evaluation. This is because the absorption edge of the interfering element, then separated only by about 3 keV from the edge of the analyte, will fall into the fitting region denoted in Fig. 3 by F_D. A practical example for this situation refers to the influence of Np on the Pu assay. A recent study has shown that the existing software designed for dual-element samples with ΔZ = 2 can also handle this case very well /7/.

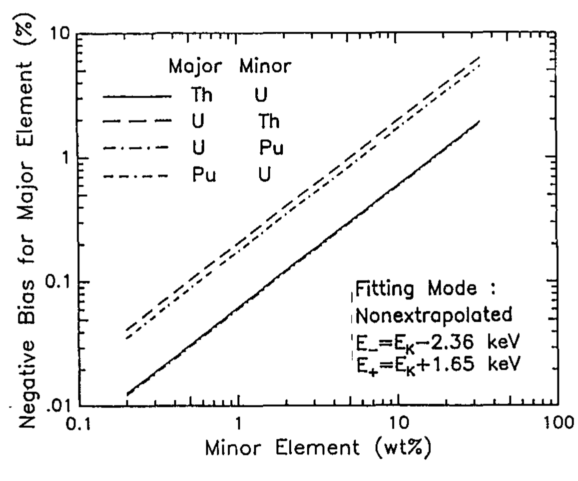

Fig. 4 : Calculated effect of minor elements on the KED result for the uncorrected non-extrapolated fitting mode.

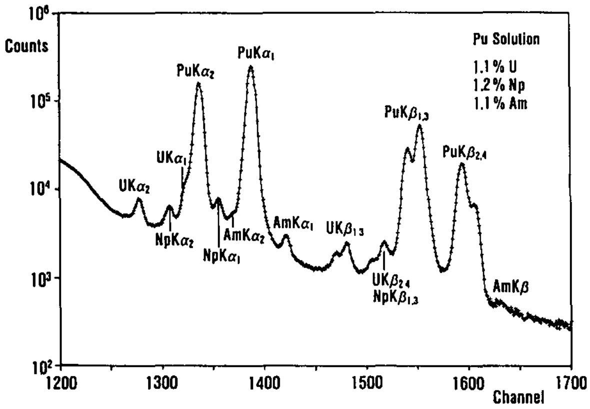

Fig. 5 : Detection of minor elements in a Pu sample by XRF.

Element Ratios from XRF

The XRF channel is utilized for the analysis of minor elements in support of the KED measurement for the main element. For this purpose it is sufficient to evaluate element ratios from the XRF spectra. They are determined from the peak areas of the respective K X-rays.

The calibration factor required for the conversion of the measured peak area ratio into the element ratio slightly depends on the heavy element concentration in the sample.. An example for this dependence, calculated from basic physical data, is shown in Fig. 6. The software for data evaluation calculates the correct calibration factor for the actual sample composition. The necessary information about the heavy element concentrations is determined to the required degree of accuracy from the XRF measurement itself, or it is obtained from the parallel KED measurement. Another correction, required for the relative detection efficiency as a function of energy, is also determined from the measured XRF spectrum. In this way the calibration for the element ratio measurement is actually reduced to the determination of a single calibration factor.

4. Performance Studies

This chapter summarizes relevant performance studies so far carried out with the K-edge/XRF analyzer at SAL. The emphasis was put on the evaluation of performance characteristics for the KED channel of the analyzer, because it provides the reference measurement for most applications. The studies carried out on a range of samples provide a fairly good overview on the overall instrument performance.

Calibration for KED

The initial set-up for KED incorporated a relatively short collimator, 1.5 cm long with a beam hole diameter of 0.3 cm, between sample and detector. Calibration runs conducted with this configuration on uranium solutions in 4 cm cells revealed an unexpectedly large nonlinearity of the instrument response (Fig. 7). Similar measurements performed on 2 cm cells did not show this behaviour.

The cause has been identified as a result of insufficient beam collimation between sample and detector, which allowed a significant fraction of the forward-peaked coherently scattered radiation reaching the detector. As a matter of fact it was found that for measurements on 4 cm cells the solid angle subtended by the collimator between sample and detector must not significantly be larger than about 10^{-4} sr in order to maintain an approximate linear calibration curve.

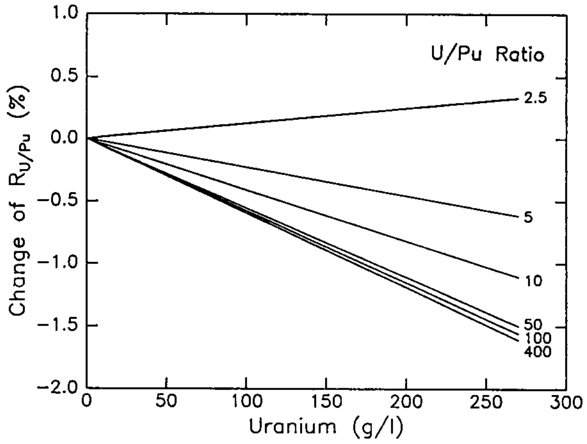

Fig. 6 : Percentage change of the XRF calibration factor for the U/Pu ratio as a function of the uranium concentration and the element ratio.

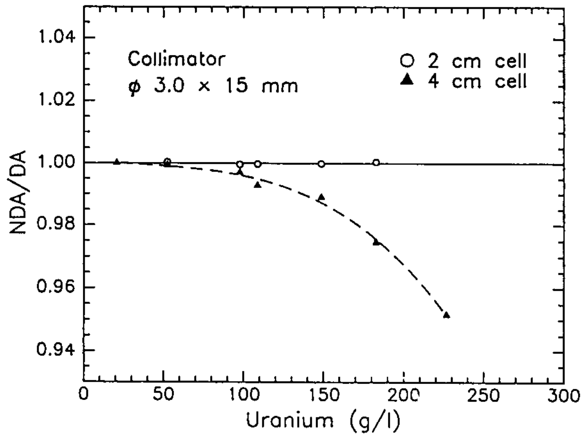

Fig. 7 : Behaviour of the KED response for medium beam collimation.

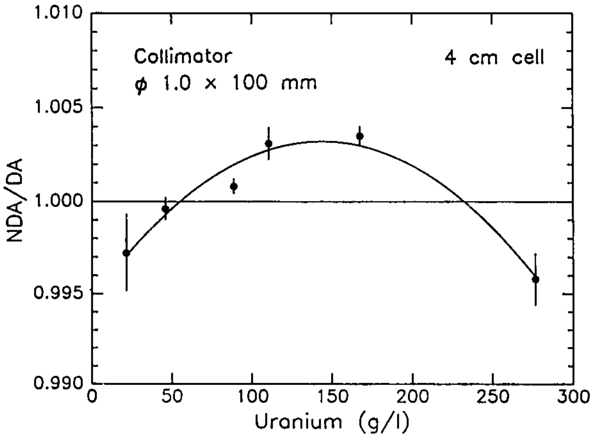

Fig. 8 : Behaviour of the KED response for tight beam collimation.

Table 1 : Calibration of KED for uranium.

2 cm Cell, $\Delta\mu$ = const		4 cm Cell, Parab. Fit to $\Delta\mu$	
DA (g/l)	KED-DA (%)	DA (g/l)	KED-DA (%)
52.32	0.034	21.48	0.016
97.62	-0.040	45.77	0.030
108.69	-0.028	88.69	-0.118
148.61	-0.013	110.46	0.033
182.83	0.044	167.45	0.053
		276.53	0.000
Stand. Dev. of Differences (%)	0.037		0.062

With tighter beam collimation (Ø 0.1 cm x 10 cm) the nonlinearity with the 4 cm cell decreased by about a factor of 10. A small residual nonlinearity of ± 0.5% remained as shown in Fig. 8. The calibration data were approximated by a parabolic fit. For a 2 cm cell the calibration curve turned out to be perfectly linear for the investigated range of concentrations (52-183 g/l). The results of the calibration runs for both cells are summarized in Table 1.

Precision for KED

The measurement precision for KED depends on the type of analysis. The precision curves in Fig. 9, experimentally established for the present system, refer to the measurement situations for single and dual-element analysis as illustrated in Fig. 3. For the optimum range of concentrations (about 80 to 150 g/l) the precision obtained for a 1000 s run varies between 0.13% and 0.18% for the indicated cases. For dual-element samples the given precision values for the major element hold for minor element weight percentages of up to 10%. Increasing beam attenuation at higher weight fractions of the minor element will slightly decrease the precision for the major element. The measurement precision is about the same for Th, U and Pu.

Uranium Solutions

A comparison between KED and DA was made for a series of uranium solution samples in order to qualify KED for the uranium analysis at SAL. The comparison measurements were done on reference materials and on dissolved UO_x powder samples. The results are summarized in Tables 2 and 3. The KED results represent averages from 5 repeat runs of 1000 s. The mean differences between KED and DA were well below 0.1% for the different sets of measurements.

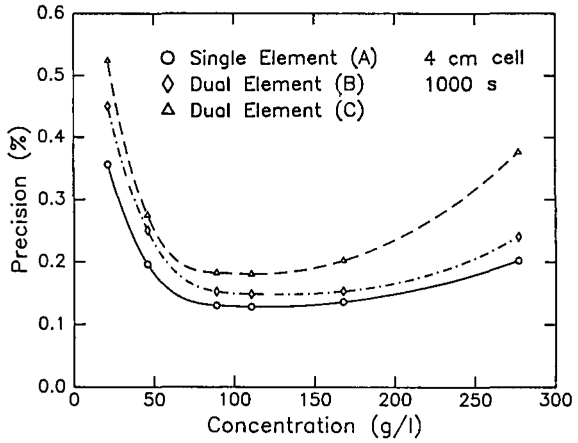

Fig. 9 : Measurement precision for KED. Cases A, B and C refer to the measurement situations illustrated in Fig. 3.

Table 2 : Differences Δ = KED-DA for uranium reference materials (2 cm cell).

Date of	Δ (%)		
Meas.	NBS 960 (40 g/l)	EC 110 (60 g/l)	NBS 960 (100 g/l)
09/27/89	0.31	0.07	0.09
09/28/89	-0.25	-0.14	-0.06
09/29/89	-0.17	0.16	-0.20
10/02/89	0.20	-0.16	0.12
10/03/89	-0.18	-0.27	-0.12
10/05/89	0.29	0.03	0.10
Mean	0.033	0.052	-0.012
SD (%)	0.260	0.163	0.134

Table 3 : Differences Δ = KED-DA for dissolved UO_x powders (2 cm cell).

DA (g U/l)	Δ (%)
84.82	0.20
84.78	0.13
87.29	0.03
87.32	0.18
87.43	-0.02
86.37	-0.08
85.71	-0.02
85.72	0.06
87.28	-0.11
87.32	-0.13
Mean	0.024
SD (%)	0.117

Th and Th-U Solutions

The performance of KED for the determination of Th was evaluated from measurements done on pure Th solutions and mixed Th-U solutions containing uranium admixtures between about 8 and 17%. The mixed samples were also analyzed by XRF for the Th/U ratio.

The KED measurements on the carefully characterized Th-U solutions offered a good opportunity to test the KED software for dual-element analysis, and to compare predicted and actually measured bias effects for the different fitting modes. The results plotted in Fig. 10 are in good agreement with the expected behaviour of the extrapolated and nonextrapolated evaluation. The slight decrease of the measured thorium concentration with increasing uranium concentration (about 0.05% per wt % uranium) as obtained from the uncorrected nonextrapolated evaluation will vanish, if the software for dual-element analysis is applied which automatically corrects for the mutual interferences. This is confirmed by the results given in Table 4. The table also lists the KED result for the minor element uranium, which is determined with a precision as indicated in Fig. 11. The standard deviation of 1.5% for the uranium results in Table 4 is in good agreement with the expected precision of about 1 to 3% for the measured range of uranium concentrations.

The measurement precision for the minor element considerably improves, if it is determined from the XRF measurement. For the given set of Th-U solutions with element ratios ranging from 5.5 to 15.5 the precision obtained for the Th/U ratio from an XRF measurement of 1000 s was 0.3% for the lower ratio, and 0.6% for the larger ratio. For these measurements, done with a tube voltage of 140 kV, the total counting rate in the XRF detector was relatively low (8 kcps). This rate, and hence the precision for the Th/U ratio, could have been increased by using a larger aperture in front of the XRF detector.

Table 4 : Differences Δ = KED-DA for Th and U in mixed Th-U solutions.

DA (g/l)		Δ (%)	
Th	U	Th	U
74.76	6.057	0.05	1.35
75.29	12.959	-0.05	1.23
78.46	6.385	-0.17	-0.87
79.32	13.539	0.13	0.52
99.48	9.122	0.19	-1.13
100.45	18.316	-0.10	0.51
101.41	18.626	0.06	-1.24
102.21	11.027	-0.36	-2.86
103.54	9.476	-0.10	0.98
105.59	9.548	-0.15	-0.93
123.62	22.142	0.19	-1.46
124.25	12.822	0.10	-3.40
124.69	12.882	0.00	0.53
126.51	21.562	0.28	0.96
Mean		0.005	-0.42
SD (%)		0.17	1.52

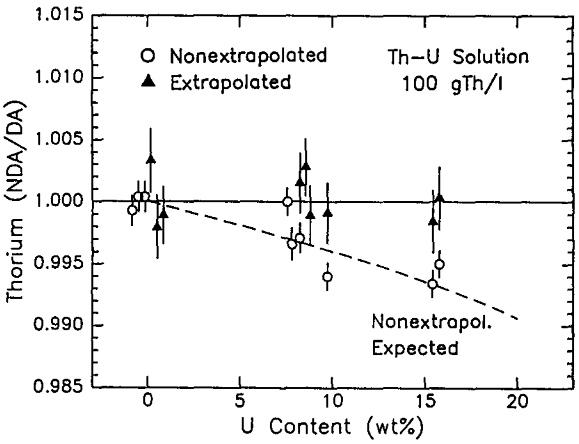

Fig. 10 : KED results for Th analysis in mixed Th-U solutions obtained from the non-extrapolated (uncorrected) and extrapolated fitting mode.

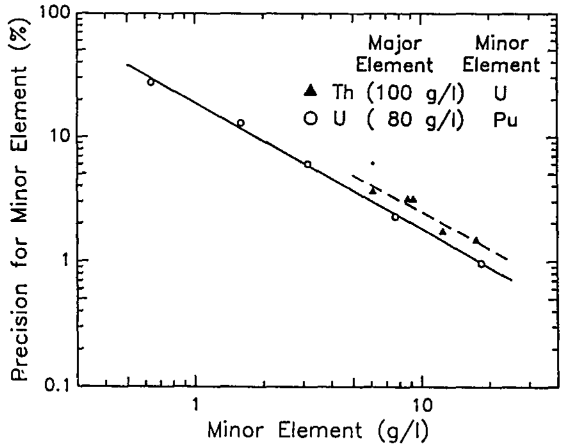

Fig. 11 : Measurement precision of KED for the minor element in dual-element samples.

U-Pu Solutions

Similar studies as for the Th-U solutions were also done on mixed U-Pu solutions. The samples used for the measurements covered a larger range of element ratios. The KED results shown in Fig. 12 include measurements on samples with U/Pu ratios ranging from 3.5 to 240.

The results for the major element uranium obtained from an uncorrected nonextrapolated evaluation agreed within less than 0.2 % with the DA values for the whole range of U/Pu ratios. Unexpectedly, they did not reveal the predicted small negative bias for lower U/Pu ratios as indicated by the dashed line in Fig. 12. On the other hand, a small positive bias was observed at the lowest U/Pu ratio for the extrapolated evaluation, which is supposed to be insensitive to matrix effects. No further studies were made on these small deviations from the expected behaviour.

The precision of the KED measurement for the minor element plutonium is plotted in Fig. 11. For improved precision it is recommended to use the XRF channel of the analyzer. The measured precision for the U/Pu ratio, based on a counting time of 1000 s, is shown in Fig. 13. For concentrated samples the measurement precision for the U/Pu ratio is about 0.7% at an element ratio of 100. This is about the measurement precision of the Hybrid Instrument for the plutonium analysis in typical reprocessing input liquors /1, 2/.

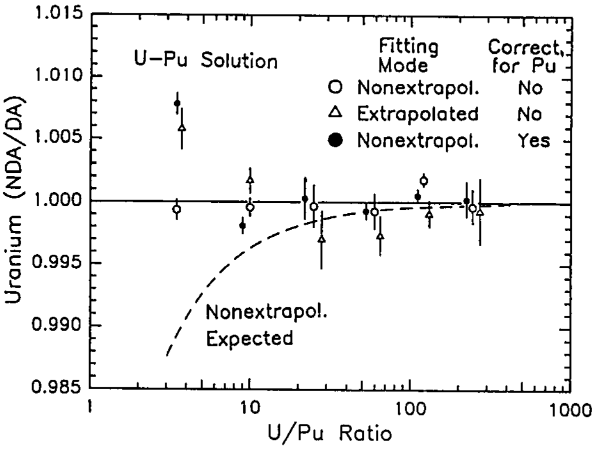

Fig. 12 : KED results for U analysis in mixed U-Pu solutions obtained from the non-extrapolated (uncorrected and corrected) and extrapolated fitting mode.

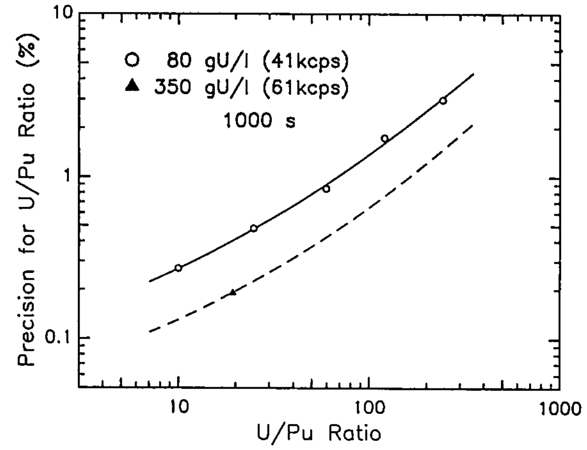

Fig. 13 : Measurement precision for the U/Pu ratio from XRF.

Mg-Size Pu Samples

Dried liquid samples containing about 4 mg of plutonium represent one type of verification samples received at SAL for analysis. The quantitative determination of plutonium in those samples, which in field are prepared from liquors by evaporation to dryness in glass vials, is usually accomplished at SAL by means of IDMS.

The design of the K-edge/XRF analyzer considered the possibility to employ its XRF channel for this analysis. For a quantitative measurement the dried samples would have to be redissolved with an internal standard, thus reducing the task of the XRF analysis to its standard application of measuring an element ratio.

The criteria for the choice of a suitable internal standard element are evident : the element should not occur in the samples to be analyzed, and its K X-ray energies should be as close as possible to those from plutonium. Thorium as a candidate has been ruled out for two reasons : First, its K_β X-rays partly interfere with the most prominent plutonium X-ray ($K_{\alpha 1}$). Second, its K_α X-rays are expected to fall into a region of strongly sloping background from inelastically scattered primary radiation, which makes a correct peak area evaluation more difficult /8/.

For this reason the use of lead as internal standard has been considered as an alternative. Some test measurements were carried out to investigate this approach. For the given excitation conditions the X-ray production rate for lead is about 5 times higher than for plutonium. Thus, working with a Pb/Pu element ratio of about 5 is usually sufficient to determine the X-ray intensity fluoresced from the internal standard with a precision of 0.1% or better. However, some difficulties were encountered in establishing reliably the overall relative detection efficiency for the Pb and Pu X-rays. The relatively large separation of the X-ray energies from both elements, and the presence of a discontinuity for the photon attenuation in between at the K-absorption edge of lead (88.0 keV), add to this problem.

From the viewpoint of the XRF measurement uranium definitely would represent the best choice as internal standard for the determination of plutonium. This option, initially ruled out because of possible interferences with uranium contaminations in the plutonium samples, is now being reconsidered.

5. Concluding Remarks

This paper has presented another example for a hybrid K-edge/XRF system designed for the measurement of nuclear materials in liquors. The principal performance characteristics of such instrumentation are now well established for a range of applications.

The system does not claim to provide the ultimate measurement accuracy nowadays achieved with carefully controlled analytical chemistry methods. But it combines a remarkable level of measurement performance for applications in its very domain with a high degree of operational and procedural simplicity. It is the combination of both features that makes an interesting tool for safeguards out of it. Up to now the scope of potential applications has not yet been exploited to its full extent.

The concept of measurement, and its practical realization, provides some flexibility for instrument installation and operation under different boundary conditions. The present paper has described a laboratory set-up coupled to a standard glovebox. In other applications such instrumentation has been adapted to heavily shielded hot cell facilities for safe measurements on highly radioactive samples.

The hybrid K-edge/XRF analyzer is built up from commercially available components. The overall instrument reliability depends, of course, on the combined reliability of those products. For the assembling of the present analyzer at SAL some latest technological products, promising a further upgrade of system performance, were selected. During the initial period of instrument operation just those components failed frequently. This kind of typical 'kinderkrankheiten' have now been mastered, but a more conservative instrument design certainly would have saved some troubles.

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STATUS OF HIGH-RESOLUTION ALPHA-PARTICLE SPECTROMETRY USING Si DETECTORS

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Abstract

The paper deals with recent work on high-resolution alpha-particle measurement and accurate analysis of complex spectra. Attention is given to a number of experimental parameters, e.g. source, detector, measurement geometry and optimization of measurement conditions. The procedure for doing accurate determination of the ^{238}Pu/($^{239+240}$Pu) activity ratio without need to analyse the fine structure, is demonstrated. Examples of analysed high-resolution spectra are given and it is shown that the alpha-particle peak shape may be considered as constant in practical energy intervals. Experimental consequences of recoil emission in case of short-lived daughters are discussed. Finally, the non-linear response of Si detectors to alpha particles is briefly mentioned.

1. Introduction

Alpha-particle spectrometry with Si detectors has become an accurate technique for analysis, certification of reference materials and decay-data measurements, especially in actinide research /1-8/. The technique has also found large application in environmental measurements because of the typical very low background. With respect to measurement accuracy, the main corrections and uncertainties result from peak tailing. The improvement of peak-fitting models and detector resolution have significantly reduced this tailing problem /9-11/.

With the introduction of ion-implanted Si detectors, improved resolution due to the thinner detector entrance window and smaller bias current (less than 10 nA), was obtained. The typical resolution of 12 keV FWHM for a 50 mm^2 Si surface-barrier detector has become almost 9 keV in the case of an ion-implanted Si detector of that size.

As a result, a number of effects which so far had been hidden, now require attention in order to exploit the improved capabilities. In addition, the gain stability of preamplifiers, in terms of a drift of the alpha-particle peak, has improved from roughly 0.3 keV/d to 0.07 keV/d. Our laboratory has been, for almost 17 years now, involved in the improvement of accuracy in alpha-particle spectrometry, especially for the measurement of emission probabilities. Much of this work was done in the frame of an IAEA Co-ordinated Research Programme, on decay data of the transactinium nuclides /12/, and via intercomparison measurements to solve problems which were identified within the International Committee for Radionuclide Metrology (ICRM).

2. Experimental Environment

A number of experimental parameters are critical in high-resolution spectrometry.

Alpha particles which are emitted from a thin layer of radioactive material suffer energy loss and straggling in the source layer and in the detector entrance window. Sources prepared by sublimation in vacuum using highly-pure radionuclides generally produce lowest straggling and peak tailing /3,13/. Source thickness could be a problem in the case of long-lived radionuclides since for a counting rate of 1 s^{-1}, a counting time of 1 day is required for 0.3 % uncertainty from Poisson statistics. For spectrometry of ^{237}Np, with a half life of 2.14 x 10^6 a, source deposits (8 mm diameter) of 76 Bq have been produced with which a peak resolution of 9.1 keV FWHM was obtained and doublet peaks of 5 keV energy separation were successfully analysed /14/.

Sublimation in vacuum is a rather expensive technique for source preparation because of the low yield of deposition and the requirement to eliminate cross contamination. For a number of analyses, quantitative sources are needed for which the technique of drop deposition with tetraethylene glycol (TEG) added as a spreading agent, is very well suited /3,13/. When using high-purity solutions, the problem of source-thickness is seldom a major one with respect to high resolution work, provided one is prepared to limit the source activity and increase the measurement time.

We have measured dead-layer thickness of 50 nm on PIPS detectors /15/ of 50 mm^2 active area (7 keV energy loss for 5.5 MeV alpha particles). Increased tailing is generally produced by border effects, e.g. the edge of the diaphragm formed by the detector housing, and the inhomogeneous electric field across the sensitive area. In some cases, peak distortion in the form of a small shoulder on the peak tailing may occur. The use of a diaphragm which covers about 1 mm of the border part of the detector eliminates the problem and improves the resolution. In order to keep the input capacitance at the preamplifier low, the latter should be directly coupled to the detector. An increase of the input capacitance by about 5 pF, due to the microdot vacuum feedthrough and BNC connector, is acceptable.

The counting geometry is an important parameter with respect to resolution and tailing. For a point source which is coaxial with the detector of unit radius, a geometry of 1 % of 4π sr is obtained at a distance of 4.92. If also the source has unit radius this distance becomes about 4.8. Little improvement in resolution and tailing can be obtained by reducing the geometry beyond 1 % of 4π sr. Peak deformation by pulse pile-up is negligible for counting rates of less than 100 s^{-1} if the electronics, especially the pole-zero adjustment is properly set.

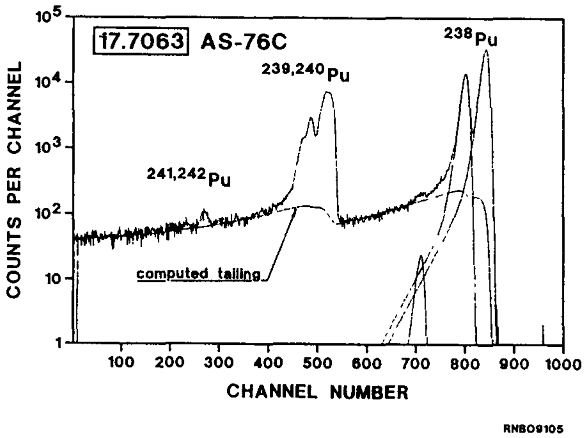

Fig. 1: Alpha-particle spectrum of a plutonium sample C used in the AS-76 interlaboratory comparison exercise, measured in June 1977. A 25 mm^2 surface-barrier detector and TEG drop-deposited source were used. The activity ratio of $^{238}Pu/(^{239+240}Pu)$ was re-analysed with the CBNM code in April 1991.

Finding out optimal amplifier and bias settings for minimum noise is very much eased by monitoring, without a source and pulser, the noise level at the output of the main amplifier using a rms voltmeter. A typical value for a 50 mm^2 detector is 1.5 mV rms. However, the bias for minimal noise does not always meet the requirement of saturation as to minimize recombination of charge carriers. Peak positions, at optimal bias with respect to noise, still vary with detector bias, depending on the detector material, from 1.5 keV/V to less than 0.1 keV/V. For this reason, the detector bias should be kept unchanged for longer periods of time to avoid the problem of poor resettability which still characterises some modern bias supplies.

Temperature effects at the detector and preamplifier can be reduced by using a thermostat. In our particular environment, the temperature effect for the alpha peak (+0.3 keV/°C) was less of a problem than it was for the pulser peak (-1 keV/°C). Alpha-particle peak drift, which is characteristic for an aging field-effect transistor, implies that, in long-term measurements, the stability of the pulser peak is not a measure for the stability of the alpha peak.

The performance of the measurement system will be badly affected in case of poor pole-zero adjustment at the main amplifier. A clamp circuit facilitates inspection of the pulse tail in the oscilloscope near the baseline in order to eliminate the slightest overshoots /16/. Alpha pulses always show a small undershoot; the pulser undershoot is larger.

The vacuum system should provide at least 0.1 Pa and be essentially free from oil vapour. In case of a faulty manipulation by which oil droplets from the rough pump may backstream into the measurement chamber and deposit on the detector or source, the resolution degrades completely. The front of a PIPS can be cautiously flushed with acetone to clean off the oil film.

The importance of isolating the vacuum system galvanically from the measurement part and the use of a single earth connection for the latter must be stressed, to avoid extensive noise.

3. Examples of Analysed Spectra

The many overlapping peaks are a characteristic feature of most alpha-particle spectra. In the course of a measurements programme on alpha-particle emission probabilities, the CBNM and CIEMAT codes /9,11/ for peak fitting have been extensively tested and intercompared /14/. The development of a code which does not require the trained eye of the spectrometrist is not for the immediate future. In the context of the present paper the performance of the CBNM code will be reviewed.

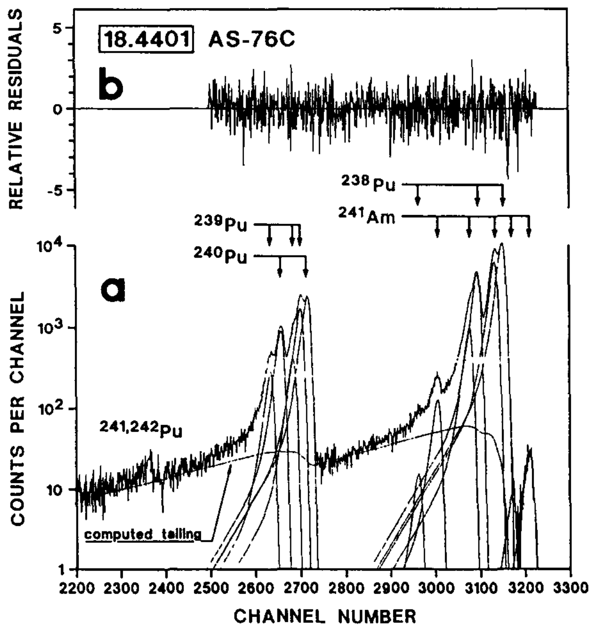

Fig. 2: The old source, used for the measurement in fig. 1, was re-measured and analysed. a: spectrum measured with a 50 mm^2 PIPS detector, showing the analysed peaks. A magnetic field was used to reduce coincidence summing of alpha particles and conversion electrons. Resolution is 12.4 keV FWHM. b: spectrum of the relative residuals.

In the period 1976 to 1978, considerable effort was spent by four laboratories to characterize plutonium samples for the AS-76 interlaboratory comparison /17/. As a result, this material became of the quality of a reference material with respect to the plutonium isotopic composition and the activity ratio R = ^{238}Pu/($^{239+240}$Pu). One of the old spectra was re-analysed for R using the CBNM code. Figure 1 shows the spectrum which was obtained from a TEG drop-deposition source, measured with a 25 mm^2 surface-barrier detector (June 1977). The code computes a tailing distribution which fits the long-range tailing. This tailing distribution is the convolution of the measured spectrum and a linear function of the exponential function. The procedure does not require information on the fine structure in the multiplets and has negligible effect on the ratio of the two groups in the spectrum. By subtracting the computed tailing, the two groups of peaks become fully separated which facilitates the calculation of their ratio. The value R = 2.976(12) from the present analysis is in full agreement with the characterization value 2.985(3). /17/. The ^{238}Pu peaks were also analysed and relative peak intensities obtained were: 0.709(1), 0.2906(10) and 0.0005(1) which, for the two major peaks, is in good agreement with the known alpha-particle emission probabilities (Pα) of this nuclide /12,18/. The standard deviations are given in brackets. A peak resolution of 14.6 keV FWHM was found for this measurement.

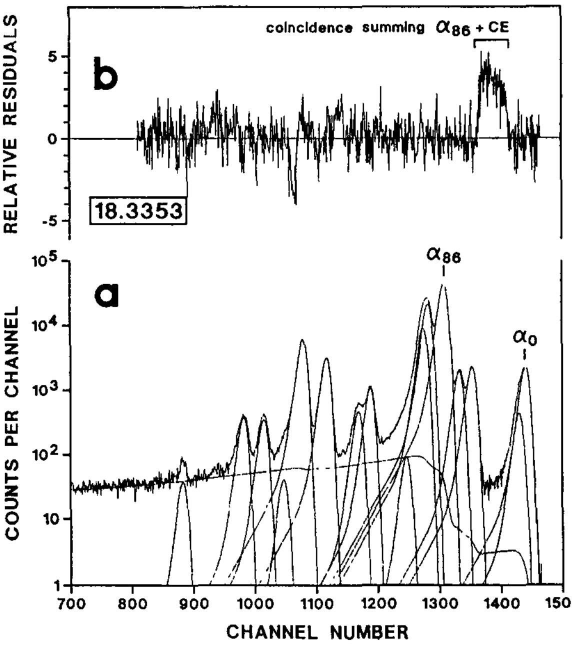

Fig. 3a: Measured alpha-particle spectrum of ^{237}Np showing the analysed peaks and the computed tailing distribution. b: Spectrum of relative residuals which shows the effect of coincidence summing produced by alpha particles and conversion electrons.

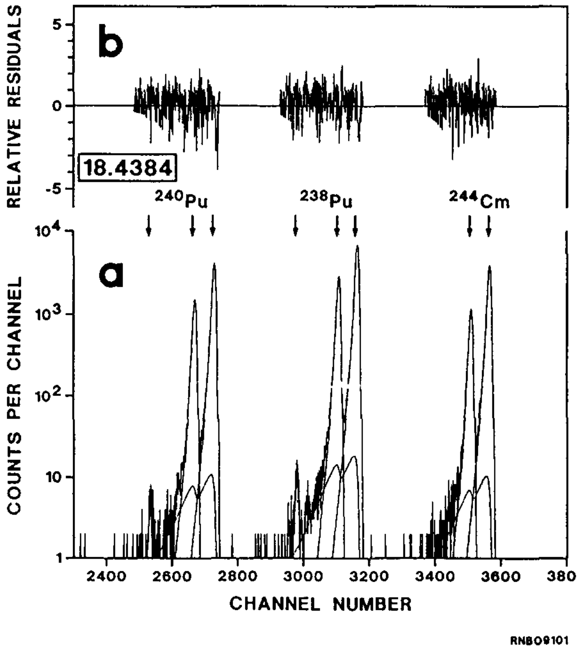

Fig. 4. a: Spectrum from a mixed source produced by sublimation in vacuum. All peaks have been analysed; a resolution of 8.6 keV FWHM was obtained in a 6 h measurement.
b: Spectrum of the relative residuals.

The old source which was used for that spectrum was re-measured using a 50 mm^2 PIPS detector, and analysed. The result is shown in fig. 2. The peak resolution of 12.4 keV FWHM this time is better than that for the spectrum of fig. 1. The fitting of the ^{238}Pu and ^{241}Am peaks provided the peak-shape parameters which were also used for the ^{239}Pu and ^{240}Pu analysis. The measured ^{241}Am/^{238}Pu activity ratio as of April 4th 1991 was 0.50(1) which has to be compared with the calculated ingrowth of 0.507 /17/. The new analysis of ^{239}Pu/^{240}Pu = 0.73(2) has to be compared with the AS-76 reference value of 0.720(1), derived for the above reference date. The spectrum of the relative residuals in fig. 2b shows the difference between the experimental spectrum (after subtraction of the computed tailing) and the fitting, relative to the standard deviation for the counts in the channel. In this case, peaks were analysed without making use of the known Pα for these nuclides. It is expected that the use of the Pα data in the fit will in general improve the result.

Another example is the analysed ^{237}Np spectrum shown in fig. 3. It was measured with a 50 mm^2 PIPS detector with an overall resolution of 9.1 keV. A particular difficulty of this spectrum were the two doublets with for both an energy difference of only 5 keV. The relative residuals (fig. 3b) reveal the effect of coincidence summing of alpha particles and conversion electrons despite the use of a magnetic field to stop conversion electrons and the 1 % geometry. Further details can be found in ref. /14/.

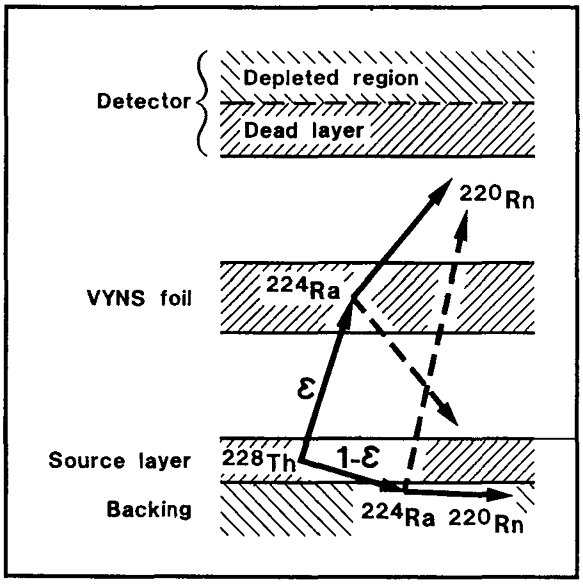

Fig. 5. To the process of recoil emission and implantation in vacuum.

The spectrum of fig. 4 shows a recent measurement of a mixed-actinides source. The measurement geometry was 0.5 % of 4π sr, duration 6 h, and the resolution was 8.6 keV FWHM. The ^{244}Cm group was first analysed and the peak-shape parameters obtained in this fit were also used for the analysis of the two other groups. The residuals in fig. 4b show that, at least in an energy range of 640 keV, the assumption of constant peak shape holds very well. Relative peak intensities obtained were: for ^{244}Cm: 0.770(5), 0.230(4); for ^{238}Pu: 0.709(3), 0.290(3), 0.00096(10); and for ^{240}Pu: 0.724(4), 0.275(4), 0.00073(13). The ^{240}Pu contained some ^{239}Pu. There is good agreement with published Pα data /12,18/.

When measuring radionuclides which produce short-lived daughters, a number of precautions and subsequent corrections have to be considered. Figure 5 shows schematically what happens when a ^{228}Th source is in the vacuum chamber. When the source is sufficiently thin, a substantial fraction ε of the recoiling ^{224}Ra ions, nearly 50 % for a vacuum sublimed source, is ejected by the source and could be implanted into the detector if not stopped by a thin foil. A VYNS foil of 15 µg/cm^2 is sufficient to stop the recoils which have an energy of about 100 keV. The other fraction, 1-ε, will produce a typical profile of implants in the source and backing. The VYNS foil is transparent to ^{220}Rn ions. Most of the radon fraction in the source and backing, however, does not leave the source.

When no VYNS is used, recoils produced in every stage of the decay chain will be implanted in the detector were the geometrical efficiency for detection of the emitted alpha particle is nearly 0.5. For modern detectors, which have a very thin dead layer, the recoils can reach the depleted region and produce a small satellite peak /19/. A consequence of the implantation profile which builds up during the measurement is that the resolutions of the alpha peaks differ for the different members of the decay chain. Furthermore, the measured "equilibrium state" in the source is changing, except for the ^{224}Ra for which the VYNS is part of the "sealed" source. When the open source is stored under atmospheric conditions the ^{224}Ra recoils are recaptured on top of the source layer.

Identification of alpha-particle emitters in the spectrum is guided by their estimated peak energies. If the energy of the unknown peak can be obtained by linear interpolation, the estimate will in general be quite reliable. However, estimates obtained by extrapolation are much less reliable since they are based on an estimation of the slope in a given point of the response curve. The fig. 6, which has been reproduced from ref. /20/, gives a magnified view of the non-linear response of the detector to alpha particles.

It can be seen that the apparent energy difference ΔE_{exp}, obtained by considering the digital offset used in the measurement and a linear system, is smaller than the energy difference ΔE_h measured by the detector. The error which results from neglecting the non-linear response is approximately 4 keV for a peak-energy difference of 650 keV. However, the error introduced by an extrapolation in case the reference peaks are too close together, and the unknown peak is at a much larger energy distance from them, may be much larger.

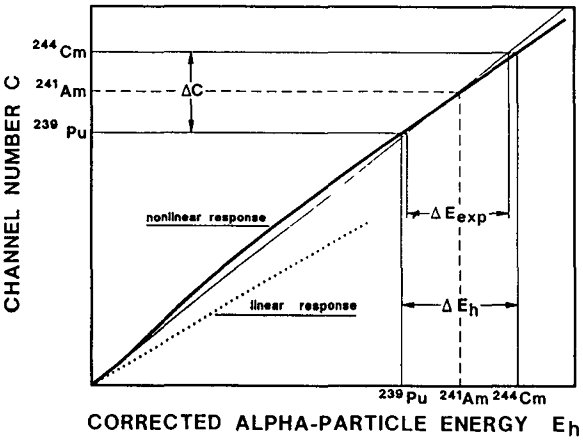

Fig. 6. Non-linear response of an ion-implanted Si detector to alpha particles. The non-linearity parameter has been arbitrarily multiplied by a factor 14 to show the effect (from ref. /20/).

4. Conclusions

Alpha-particle spectrometry has become an accurate technique for measuring decay data and for analysis. It should, however, be kept in mind that the high performance of codes for peak fitting cannot compensate for medium quality measurements. In this respect, the code cannot analyse beyond the limits of specific information available in the spectrum. One can say that what the well-trained eye is unable to reveal, the code can most often not resolve. Accurate calculation of the ^{238}Pu content is possible without need for resolving the fine structure in the spectra. Finally, the confirmation that routinely made TEG drop-deposition sources are stable with time and can be of fairly high quality for spectrometric work, is certainly an important fact.

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PREPARATION AND CERTIFICATION OF REFERENCE MATERIALS OF URANIUM ISOTOPES NITRIC SOLUTIONS

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Abstract

The report presents the results of preparation and certification of three reference materials (RM) of ^{235}U, ^{236}U, ^{238}U, and RM of ^{233}U, ^{235}U, ^{238}U to ^{236}U isotope ratios.

The certified values of the first three samples are the mass fractions of the main uranium isotopes in solutions (~10 ± 0.004 mg/g), for the last one, they are the molar ratios of isotopes ^{233}U, ^{235}U, ^{238}U to ^{236}U (~1 ± 0.0004).

The starting materials were the high enriched RM's of uranium isotopic composition in U_3O_8 form dissolved in HNO_3 (^{235}U>99.99%, ^{236}U>99.97%, ^{238}U>99.9999%).

The certification was done by potentiometric titration method and by isotope dilution mass spectrometric method (IDMS) with internal standard.

The produced RM are intended for use in mass spectrometric measurements of the mass fraction of uranium in uranium containing materials by the isotope dilution method, as well as for calibration of mass spectrometers.

1. Introduction

At present the isotope dilution mass-spectrometric method (IDMS) is one of the most widespread destructive methods for analysis of spent nuclear fuel for determination of the uranium and plutonium content in it. Correct application of the method requires reference materials (RM), the so called spikes. First of all, the RM are used for control of correctness of the analysis results, i.e. for account of systematic errors arising in different stages of analysis: chemical operations of separation, dilution, evaporation, etc. as well as in mass-spectrometric measurements due to the mass discrimination effect. RM can be used also for calibration of mass- spectrometers, especially of multicollector devices of the MAT 261 type.

Most widespread spikes are RM of uranium-233 solution - NBL 111 and uranium-235 solution - NBL 135. In the USSR a RM of the uranium-233 solution - ГСО 3213-85 - has been produced.

Recently an increased interest arise in application of double spikes. In particular, the IAEA initiated the production of RM solution enriched with ^{233}U, ^{236}U,^{242}Pu,^{244}Pu - QS 85 and QS 87 /1/.

The present work is devoted to preparation of three types of RM solution-spikes ^{235}U, ^{236}U,^{238}U and of isotopic composition RM containing ^{233}U,^{235}U, ^{236}U and ^{238}U in approximately equal amounts.

The work was done within the framework of the Soviet support programme to IAEA (task SSR-3-3) and, partly, within the scope of the research contract 5065/TC.

2. Preparation and Certification of RM Spike Solutions of ^{235}U, ^{236}U and ^{238}U

Preparation of the RM starting materials

The starting materials were the state RM's of uranium isotopic composition in U_3O_8 form dissolved in HNO_3. The certified values are given in Table 1.

Table 1

Isotope composition of uranium for starting materials (molar fraction, %)

	234	235	236	238
ГСО				
2706	0.0042	99.9942	0.0015	<0.00015
-83	±0.0001	±0.0002	±0.0001	±0.00015
4213	<0.00001	0.0043	99.9732	0.0225
-87	±0.00001	±0.0001	±0.0002	±0.0002
2681	<0.000001	0.000011	<0.000001	99.999987
-83	±0.000001	±0.000005	±0.000001	±0.000005

The solution were prepared so as to be mass fraction of uranium in the solution ~ 1%. After mixing the solutions were filled into ampoules of borosilicate glass or pyrex glass. Each ampoule was sealed off immediately after filling.

Certification analysis

Potentiometric titration of uranium was used as the main method for certification of RM. It was based on the Davies - Gray - NBL method /2/ modified to determine 5-10 mg of uraniumin a sample /3/. The titration was done by an automatic titrator Mettler DL 40 RC.

The value of the RSD didn't exceed 0.04 %. The total error included also the systematic error, connected with uncertainty of RM certification value used (for example for NBS 960 this uncertainty is equal 0.017 %). The total error of analysis result was equal 0.03 ÷ 0.04 % with an acceptable number of titrations (about 20).

Besides for analysis of RM ^{236}U and ^{238}U a mass- spectrometric isotope dilution method with internal standard (mass-spectrometer TH-5) was used.

Results of potentiometric titration

6-9 ampoules with each type of RM solution were selected for analysis. After opening up an ampoule 3 aliquots of the solution with exactly known mass (weighing with an error 0.05 mg) were taken from the ampoule. The mass of each aliquot was about 1 g (about 10 mg of uranium). This aliquots were titrated.

Simultaneously with the solution of the sample under development the solutions prepared of samples of ГСО 2396-83 and NBS 960 were titrated.

The order of titration was as follows: an aliquot of a ГСО 2396-83П or NBS 960 solution was titrated, then an aliquot of the sample being developed, then again an aliquot of ГСО 2396-83П or NBS 960, etc. Such an alternative order of titration of samples and RM, enables to reach a maximum possible identify of the conditions of measurements of the developed sample and RM. Thus the results of each measurement were calibrated with use of two different RM.

This procedure reduced the error owing to attenuation of the influence of such factors as the change of the titer of the potassium dichromate solution as a result of evaporation, the titration end point shift connected with change of temperature, pressure, reagents' composition, etc.

The mass fraction of uranium isotope in the RM solution was calculated by the formula:

$$A^{i}_{n}=\frac{1}{2}\left(\frac{Y^{RM}_{o,n-1}}{Y^{RM}_{n-1}}+\frac{Y^{RM}_{o,n+1}}{Y^{RM}_{n+1}}\right)\cdot Y_n\cdot\frac{M_i}{238.0289}\cdot\mu_i \quad (1)$$

where A^{i}_{n} is the mass fraction of uranium isotope i in the n-th aliquot, %;

$Y^{RM}_{n-1}, Y^{RM}_{n+1}$ - the results of the (n-1)-th and (n+1)-th measurements of the mass fractions of uranium in RM solutions (ГСО 2396-83П or NBS 960), %;

$Y^{RM}_{o,n-1}, Y^{RM}_{o,n+1}$- the mass fraction of uranium in RM solutions calculated proceeding from certified values and the results of preparing the solutions , %;

Y_n - the result of the n-th measurement of the mass fraction of uranium in the sample solution under development;

M_i g/mole - the molar mass of uranium in the RM under development;

238.0289 g/mole - the molar mass of uranium of natural isotope composition (ГСО 2396-83П or NBS 960);

μ_i - the mass fraction of uranium isotope i in the uranium of the RM under development. The results of analysis are presented in Table 2.

Table 2

Results of potentiometric titration (mass fraction of uranium isotope i in developed RM solution)

	Sample								
Amp.	^{235}U			^{236}U			^{238}U		
	$\bar{A}_t$	SD· 10^5	n_t	$\bar{A}_t$	SD· 10^5	n_t	$\bar{A}_t$	SD· 10^5	n_t
1	0.95341	27	2	0.98815	25	4	0.92787	17	5
2	0.95231	5	2	0.98794	24	4	0.92838	19	4
3	0.95299	41	3	0.98758	24	4	0.92810	46	5
4	0.95269	16	3	0.98825	18	5	0.92827	30	4
5	0.95260	56	3	0.98788	21	3	0.92816	30	5
6	0.95331	24	2	0.98852	41	4	0.92825	32	5
7	0.95290	17	3						
8	0.95295	32	3						
9	0.95259	29	3						
A	0.95283			0.98807			0.92816		
ε	0.00020			0.00034			0.00013		

Validation of certified values and their errors

All results were checked on distribution normality, gomogenity of variances and mean values (one way variance analysis). The mean values for all RM are taken as the certified values. For ^{235}U and ^{238}U all results were uninform. The random error in this case was calculated by the formula /4/:

$$\epsilon=t_{0.95,f}\left(\frac{1}{N(N-1)}\left(\sum_t f_t S_t^2+\sum_t n_t(\bar{A}_t-\bar{\bar{A}})^2\right)\right)^{0.5} \quad (2)$$

For ^{236}U the mean values were nonuniform, i.e. the variance between the ampoules is significantly greater than the variance inside the ampoules. The random error in this case was calculated by formula:

$$\epsilon = t_{0.95,5}\,(\sum_t n_t(\bar{A}_t - \bar{\bar{A}})^2 \,/\, 5 \sum_t n_t)^{0.5} \qquad (3)$$

The systematic error is determined by the certification errors of the applied RM - ГСО 2396-83П (0.0002%) and NBS 960 (0.00017%), and by the weighing errors in preparation of the solution. For a confidence level 0.95 it is true

$$\theta = 1.1\cdot A\,(\delta^2 m_{RM} + \delta^2 m_S + \delta^2 A_{RM})^{0.5} = = 0.00022\ \% \qquad (4)$$

where $\delta m_{RM} = 5\cdot 10^{-5}$ - the relative error of weighing the RM samples,

$\delta m_S = 5\cdot 10^{-7}$ - the relative error of weighing the RM solution,

$\delta A_{RM} = 2\cdot 10^{-4}$ - the relative error of the RM certified values.

The total error of the certified value, according to /4/ will be equal

$$\Delta = K\,(\,\theta + \epsilon\,) \qquad (5)$$

K depends on $\theta/S(\bar{\bar{A}})$ and on the value of confidence level. For P = 0.95 and $\theta/S(\bar{\bar{A}})$~2, K " 0.7.

The values of error components are given in Table 3.

Table 3

The characteristics of the RM solution

Mass fraction of isotope in the RM solution, %					
Samp.	Certified value, $\bar{\bar{A}}$	$S(\bar{\bar{A}})$	ϵ	θ	Δ
^{235}U	0.9528	0.00009	0.00020	0.00022	0.0003
^{236}U	0.9881	0.00013	0.00034	0.00021	0.0004
^{238}U	0.9282	0.00006	0.00013	0.00021	0.0003

Results of mass-spectrometric measurements

A variant of the method of isotope dilution with internal standard was used in this case. We use the next double spikes

- for the RM ^{236}U - the double spike $^{235}U/^{238}U$,
- for the RM ^{238}U - the double spikes $^{235}U/^{236}U$ and $^{233}U/^{236}U$.

The double spike solution $^{235}U/^{238}U$ was prepared by mixing of weighted solution obtained by dissolving RM NBS 960 and weighted solution of ^{235}U. The double spike $^{235}U/^{236}U$ was prepared by mixing of solutions of ^{235}U and ^{236}U (table 3). The RM ГСО 4383-88 of isotope ratio $^{233}U/^{236}U$ was used as the spike in the second case. The ratios $^{235}U/^{238}U$, $^{235}U/^{236}U$ and $^{233}U/^{236}U$ were equal approximately to one. The solutions of double spikes were added to developed RM solution and the isotope ratios in obtained mixtures were measured.

The relation between the measured and "true" isotope ratios in mass-spectrometric measurements is determined by the formula /1/:

$$R^{i/j} = R^{i/j*}\cdot(1 + \Delta\cdot F) \qquad (6)$$

were $R^{i/j}$, $R^{i/j*}$ are "true" and measured ratios of isotopes i and j,

$\Delta = M_i - M_j$ - the difference of molar masses of isotopes i and j,

F - discrimination factor per unit of the mass difference.

For the mixture $^{235}U/^{236}U/^{238}U$ (developed RM - ^{236}U, double spike -$^{235}U/^{238}U$) one may write

$$R_m^{5/6} = \frac{C_t\cdot N_t + R_n^{5/6}\cdot C_n^{6}\cdot N_n}{C_t^{6}\cdot N_t + C_n^{6}\cdot N_n} = R_m^{5/6*}\cdot(1 - F) \qquad (7)$$

$$R_m^{8/6} = \frac{C_t^{8}\cdot N_t + R_n^{8/6}\cdot C_n^{6}\cdot N_n}{C_t^{6}\cdot N_t + C_n^{6}\cdot N_n} = R_m^{8/6*}\cdot(1 + 2F) \qquad (8)$$

where C_t^i - the concentration of isotope i in double spike solution, mol/g,

C_n^6 - the concentration of uranium-236 in the solution of the RM under development, mol/g,

N_t, N_n- the mass of the solutions of double spike and RM. The index m refers to the mixture, n - to the RM, t - to the double spike.

Excluding F from (7) and (8), we obtain an equation with one unknown C_n^6:

$$C_n^{6} = \frac{N_t(2C_t^{5}/R_m^{5/6*} + C_t^{8}/R_m^{8/6*} - 3C_t^{6})}{N_n(3-2R_n^{5/6}/R_m^{5/6*} - R_n^{8/6}/R_m^{8/6*})}\cdot\frac{100}{236.0456} \qquad (9)$$

Here $C_n^6$is expressed in %.

For RM solution of ^{238}U analysed with use of the double spikes $^{235}U/^{236}U$ and $^{233}U/^{236}U$ the formulas looks as

$$C_n^{8} = \frac{N_t(2C_t^{5}/R_m^{5/6*} + C_t^{8}/R_m^{8/6*} - 3C_t^{6})}{N_n(3/R_m^{6/8*} - 2R_n^{5/8}/R_m^{5/6*} - 1/R_m^{8/6*})}\cdot\frac{100}{238.0508} \qquad (10)$$

$$C_n^{8} = \frac{N_t^{6}\,(5C_t^{6} + 3C_t^{8}/R_m^{8/6*} - 2C_t^{3}/R_m^{3/6*})}{N_n\,(3/R_m^{8/6*} + 2R_n^{3/8}/R_m^{3/6*} - 5R_n^{6/8})} \cdot \frac{100}{238.0508} \qquad (11)$$

The characteristics of double spikes C_t^{i} and isotope composition of developed RM's $R_n^{i/j}$ (table 1) are known. After measurement of the ratios in mixtures $R_m^{i/j*}$ one may find the mass fractious of corresponding isotopes in developed RM solutions (table 4).

Table 4

Results of IDMS analysis
(mass fraction of isotope in RM solution, %)

Ampoule	1	3	4	5	Mean
$C_n^{6}(5/8)$	0.98839	0.98742		0.98841	0.98807
SD	0.00025	0.00008		0.00011	0.0006
$C_n^{8}(5/6)$	0.92858	0.92779	0.92842		0.92826
SD	0.0003	0.0004	0.0007		0.0004
$C_n^{8}(3/6)$	0.92775	0.92767	0.92760		0.92767
SD	0.00014	0.00020	0.0003		0.00008

Taking into account a systematic error connected with the error of certified values of double spikes we can see nonsignificant differences between results of potentiometric and mass-spectrometric measurements (Tables 3, 4).

The developed RM solutions has been registered as

^{235}U - ГСО 4343-88
^{236}U - ОСО 95.048-88
^{238}U - ОСО 95.057-89

3. Preparation and Certification of RM of Ratios $^{233}U/^{236}U$, $^{235}U/^{236}U$, $^{238}U/^{236}U$

Preparation of the starting material

RM of uranium isotope composition (ГСО 2702-83) in the form of U_3O_8 and RM of isotope ratio $^{233}U/^{236}U$ and the mass fraction of uranium-233 (ГСО 4383-88) were used as the starting materials. The certified values of these RM are given in Table 5.

The mass fraction of isotopes in the ГСО 4383-88 solution are

^{233}U - (0.09917 ± 0.00007) %,
^{236}U - (0.09873 ± 0.00007) %,
U - (0.20080 ± 0.00016) %.

Table 5

Certified values of uranium isotope composition (molar fraction, %) and the molar ratios of uranium isitopes in RM

Isotope	ГСО 2702-83	ГСО 4383-88
233	-	49.703
234	0.367 ±0.002	1.3549
235	49.588 ±0.012	0.0482
236	0.4569 ±0.0005	48.853
238	49.588 ±0.012	0.0415
235/238	1.0000 ±0.0005	-
233/236	-	1.0174 ±0.0006

U_3O_8 (ГСО 2702-83) was dissolved in HNO_3. The exactly known mass of this solution was mixed with exactly known mass of ^{233}U (ГСО 4383-88) solution. After thorough mixing solution was packed into glass ampoules per 1 ml of the solution in each.

Certification analysis

Seven ampoules of the RM solution were selected for analysis. Mass-spectrometric measurements were carried out with TH-5 mass spectrometer, getting measured isotope ratios in the mixture.

Treatment of results.
Validation of the certified values

For evaluation of the certified characteristics - molar ratios of isotopes $^{233}U/^{236}U$, $^{235}U/^{236}U$, $^{238}U/^{236}U$ in the mixture the calculation method described in /1/ was used. In this case ГСО 4383-88 was considered as sample with known isotope composition, and ГСО 2702-83, as RM - internal standard. It is possible to write on the basis of the material balance equation /1, 5/:

$$\frac{N_n^{6}}{N_t^{6}} = \frac{R_m^{3/6} - R_t^{3/6}}{R_n^{3/6} - R_m^{3/6}} = \frac{R_m^{5/6} - R_t^{5/6}}{R_n^{5/6} - R_m^{5/6}} = \frac{R_m^{8/6} - R_t^{8/6}}{R_n^{8/6} - R_m^{8/6}} \qquad (12)$$

where N_n^6, N_t^6 - the numbers of moles of uranium-236 in the sample and the RM,

$R_n^{i/j}$, $R_t^{i/j}$, $R_m^{i/j}$ - the isotope ratios in the sample, the RM and the mixture.

The relation between the "true" $R^{i/j}$ and the measured $R^{i/j*}$ratios is given by the expressions, for instance, for $R^{i/6}$:

$$R_n^{i/6} = R_n^{i/6*} \quad (1 + \Delta_{i/6} F_n) \qquad (13)$$

$$R_m^{i/6} = R_m^{i/6*} \quad (1 + \Delta_{i/6} F_m) \qquad (14)$$

were Δ- the difference between the isotope mass numbers of the denominator and the numerator,

F_n, F_m - the discrimination coefficients per a unit of mass difference for the sample and the mixture.

Substituting (13) and (14) into (12) we obtain a system of 3 linear equations with 3 unknowns N_n^6/N_t^6, F_n, F_m solved which find $R_m^{i/6}$. The calculation procedure is described in detail in /5/. The results of the calculation, the corrected ("true") isotope ratios in the mixture are given in Table 6.

Estimation of the certified values errors

The random error was estimated on the base of the results of certification analysis (Table 6) and formulas (2) and (3). The data were checked against the hypotheses about uniformity of variances and mean values. The confidence limits of the random errors for isotope ratios in the mixture were

(233/236) = 0.00018,
(234/236) = 0.00008,
(235/236) = 0.00012,
(238/236) = 0.00012.

The systematic error in this case was caused by the errors of the certified characteristics of ГСО 2702-83, the mixture's component used as the internal standard in the certification calculations. The systematic error was estimated in the way described in /5/. It was supposed that the molar fractions of isotopes in ГСО 2702-83 were random values having normal distribution with mean values indicated in the certificate (Table 5) and SD equal to the errors of the certified molar fractions. Drawing by means of a random number generator the values of the isotopes' molar fractions independent of one another, thus we obtained a "set" of reference materials (internal standards). The whole "set" of internal standards (about 200 values) was used in calculation for the sample (ГСО 4383-88) and the mixture with present isotope compositions.

Table 6

Results of analysis of the mixture

	Measured isotope ratios in the mixture			
Amp.	233/236	234/236	235/236	238/236
1	1.0094	0.03462	0.95645	0.95576
2	1.0095	0.03473	0.95639	0.95525
3	1.0107	0.03460	0.95687	0.95444
4	1.0105	0.03459	0.95653	0.95474
5	1.0096	0.03472	0.95619	0.95523
6	1.0090	0.03462	0.95659	0.95563
7	1.0081	0.03460	0.95588	0.95577
Mean	1.0095	0.03464	0.95641	0.95526
SD	0.0009	0.00006	0.0003	0.0005
	Corrected isotope ratios in the mixture			
Amp.	233/236	234/236	235/236	238/236
1	1.0088	0.03460	0.95626	0.95613
2	1.0084	0.03472	0.95606	0.95592
3	1.0083	0.03454	0.95610	0.95597
4	1.0088	0.03455	0.95598	0.95584
5	1.0087	0.03470	0.95591	0.95578
6	1.0082	0.03460	0.95632	0.95618
7	1.0082	0.03460	0.95589	0.95575
Mean	1.0085	0.03462	0.95607	0.95593
SD	0.0003	0.00007	0.00017	0.00017

As the result of the calculation a "set" of values $R_m^{i/6}$ for the mixture was received, the number of the values $R_m^{i/6}$ being equal to the number of used internal standards.

The values of SD calculated for the obtained $R_m^{i/6}$ were considered as the estimations of the systematic errors of the isotope ratios in the mixture. They were

θ(233/236) = 0.00033,
θ(234/236) = 0.00019,
θ(235/236) = 0.00020,
θ(238/236) = 0.00020.

The total errors of the isotope ratios were calculated by means of formula (5).

Table 7

Certified values of RM of isotope ratios

	$^{233}U/^{236}U$	$^{234}U/^{236}U$	$^{235}U/^{236}U$	$^{238}U/^{236}U$
Molar ratio	1.0085	0.03462	0.95607	0.95593
Total error	0.0004	0.00021	0.00023	0.00023

The certified values of the molar ratios $^{233}U/^{236}U$, $^{234}U/^{236}U$, $^{235}U/^{236}U$, $^{238}U/^{236}U$, and their total errors for a probability 0.95 are given in Table 7.

The RM of isotope ratios has been registered as a branch RM OCO 95.060-89.

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ELIMINATION DES CHLORURES DANS LA METHODE DE DOSAGE PRECIS DE L'URANIUM OU DU PLUTONIUM, UTILISANT LES IONS TITANEUX COMME REDUCTEUR

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Abstract

The Corpel and Regnaud's procedure for the precise determination of uranium and plutonium, using titanous (III) chloride as reductant has been modified in order to be compatible with the throwing out standards in nuclear plants.

The removal of chloride reagents has been studied. On the original method, there are two : titanous chloride and ferric chloride.

We propose titanous sulphate and ferric nitrate as substitution reagents.

As commercial titanous sulphate can't be found, an easy procedure has been set and described with storage conditions: experimental conditions have been optimized and adapted for manufacturing on a laboratory scale.

1. Introduction

L'enjeu économique, le respect de l'environnement et la sécurité en entreprise rendent de plus en plus nécessaire un contrôle analytique précis des procédés industriels.

Par les risques supplémentaires qui lui sont propres (sûreté, criticité), l'industrie nucléaire est sans doute parmi les plus exigeantes dans ce domaine. Ainsi, les matières nucléaires sont l'objet de contrôles nationaux et internationaux, assurant la validité des mesures de l'exploitant. Ces analyses sont obtenues par des méthodes physiques, de plus en plus utilisées aujourd'hui, mais aussi chimiques qui, comme le montre cet article, présentent toujours un intérêt certain. Au CEN/FAR, le Laboratoire de Métrologie des Matières Nucléaires (LAMMAN) dont les compétences dans le domaine de l'analyse précise sont reconnues, utilise de façon courante des méthodes chimiques pour les dosages précis de l'uranium et du plutonium. Pour adapter ces méthodes à l'évolution des normes dans les installations nucléaires, de nouvelles procédures de mise en oeuvre de ces méthodes ont été mises au point ; c'est le cas de la méthode d'analyse précise de l'uranium et du plutonium, utilisant les ions titaneux comme réducteur, dans laquelle l'utilisation des ions chlorure a été éliminée.

2. Intérêt de l'analyse chimique

Prenons l'exemple des installations de retraitement : tout le long du procédé, des contrôles analytiques destinés à en vérifier le bon fonctionnement sont installés.

Toutes ces analyses sont évidemment coûteuses et la tendance actuelle est de développer des méthodes non-destructives utilisables en contrôle en ligne, celui-ci présentant le double avantage d'être automatisé et de ne pas produire d'effluents.

Cependant, la détection de dérive est difficile et la précision est souvent moins bonne que celle des méthodes chimiques concurrentes ; d'ailleurs, les méthodes physiques sont en général étalonnées au moyen de références préparées par chimie.

En pratique, les méthodes destructives sont encore les méthodes de choix pour les mesures liées aux bilans.

En effet, à ce stade du procédé, l'objectif est de comptabiliser le plus précisément possible la masse de matière. Celle-ci est estimée à partir de la relation :

$$\boxed{M = C \times \rho \times V}$$

où :
C est la concentration massique de la solution,
ρ est la masse volumique de la solution,
V est le volume de solution contenu dans la cuve.

A titre d'exemple, une incertitude de 1 % sur une tonne de matière correspond à 10 kg, ce qui est inacceptable. Les performances actuelles sont de l'ordre de 0,1 % en valeur relative, soit 1 kg pour 1 tonne /1/ et /2/.

L'objectif visé pour l'avenir est d'obtenir une incertitude relative de 0,01 %, soit 100 g pour 1 tonne.

Il est clair que pour obtenir la meilleure précision sur la masse, chaque facteur doit être minimisé. Les performances actuelles de chacun d'entre-eux sont de l'ordre de :

- 5.10^{-2} % pour la mesure de la masse volumique et du volume,
- 5.10^{-2} % à 10^{-1} % pour la mesure de la concentration selon les conditions opératoires.

La substitution de ces réactifs pouvant entraîner une diminution des performances des analyses précises, nous avons étudié leur effet sur les résultats de dosage, en terme de reproductibilité.

Remplacement du chlorure titaneux

La majeure partie de l'étude a consisté à optimiser le mode opératoire de préparation de sulfate titaneux. L'objectif est d'obtenir une solution de titane (III) suffisamment concentrée, de concentration équivalente au chlorure titaneux, c'est-à-dire environ molaire, et suffisamment stable dans le temps, tout en s'assurant que les modifications apportées n'ont pas une influence néfaste sur les performances du dosage :

- blanc faible et reproductif,
- dosage le plus exact et reproductible possibles.

Le principe de préparation consiste en une oxydation ménagée de titane métal par de l'acide sulfurique 3M.

Les différents paramètres mis en jeu dans cette dissolution oxydante ont été testés :

- température et temps d'attaque,
- concentration en acide sulfurique,
- rapport "masse titane/ volume d'acide".

L'exploitation des résultats par une analyse de variance montre que les conditions de mise en solution n'influencent pas la qualité des dosages : exactitude et reproductibilité sont comparables, voire meilleures que celles obtenues avec $TiCl_3$.

Ainsi, dans le cas du dosage de l'uranium :

- l'intervalle de confiance du blanc de réactifs est (0,1350 ± 0,0036)g de solution cérique, alors que pour les dosages d'uranium, on utilise environ 50 g de Ce(IV) ; le blanc n'entraîne pas d'incertitude notable sur le dosage ;
- la reproductibilité du dosage de l'uranium est de l'ordre de 0,1 % en valeur relative pour 4 à 5 essais.

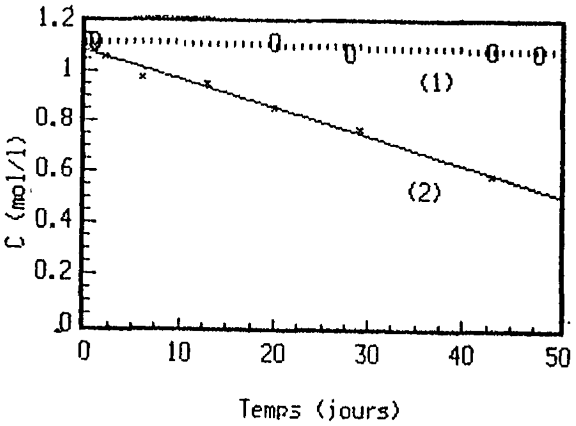
Néanmoins, pour obtenir une solution d'ions titaneux suffisamment concentrée (~ 1 $mol.l^{-1}$) tout en disposant des quantités de réactifs nécessaires à des dosages de routine (par exemple 250 ml), les conditions de mise en solution sont plus restrictives.

Un mode opératoire, facile à mettre en oeuvre, a été défini pour préparer des lots de 250 ml de solution de Ti(III) de capacité réductrice suffisante.

Il consiste en une attaque à chaud, en milieu sulfurique 3M, de titane métal de grande pureté (99,98 %) ; on procède en deux paliers de température : le premier palier, à faible température (~ 50°C), permet de déliter l'éponge de titane et facilite ainsi la mise en solution durant le deuxième palier à température plus élevée (~ 80°C).

Stockage des solutions d'ions titaneux

L'étude de la variation de la capacité réductrice en fonction du temps, dans différentes conditions de conditionnement, montre qu'il est souhaitable de fractionner la solution de Ti(III), une fois filtrée, en flacons de 25 ou 50 ml (selon les besoins du laboratoire), bien remplis et protégés de la lumière (cf. figure 1).



(1) Solution conservée à l'abri de l'air (c'est-à-dire dans un flacon bien rempli)
(2) Solution conservée sans précautions particulières (c'est-à-dire dans un flacon peu rempli)

Figure 1 : Variation de la capacité réductrice de solution de Ti(III) conservées dans différentes conditions (1) et (2), en fonction du temps.

Remplacement du chlorure ferrique par le nitrate ferrique

Une étude systématique a été menée dans le cas du dosage de l'uranium. Elle montre que le remplacement du chlorure ferrique par du nitrate ferrique de concentration équivalente (soit 1,7 $mol.l^{-1}$ environ) n'altère ni la justesse, ni la reproductibilité des dosages (cf. tableaux 1 et 2).

	Moyenne* (g)	Ecart-type** (g)
$FeCl_3$	0.2185	0.0106
$Fe(NO_3)_3$	0.2025	0.0039

* Masse moyenne, sur 5 mesures, de solution cérique consommée pour le dosage du blanc
** Ecart-type correspondant

Tableau 1 : Comparaison du blanc de réactif obtenu en utilisant soit du chlorure ferrique, soit du nitrate ferrique

Cette remarque montre qu'il est nécessaire de disposer de mesures les plus précises possibles pour la concentration massique, le volume et la masse volumique.

3. Le Laboratoire de Métrologie

En réponse à ces besoins, le CEA a créé une structure : le Laboratoire de Métrologie des Matières Nucléaires (LAMMAN), dont quelques unes des réalisations marquantes sont résumées ici :

- préparation et conditionnement en ampoules de verre scellées d'échantillons de référence pour des circuits interlaboratoires, à la demande de la CETAMA (circuit EQRAIN),
- préparation et conditionnement de solutions de référence "secondaires", pour étalonner des méthodes d'analyse (réalisations pour l'IPSN, EURATOM, ...),
- participation à la certification de matériau de référence dans le cadre de la CETAMA,
- mesures de volumes de cuves et de masses volumiques pour COGEMA, en collaboration avec l'IPSN.

Evidemment, l'amélioration des performances des méthodes est un souci permanent.

Les principales caractéristiques qui rendent compte des performances des analyses précises sont :

- la justesse,
- la reproductibilité.

sachant que l'objectif final est de déterminer la concentration massique à mieux que 0,1 % en valeur relative.

Pour maintenir, voire améliorer, les performances d'une méthode analytique, ces deux facteurs doivent être maîtrisés. Comment cela est-il réalisé en pratique ?

Une des premières conditions est d'utiliser une méthode éprouvée. Tout particulièrement pour maîtriser le critère de reproductibilité, il faut travailler toujours dans les mêmes conditions parfaitement définies, avec des opérateurs bien formés et soucieux des précautions à suivre.

En ce qui concerne le critère de justesse, il faut utiliser une méthode de traçabilité démontrée, c'est-à-dire qui se réfère à des étalons primaires élaborés par des laboratoires reconnus /3/.

4. Adaptation de la méthode d'analyse précise

Principe de la méthode

Nous allons maintenant illustrer ces idées par l'exemple d'une des principales méthodes d'analyse précise de l'uranium et du plutonium, utilisée dans notre laboratoire. C'est la méthode Corpel et Regnaud, dite "méthode au titane", qui met en jeu le couple rédox Pu(III)/Pu(IV) et U(IV)/U(VI) /4/ et /5/.

Le principe de la méthode est résumé ici dans l'exemple de l'uranium. Les étapes suivantes sont successivement mises en jeu :

- réduction de l'uranium au degré d'oxydation + IV, en milieu sulfonitrique, par du chlorure titaneux ajouté en excès, et destruction de l'excès de Ti(III) par les ions nitrate,
- oxydation de l'U(IV) par les ions ferriques et dosage des ions ferreux produits,
- titrage au moyen d'une solution cérique étalonnée ; le point équivalent est détecté photométriquement par décoloration de l'orthophénanthroline ferreuse.

Les permormances de la méthode, sont les suivantes :

- sur une moyenne de 5 mesures, l'intervalle de confiance de la concentration massique de la solution considérée est déterminé à mieux que 0,1 % en valeur relative (probabilité : 0,95), les prises d'essais étant comprises entre 40 et 80 mg de matière,
- sur des séries plus importantes de mesures, on peut obtenir des résultats à quelques 10^{-2} %.

L'inconvénient de cette méthode est qu'elle produit des effluents chlorés qui proviennent de certains réactifs utilisés, à savoir le chlorure titaneux et le chlorure ferrique, ce qui la rend incompatible avec les normes de rejets des installations nucléaires.

En outre, la qualité des lots commerciaux de chlorure titaneux est apparue moins satisfaisante qu'auparavant : en particulier, la reproductibilité des "blancs" de réactifs s'est dégradée, ce qui altère les performances de la méthode.

Afin de rendre cette méthode compatible avec les normes de rejets des installations nucléaires vis-à-vis des ions chlorure, nous avons étudié le remplacement de ces réactifs chlorés.

	Moyenne* (g)	Ecart-type** (g)
$FeCl_3$	5.291	0.0096
$Fe(NO_3)_3$	5.2907	0.0072

* Masse moyenne, sur 5 mesures, de solution cérique consommée pour le dosage du blanc.
** Ecart-type correspondant.

Tableau 2 : Comparaison du dosage d'une solution de référence d'uranium, de concentration massique égale ((5,29 ± 0,01) $g.kg^{-1}$, en utilisant soit du chlorure, soit du nitrate ferrique

5. Conclusion

L'élimination des ions chlorure dans la méthode de dosage précis du plutonium ou de l'uranium est une avancée importante puisqu'elle permet de minimiser les problèmes induits par la gestion ultérieure des effluents, sans en altérer les performances.

Le mode opératoire de préparation de sulfate titaneux est simple à mettre en oeuvre et adapté à la préparation des quantités nécessaires aux dosages de routine.

Cette méthode de dosage est à préconiser en usine pour les analyse de bilan des produits finis (PuO_2 et nitrate d'uranyle), en remplacement de la méthode CuCl (pour le PuO_2) qui est responsable à l'heure actuelle de 20 à 25 % des effluents chlorés des usines.

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DEVELOPMENT OF ISOTOPE DILUTION GAMMA-RAY SPECTROMETRY FOR PLUTONIUM ANALYSIS*

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Abstract

We are studying the feasibility of determining the plutonium concentration and isotopic distribution of highly radioactive, spent-fuel dissolver solutions by employing high-resolution gamma-ray spectrometry. The study involves gamma-ray plutonium isotopic analysis for both dissolver and spiked dissolver solution samples, after plutonium is eluted through an ion-exchange column and absorbed in a small resin bead bag. The spike is well characterized, dry plutonium containing ~98% of ^{239}Pu. By using measured isotopic information, the concentration of elemental plutonium in the dissolver solution can be determined. Both the plutonium concentration and the isotopic composition of the dissolver solution obtained from this study agree well with values obtained by traditional isotope dilution mass spectrometry (IDMS). Because it is rapid, easy to operate and maintain, and costs less, this new technique could be an alternative method to IDMS for input accountability and verification measurements in reprocessing plants.

1. Introduction

Isotope dilution mass spectrometry (IDMS) has long been the most accepted technique for determining the plutonium content of input spent-fuel dissolver solutions in reprocessing plants./1/ However, IDMS is time consuming, sample preparation is lengthy, and the equipment and operation are costly. Recently, a hybrid K-edge and K x-ray fluorescence (XRF) densitometer for determining the uranium and plutonium element concentrations of dissolver solutions was developed at Kernforschungszentrum Karlsruhe./2/ However, the hybrid instrument does not measure isotopic compositions. To avoid IDMS's disadvantages, but yet deliver acceptable measurement accuracy and precision, and to complement the hybrid K-edge/K-XRF measurement of plutonium concentration, we are developing a new technique—isotope dilution gamma-ray spectrometry (IDGS)—for simultaneously measuring the plutonium concentration and isotopic composition of highly radioactive fuel-dissolver solutions. IDGS is similar to IDMS except that the isotopic distributions of both unspiked (unknown dissolver solution) and spiked (by adding to the dissolver solution a spike of well-characterized plutonium) samples are measured by high-resolution gamma-ray spectrometry rather than mass spectrometry, and that sample preparation is simpler for IDGS. Gamma-ray measurements of highly radioactive dissolver solutions from reprocessing plants require rapid and efficient separation of plutonium from fission products and other actinides. A two step ion-exchange separation was developed to obtain satisfactory purification and recovery of plutonium for the IDGS measurement. Spectral analysis for the required full-energy peak areas and isotope ratios is accomplished by well-established methods. The isotopes ^{236}Pu, ^{238}Pu, ^{239}Pu, and ^{240}Pu are all good candidates as a known spike for the IDGS technique. However, for reasons of cost and availability, ^{239}Pu is the best choice. Two proof-of-principle experiments have been carried out at the Tokai Reprocessing Plant. We used large-size dry (LSD) spikes of ^{239}Pu (97.9%), prepared by the International Atomic Energy Agency's (IAEA) Safeguards Analytical Laboratory (SAL), for our experiments./3/ Their certified isotopic compositions (in weight percent) are listed in Table I.

This paper describes IDGS measurement principles and the preparation of the resin bead sample, and discusses plutonium element concentrations and isotopic compositions of dissolver solutions obtained by IDGS.

2. Measurement Principles

Plutonium Isotopic Composition

The measurement method of plutonium isotopic ratios is based on high-resolution, gamma-ray spectrometry techniques. In general, the atom ratio $N(m)/N(n)$ of two isotopes m and n can be determined by measuring their respective gamma rays a and b.

$$R = \frac{N(m)}{N(n)} = \frac{A(a)}{A(b)} \cdot \frac{I(b)}{I(a)} \cdot \frac{T_{1/2}(m)}{T_{1/2}(n)} \cdot \frac{\varepsilon(b)}{\varepsilon(a)} , \qquad (1)$$

where A = full-energy peak areas,
I = absolute emission probability of gamma rays,
$T_{1/2}$ = half-life of isotope, and
ε = relative efficiency of selected gamma rays, including detector intrinsic efficiency, counting geometry, and attenuation.

TABLE I. Plutonium Isotopic Abundances (wt %) of LSD Spikes

Isotope	1st Experiment	2nd Experiment
^{238}Pu	0.00245	0.00247
^{239}Pu	97.921	97.914
^{240}Pu	2.0604	2.0628
^{241}Pu	0.0141	0.0138
^{242}Pu	0.00132	0.00543

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Because of the small sample volumes (containing less than 1 mg of plutonium) in these experiments, the isotopic ratios ^{238}Pu/^{239}Pu, ^{240}Pu/^{239}Pu, and ^{241}Pu/^{239}Pu are determined by measuring the high-intensity, low-energy gamma-ray ratios 43.48 keV/51.63 keV, 45.23 keV/51.63 keV, and 148.6 keV/ 129.3 keV, respectively /4,5/. The absolute ^{238}Pu, ^{239}Pu, ^{240}Pu, and ^{241}Pu fractions in the sample can then be determined by combining isotopic ratios and correcting for the ^{242}Pu content, which is predicted by isotope correlation techniques /6/, that work well for material from known reactor types. All gamma-ray peak areas are calculated by using a channel-by-channel summation method with a linear, straight-line background subtraction. Minor interferences in the full-energy peaks are taken into account in the assay equations. The $\varepsilon(b)/\varepsilon(a)$ ratios are also determined from internal spectral information; the factors $[I(b)/I(a)]$ and $[T_{1/2}(m)/T_{1/2}(n)]$ are known and thus the R may be computed.

Total Concentration of Plutonium

By measuring the isotopic compositions of both unspiked and spiked dissolver solution samples, the concentration of plutonium in the unknown dissolver solution, C_u, can be determined. Let W^i, C, and V be the weight fraction of isotope i, concentration (g/ℓ), and volume (ℓ), respectively, and the subscripts u, s, and m stand for the unspiked sample (dissolver solution), spike (LSD spike), and spiked sample (mixture of dissolver solution and LSD spike), respectively. Assuming first a mixture of spike solution and unknown solution, the weight fraction of isotope i in the spiked (mixed) samples is

$$W_m^i = \frac{W_s^i C_s V_s + W_u^i C_u V_u}{C_s V_s + C_u V_u} \quad . \tag{2}$$

This is the basic isotope dilution relation. However, in applying the IDGS method to the plutonium concentration of dissolver solutions it is advantageous to work primarily in terms of *isotopic ratios*. The reason for this is that ^{242}Pu does not emit a usable gamma ray and its normally small fraction must be estimated by a correlation technique in order to convert the directly and accurately measured isotopic ratios to absolute fractions. Furthermore, when using ^{239}Pu as the spike we get optimal results when using the ^{240}Pu/^{239}Pu ratio. Thus, using $i = 0$ for ^{240}Pu and $i = 9$ for ^{239}Pu, we use Eq. (2) to write

$$\frac{W_m^0}{W_m^9} = \frac{W_s^0 C_s V_s + W_u^0 C_u V_u}{W_s^9 C_s V_s + W_u^9 C_u V_u} \quad . \tag{3}$$

Solving for the plutonium concentration of the unknown dissolver solution, C_u, we obtain /7/

$$C_u = \frac{M_s}{V_u} \cdot \frac{W_s^9}{W_u^9} \cdot \frac{R_m - R_s}{R_u - R_m} \tag{4}$$

where M_s = Mass of plutonium in the spike,

R_m = W_m^0/W_m^9, the ^{240}Pu/^{239}Pu ratio in the spiked sample,

R_s = W_s^0/W_s^9, the ^{240}Pu/^{239}Pu ratio in the spike, and

R_u = W_u^0/W_u^9, the ^{240}Pu/^{239}Pu ratio in the dissolver solution sample.

In this equation, the values of M_s, W_s^9, R_s, and V_u are known. Therefore, only the values of R_u and W_s^9 in the unspiked dissolver-solution sample and R_m in the spiked sample need to be measured by gamma-ray spectrometry.

The precision and accuracy to be expected from IDGS measurements of C_u are obviously important. There is confidence that the random and systematic errors of M_s, V_u, W_s^9, and R_s are all <0.1%. Preliminary results indicate that the systematic error in C_u introduced by the gamma-ray spectrometric measurements of R_m, R_u, and W_s^9 is a small fraction of a percent. The random errors arising in the measurements of R_m, R_u, and W_s^9 will dominate the precision. An approximate expression, of reasonable accuracy, giving the relative standard deviation (RSD) of C_u in terms of the RSDs of R_m, R_u, and W_s^9 can be easily derived. R_u and W_s^9 are not fully independent statistically because of the way their values are extracted from the gamma-ray spectra, but they are "near independent." If we assume independence and propagate by usual methods we obtain

$$RSD(C_u) \cong \left[\frac{R_m^2 (R_u - R_s)^2}{(R_m - R_s)^2 (R_u - R_m)^2} RSD^2(R_m) + \frac{R_u^2}{(R_u - R_m)^2} RSD^2(R_u) + RSD^2(W_u^9) \right]^{1/2} \tag{5}$$

This expression is slightly conservative and quite accurate when compared to results of RSDs computed from replicate assays. The analysis routines that produce R_m, R_u, and W_s^9 also give good estimates of their RSDs for use in Eq. (5).

3. Resin Bead Sample Preparation

Two proof-of-principle experiments have been carried out at the Tokai Reprocessing Plant. Because IDGS measurements of highly radioactive dissolver solutions require rapid and efficient separation of plutonium from fission products and other actinides, we have developed a two-step ion-exchange method to purify and recover plutonium on resin beads. The procedure for preparing the resin bead sample is shown in Fig. 1 for the spiked sample (solid line) and the unspiked sample (dashed line). Spiked samples were prepared by mixing dissolver solutions (1 mℓ) with LSD spikes and then completely dissolving them at 90°C with 3 M HNO$_3$ (5 M HNO$_3$ and 0.01 M HF were used in the first experiment). After evaporation and redissolution with 8 M HNO$_3$, plutonium in spiked solutions was completely adjusted to tetra-valency with Fe(II) and NaNO$_2$. Because the dissolver solution used in the first experiment was over one month old, the valency was not adjusted. For each batch of dissolver solution, a 1-mℓ aliquot was also taken as an unspiked sample to be used for the plutonium isotopic composition measurement. Each spiked and unspiked sample was split into two or more subsamples. Each subsample was individually passed through the anion exchanger column (BIO-RAD AG-MP1 NO$_3$ form, 0.5 mℓ, 5 mm ϕ), followed by washing with 8 M HNO$_3$ to remove fission products and americium and 3 M HNO$_3$ to remove uranium. To better separate plutonium, we approximately

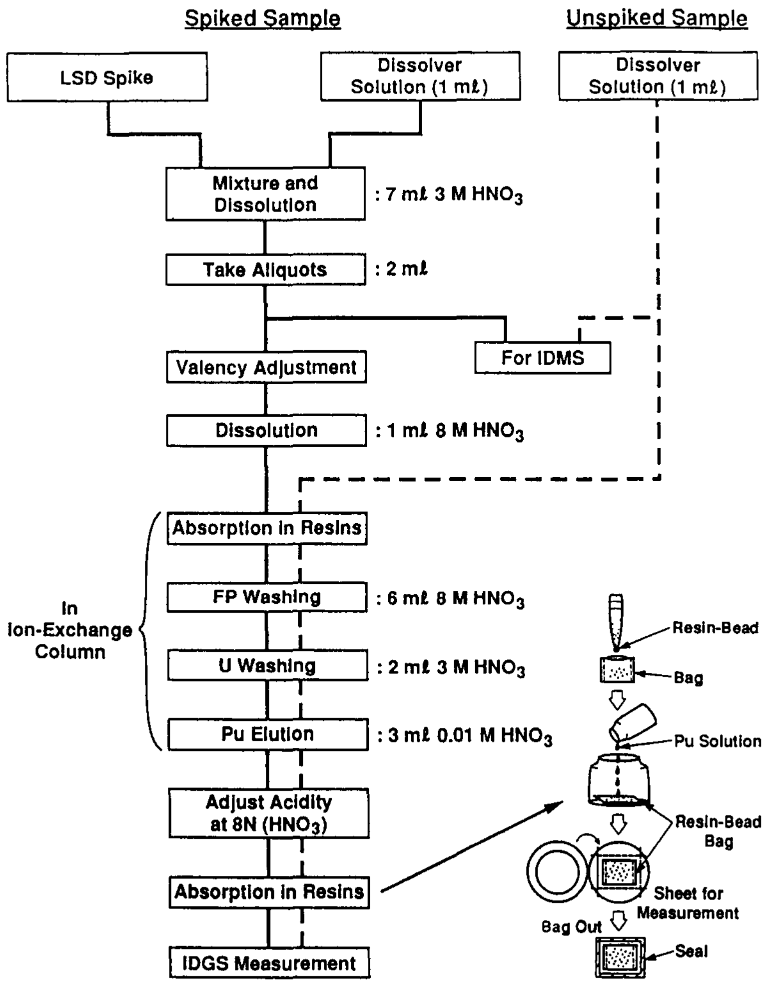

Fig. 1. The resin bead sample preparation procedure used in the second experiment for the spiked (solid line) and unspiked (dashed line) samples. This procedure is slightly different from the first experiment described in the text.

doubled the amount of 8 M HNO_3 solution used in the second experiment. Plutonium was eluted with 0.01 M HNO_3, the acidity was adjusted with 8 M HNO_3, and the plutonium was then absorbed on resin beads contained in small gauze bags. Each resin bead sample was placed in a plastic bag, removed from the glove box, and placed directly against the end cap of the high-purity germanium detector for gamma-ray measurements. The sample preparation flow originally proposed had been such that a certain amount of resins containing plutonium were taken from the ion exchange column for gamma measurement. It was, however, found that ruthenium, one of the major fission products, was hardly rinsed from the first stage of the ion separation. Therefore, the idea of plutonium elution-reabsorption mentioned above was adopted.

In the first experiment, we spiked four 1mℓ samples of input dissolver solution from one batch with four LSD spikes. Each LSD spike contained ~4.5 mg of plutonium. In the second experiment, six spiked samples were made from three different batches of dissolver solution. Two 1-mℓ dissolver-solution samples from each batch were individually spiked to make six spiked samples. The mass of plutonium in each spike was ~2 mg. The spike-to-dissolver-solution plutonium ratio changed from ~4:1 in the first experiment to ~2:1 in the second experiment to decrease the amount of spike used. In addition to plutonium masses, isotopic compositions of the spikes were also slightly different in the two experiments as shown in Table I.

4. Results and Discussion

Plutonium Isotopic Compositions

Figure 2 shows the low-energy gamma-ray spectra of (a) the pure LSD spike, (b) the unspiked (unknown) dissolver solution, and (c) the spiked dissolver solution from the first experiment. Notice the dramatic differences in gamma-ray intensities among the three samples. In the second experiment, gamma-ray peaks of ^{239}Pu (at 38.66 and 51.63 keV) are relatively smaller compared to gamma-ray peaks of ^{238}Pu (43.48 keV) and ^{240}Pu (45.23 keV) because of the smaller spike-to-dissolver solution ratio. Table II shows the gamma-ray plutonium isotopic compositions (in weight percent) for the dissolver solutions. All IDGS data shown are averages from two or three independent measurements of subsamples. A few subsamples of sample 1 in the first experiment were contaminated by fission products; data from these are not included in the averages. The K

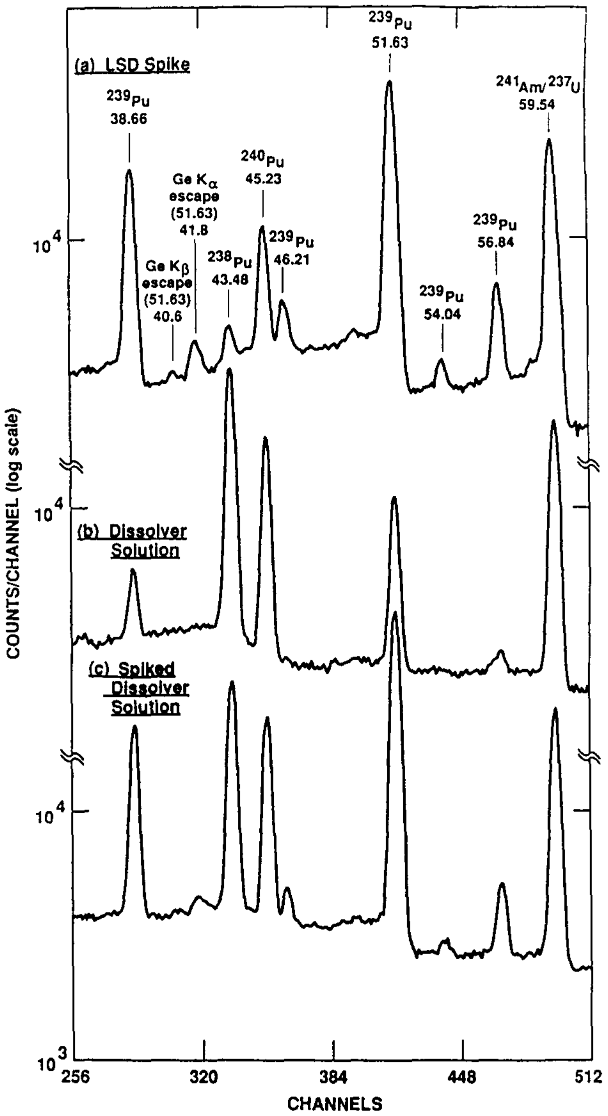

Fig. 2. Low-energy gamma-ray spectra of (a) pure LSD spike, (b) unspiked dissolver solution, and (c) spiked resin bead samples in the first experiment.

Table II. Plutonium Isotopic Abundances (wt %) of Dissolver Solutions as Determined by IDGS

Batch No.	^{238}Pu	^{239}Pu	^{240}Pu	^{241}Pu	^{242}Pu
1	1.50	56.68	27.02	9.62	5.18
2	1.50	56.63	27.14	9.56	5.18
3	1.53	58.22	25.13	10.25	4.87
4	1.55	56.63	26.75	9.83	5.25

x-rays from fission products directly interfere with the plutonium low-energy gamma rays to affect the accuracy of the measurement. Furthermore, the dramatically increased continuum background produced by higher energy gamma rays from fission products reduces the precision of measurement for a given counting time and geometry for the same mass of plutonium. Figure 3 shows a comparison of the low-energy gamma-ray spectrum from a fission-product-contaminated resin bead sample (dotted spectrum) with the spectrum from the same sample after it was rewashed to remove fission products. No low-energy gamma rays from fission products interfered in the second experiment (samples 2-4). This may be due to the double amount of 8 M HNO_3 solution used in the fission-product washings. However, some weak high-energy gamma rays from ruthenium that produce a higher continuum background were observed in spectra in both experiments. Figure 4 compares a dissolver solution spectrum with the higher continuum background with a pure LSD spike spectrum.

Table III(A) shows the comparison of the plutonium isotopic compositions of dissolver solution as determined by IDGS and IDMS. The agreement between IDGS and IDMS is generally good, especially so for the ^{240}Pu/^{239}Pu ratio (average IDGS/IDMS ratio is 0.993) and the weight percent of ^{239}Pu (average IDGS/IDMS ratio is 0.999), which are important for calculating the total plutonium concentration. Using Table III(B) we can compare the IDGS and IDMS plutonium isotopic measurements of the pure LSD spike used in the second experiment. The agreement between the IDGS method and the IDMS method is excellent except for ^{241}Pu, which has a very low fraction in the spike. Of course, gamma-ray measurements of the isotopic fractions in the spike need not be made, but were made in this case to increase confidence in the gamma-ray plutonium isotopic measurements.

Total Plutonium Concentration

From Eq. (4), the total plutonium element concentration of the unknown dissolver solution can be calculated by using measured ^{240}Pu/^{239}Pu values for the spiked solution (R_m) and for the unspiked dissolver solution (R_u), the measured weight percent of ^{239}Pu in the unspiked dissolver solution (W_u^9), and certified values for W_s^9 and ^{240}Pu/^{239}Pu (R_s) for the LSD spike. The results of the plutonium element concentrations from IDGS and IDMS are compared in Table IV. The average IDGS/IDMS ratio is 1.0005 and the sample RSD of the average ratio is 0.12%, indicating that any bias between IDMS and IDGS methods is ≤0.2%. Because each of the 10 IDGS plutonium concentration values in Table IV was computed from the average isotope ratios from an unequal number of counts of varying times, the sample RSD of the 10 ratios just mentioned says little about the precision of single measurements. The precision will be a function of sample plutonium masses, detector efficiencies, count times, and isotopic distributions. For the 1000-mm^2 detector used, with ~0.5 mg of plutonium in the spiked samples and ~0.2 mg of plutonium in the unspiked samples (although less than 60% was trapped on the resin beads), and 100-min counts on both spiked and unspiked resin bead samples, Eq. (5) estimates the RSD of the random error to be ~1.2%. By optimizing the sample preparation and perhaps using somewhat larger samples of dissolver solution, the precision could be 1.0% for 1-h count times on both samples.

5. Conclusions

In summary, we have demonstrated for the first time the simultaneous determination of plutonium concentration and isotopic composition of spent-nuclear-fuel dissolver solutions from a reprocessing plant by the IDGS technique.

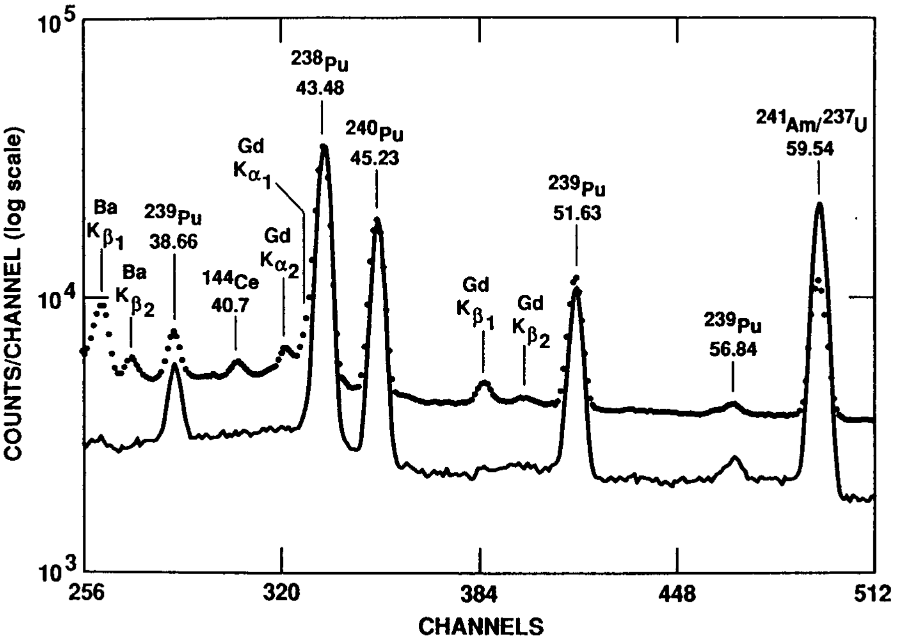

Fig. 3. Comparison of the low-energy gamma-ray spectrum from a fission-product-contaminated, unspiked resin bead sample (dotted spectrum) and from the same sample after the fission products were washed again (solid spectrum).

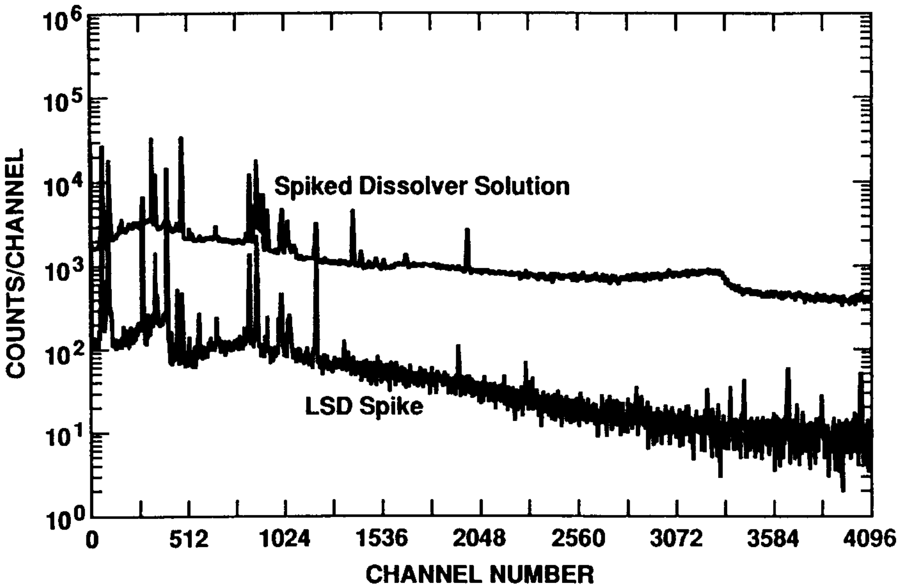

Fig. 4. Comparison of continuum background in gamma-ray spectra of (a) LSD spike and (b) spiked dissolver solution resin bead samples.

TABLE III. Comparison of Plutonium Isotopic Abundances as Determined by IDGS and IDMS

Batch No.	IDGS/IDMS ^{238}Pu	^{239}Pu	^{240}Pu	^{241}Pu	^{242}Pu	^{240}Pu/^{239}Pu
A. Dissolver Solution						
1	1.149	0.997	0.993	1.003	1.029	0.995
2	1.152	0.996	0.997	0.996	1.028	1.000
3	1.123	1.005	0.989	1.013	0.950	0.984
4	1.125	0.997	0.991	1.007	1.034	0.994
Av	**1.137**	**0.999**	**0.993**	**1.005**	**1.010**	**0.993**
B. LSD Spike						
	0.984	1.000	1.001	1.275	—	1.001

TABLE IV. Comparison of Total Plutonium Concentrations of Dissolver Solutions as Determined by IDGS and IDMS

Sample	IDGS (g Pu/L)	IDMS (g Pu/L)	IDGS/IDMS
A. First Experiment			
1	0.9702	0.9769	0.9931
2	0.9750	0.9778	0.9971
3	0.9838	0.9822	1.0016
4	0.9811	0.9806	1.0005
Average			0.9991
B. Second Experiment			
5	1.2968	1.298	0.9991
6	1.2933	1.298	0.9964
7	1.3852	1.375	1.0074
8	1.3776	1.375	1.0019
9	1.2713	1.266	1.0042
10	1.2708	1.266	1.0038
Average			1.0021

Because IDGS is adequately accurate and precise, easy to operate and maintain, and costs less than IDMS, this new technique could be implemented as an alternative method for input accountability and verification measurements in reprocessing plants. It could also complement K-edge/K-XRF measurement of plutonium concentration by providing the isotopic distributions of dissolver solutions. In this case, because only dissolver solution is to be measured and analyzed, no spiked sample is involved. The IDGS technique may also provide useful accountability and verification information on intermediate-process and hot-waste-streams, which are important for near-real-time accounting at reprocessing plants.

IDGS could also be a potential on-site verification method for IAEA inspections. At present, spiked and unspiked aliquots from each dissolver batch are prepared by the plant operators and given to the inspectors for shipping to the SAL for IDMS analysis. The turnaround time in getting the results of these analyses is usually more than one month because of difficulties in shipping plutonium-bearing samples. A simple, prompt, verification analysis for each dissolver batch that could be done at reprocessing plants would be important. By implementing the IDGS technique for on-site verification, the IAEA and domestic inspectors could promptly detect anomalies and significantly reduce the number of samples sent to the SAL for IDMS analysis.

6. Acknowledgment

The authors wish to thank E. Kuhn of the IAEA for providing us with the LSD spikes and for helpful discussions.

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THE STATE-OF-THE-PRACTICE IN LOW ENRICHED UF_6 ISOTOPIC MEASUREMENTS IN THE EUROPEAN COMMUNITY : RESULTS OF REIMEP UF_6

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Abstract

After the interruption of the SALE Programme, CBNM has established a Regular European Interlaboratory Measurement Evaluation Programme (REIMEP) with the support of the ESARDA Working Group for Techniques and Standards for Destructive Analysis (WGDA), the ESARDA Working Group for Techniques and Standards for Non-Destructive Analysis (WGNDA) and the IAEA.

On the basis of a questionnaire with answers from 41 laboratories, 36 laboratories have announced their interest for such a programme. In this paper we report on the 1986/87 round of the programme establishing the measurement capability or "State-Of-the-Practice" in UF_6 isotopic measurements by methods left at the discretion of the participants (thermal ionization mass spectrometry, electron impact mass spectrometry and gamma-ray spectrometry).

Pictures of the "State-Of-the-Practice" are presented as graphs displaying participants' results.

1. Introduction

With the support of the ESARDA Working Group for Techniques and Standards for Destructive Analysis (WGDA), CBNM organized an Interlaboratory Measurement Evaluation Programme on UF_6 in 1982 amongst UF_6 measurement laboratories in Western Europe and reported on it in 1983 /1/.

The present paper reports on a similar programme carried out in 1986/87 as a part of a more general and regular programme called REIMEP (Regular European Interlaboratory Measurement Evaluation Programme). Through this programme, CBNM intends to provide nuclear analytical laboratories with a regular opportunity

- for an external check on the quality of their measurements;
- to establish the State-Of-the-Practice of assaying common nuclear materials for isotope and element content;
- to present a true picture of the real life interlaboratory spread of given measurements on given materials.

This paper reports on the first UF_6 round in REIMEP, ("REIMEP UF_6 1987"). As opposed to the previous UF_6 interlaboratory evaluation programme /1/, participants could choose their methods to perform the UF_6 isotopic measurements.

Consequently a number of γ-ray spectrometry laboratories chose to participate with this technique. Some of them took the occasion to purchase or loan the EC NRM 171 Nuclear Reference Material set, (5 U_3O_8 "infinitely thick" layers in Al cans with ^{235}U abundances from 0.3 % to 4.5 % certified to 0.07 % of values) /2/ in order to calibrate their measurements. Also some participants sent in results obtained by thermionic mass spectrometry.

2. Participation List

2.1 Participating laboratories with measurements by Mass Spectrometry.

1. IAEA-SAL, Mr. Deron,
   Seibersdorf Austria
2. CDTN-NUCLEBRAS, Mr. Campos,
   Belo Horizonte, Brazil
3. Balzers, Mr. Rettinghaus,
   Balzers, Liechtenstein
4. LCAC Grenoble, Mrs. Chaussabel,
   Grenoble, France
5. COGEMA, Mr. Journoux,
   Pierrelatte, France
6. FBFC, Mr. Navard,
   Romans, France
7. Eurodif, Mr. Aubeau,
   Pierrelatte, France
8. Urenco Deutschland, Mr. Abt,
   Gronau, FRG
9. KfK, Mr. Bley,
   Karlsruhe, FRG
10. RBU, Mr. von Wachtendonk,
    Hanau, FRG
11. CEC-JRC, Mr. Guzzi,
    Ispra, Italy
12. URENCO, Mr. Zeckendorf,
    Almelo, The Netherlands
13. BNFL, Mr. Ogden,
    Capenhurst, United Kingdom
14. BNFL, Mr. Hayes,
    Preston, United Kingdom
15. NBL, Mr. Bingham,
    Argonne, USA
16. Goodyear Atomic Corp., Mr. Vito,
    Piketon, Ohio USA
17. M. Marietta En. Syst., Mr. Woolridge,
    Paducah, Ohio USA

2.2 Participating laboratories with measurements by Gamma-Ray Spectrometry

1. IAEA, Mr. Dragnev,
   Wien, Austria
2. SCK/CEN, Mr. Fettweiss,
   Mol, Belgium

3. BAM, Mr. Thiele,
Berlin, FRG
4. KfK, Mr. Ottmar,
Karlsruhe, FRG
5. ENEA, Mr. Cresti,
Roma, Italy
6. JRC, Mr. Guardini,
Ispra, Italy
7. ECN, Mr. Harry,
Petten, The Netherlands
8. NBL, Mr. Bingham,
Argonne, USA
9. LANL, Mr. Reilly,
Los Alamos, USA
10. M. Marietta En. Syst., Mr. Beverly,
Paducah, Ohio USA

3. Preparation of the Material Batch, Verification of its Isotopic Homogeneity and Verification of Sample Identity with regard to Isotopic Composition

The UF_6 material for this programme was prepared by mixing 2 UF_6 materials of different isotopic composition, $^{235}U/U$ = 0.007 and 0.05 /3/, until it was homogeneous. This homogeneity was verified and results are shown in Fig. 3.1.

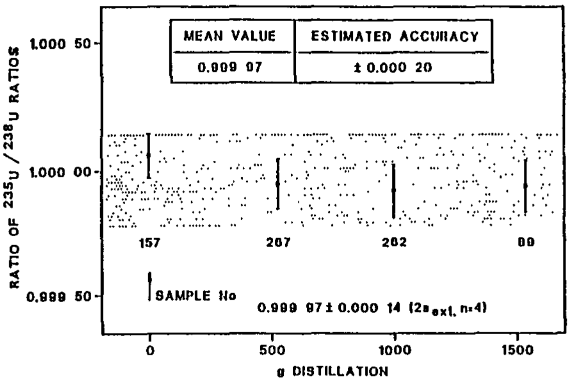

Fig. 3.1 - Verification of isotopic homogneity by sampling (157-267-262-89) during the distillation process and subsequent UF_6 mass spectrometry

To check the isotopic homogeneity of all samples issued UF_6 mass spectrometry was used on the basis that it allows intercomparison of $^{235}U/^{238}U$ ratios in two samples of materials with good precision (2s ≤ 0.02 % of ratio value) /4/. The results are displayed in Fig. 3.2 and allow to conclude to an isotopic homogeneity of the UF_6 material as bottled of better than 0.02 % (2s). This is expressed as the possible maximum variability of the $^{235}U/^{238}U$ ratios throughout all samples.

Also, all samples intended for gamma-ray spectrometry were compared to each other by gamma-ray spectrometry. Results are displayed in Fig. 3.3. It can be concluded that the maximum variability of the apparent ^{235}U abundance is 0.06 % (2s) as measured by gamma-ray spectrometry. A full report is available separately /3/.

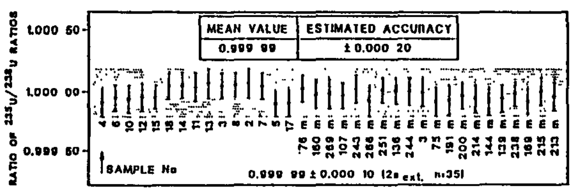

Fig. 3.2 - Verification of isotopic identity (by UF_6 mass spectrometry) of all samples distributed in the programme

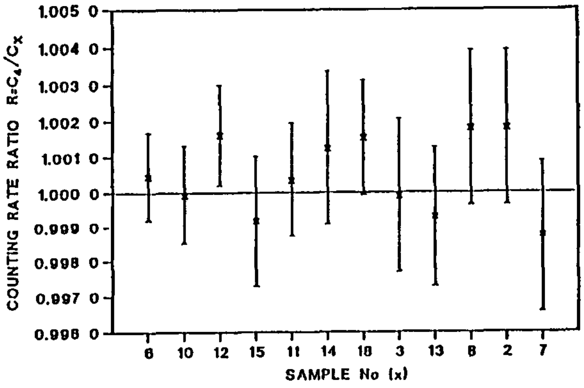

Fig. 3.3 - Verification of sample uniformity of all samples versus sample 4 by measuring the counting ratio of the 185.7 KeV gamma ray of ^{235}U

4. Characterization of the Samples Distributed

The material distributed was extensively characterized by UF_6 /5/,/6/ and thermionic mass spectrometry /7/ at CBNM for all the quantities evaluated in this programme. The sample preparation and the measuring technique of both methods were different but gave the same results within the accuracy of the measurement. A conservative uncertainty range was placed on each "declared value" which contains all uncertainties from possible uncertainty components or corrections and of the Reference Materials or synthetic isotope mixtures used in the characterization process /8/. A full report on the characterization is available separately /3/.

5. Description of the Samples for Gamma-Ray Measurements

Details of the samples for gamma-ray measurements are given in Fig. 5.1. Details of the accompanying calibration discs in monel and aluminium are given in Fig. 5.2.

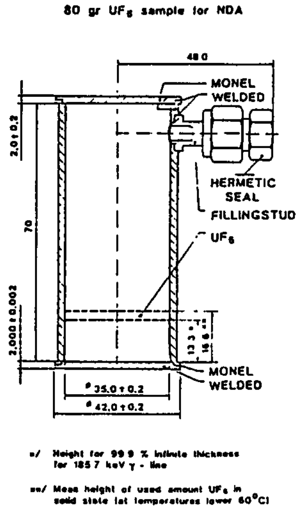

Fig.5.1 - UF6 sample γ-ray spectrometric measurements

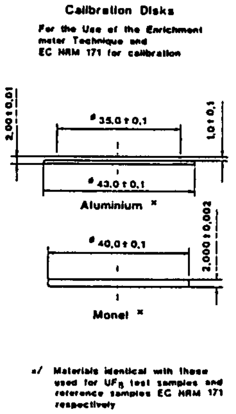

Fig. 5.2 - Calibration disks

A special note for gamma-ray participants was mailed with their sample, intending to assist them with their measurements.

6. Results

The results are graphically displayed in Figs. 6.1 to 6.8 They display the situation of measurements of various quantities.

Fig. 6.1	^{235}U/U molar ratio by mass spectrometry and by gamma-ray spectrometry
Fig. 6.2	^{235}U abundance measurements in mass % (mass spectrometry only)
Fig. 6.3	^{234}U abundance measurements in mass % (mass spectrometry only)
Fig. 6.4	^{236}U abundance measurements in mass % (mass spectrometry only)
Fig. 6.5	^{238}U abundance measurements in mass % (mass spectrometry only)
Fig. 6.6	molar isotope ratio ^{235}U/^{238}U (mass spectrometry only)
Fig. 6.7	molar isotope ratio ^{234}U/^{238}U (mass spectrometry only)
Fig. 6.8	molar isotope ratio ^{236}U^{238}U (mass spectrometry only)

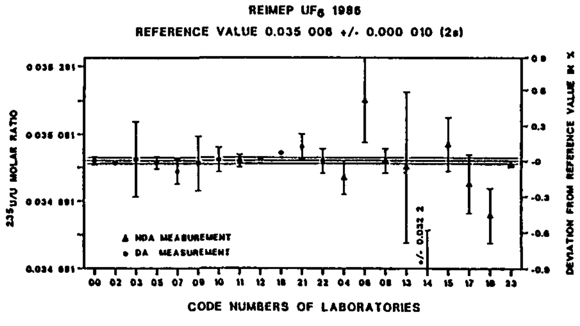

Fig 6.1 - Results of ^{235}U/U molar ratio determinations by MS (•) and γ (▲)

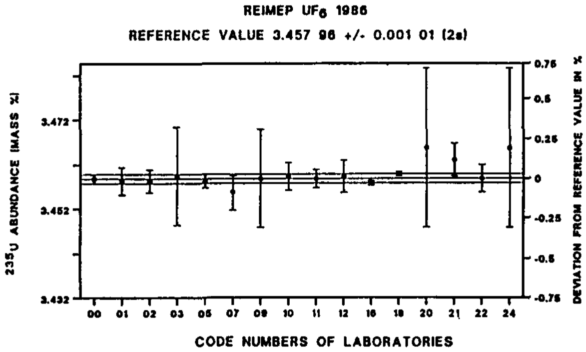

Fig. 6.2 - Results of ^{235}U abundance determinations

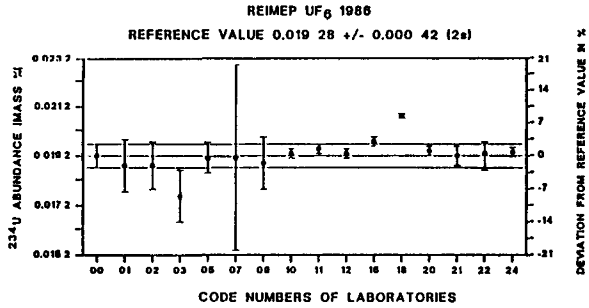

Fig. 6.3 - Results of ^{234}U abundance determinations

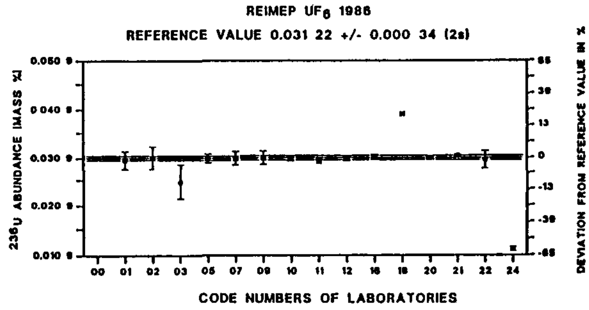

Fig. 6.4 - Results of ^{236}U abundance determinations

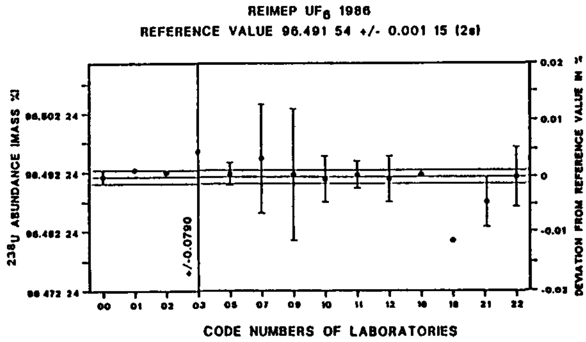

Fig. 6.5 - Results of ^{238}U abundance determinations

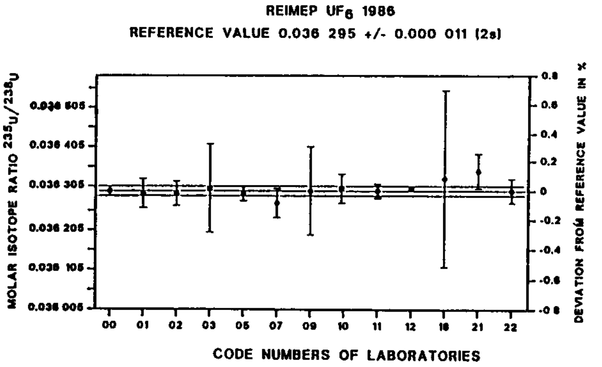

Fig. 6.6 - Results of $^{235}U/^{238}U$ molar isotope ratio determinations

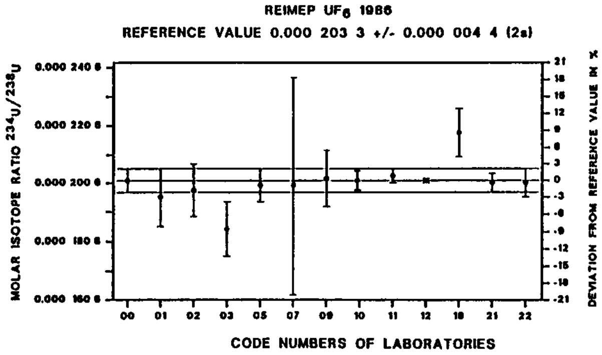

Fig. 6.7 - Results of $^{234}U/^{238}U$ molar isotope ratio determinations

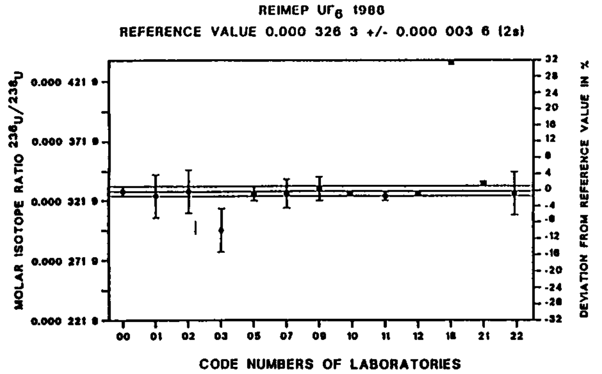

Fig. 6.8 - Results of $^{236}U/^{238}U$ molar isotope ratio determinations

7. Conclusions

1. The 1987 State-Of-the-Practice of the molar isotope ratio and mass abundance determinations in low enriched UF_6 isotopic measurements by mass spectrometry can be summarized as follows (expressed as interlaboratory spread of participants' values computed on a 2s basis and as average deviation from the Reference Value):

Molar Isotope Ratio	Spread (on 2s Basis in % of Value)	Average Deviation from Reference in % of Value
$^{234}U/^{238}U$	± 3.71	- 0.44
$^{235}U/^{238}U$	± 0.05	+ 0.01
$^{236}U/^{238}U$	± 9.94 *	+ 1.41 *

* caused by the result of one laboratory

Isotope Abundance (mass %)	Spread (on 2s Basis in % of Value)	Average Deviation from Reference in % of Value
^{234}U	± 3.25	- 0.05
^{235}U	± 0.08	+ 0.03
^{236}U	± 18.65 *	- 3.41 *
^{238}U	± 0.004	- 4.46 X 10^{-4}

* caused by the result of one laboratory

Molar Abundance	Spread (on 2s Basis in % of Value)	Average Deviation from Reference in % of Value
$^{235}U/U$	± 0.05	+ 0.01

2. The measurement capability for minor isotope abundance measurements is good and has been quantified. It is now usable for evaluating the usefulness of MIST (Minor Isotopes Safeguarding Technique).

3. The 1987 State-Of-the-Practice of $^{235}U/U$ abundances in low enriched UF_6 isotopic measurements by gamma-ray spectrometry can be summarized as follows (expressed as interlaboratory spread of participants' values computed on a 2s basis and as average deviation from the Reference Value):

Molar Isotope Abundance	Spread (on 2s Basis in % of Value)	Average Deviation from Reference in % of Value
$^{235}U/U$	± 0.98* ± 0.19**	- 0.21* + 0.000 4**

* caused by the result of one laboratory
** not taking in account the previously cited one laboratory

This denotes a drastic improvement of the accuracy of gamma-ray spectrometry over the previous situation, possibly due to the increased use of the available Reference Material.

References

/1/ P. De Bièvre, W.H. Wolters, G. Müschenborn, W. De Bolle, Proc. 5th ESARDA Symposium, Versailles, France, April 1983 p. 177.

/2/ P. De Bièvre, H.L. Eschbach, R. Lesser, H. Meyer, J. Van Audenhove (CBNM), B.S. Carpenter (NBS), "^{235}Uranium Isotope Abundance Certified Reference Material for Gamma Spectrometry. EC Nuclear Reference Material 171 Certification Report COM 4153", available from CBNM.

/3/ W. De Bolle, R. Damen and W. Lycke, "Preparation, Isotopic Characterization and Verification of Isotopic Homogeneity of UF_6 Material for the Regular European Interlaboratory Measurement Evaluation Programmes - REIMEP", Internal Report, CBNM-R-MS-96-87.

/4/ W. De Bolle, G. Müschenborn and P. De Bièvre, "The Potential of Double Collector UF_6 Mass Spectrometry for Verification of Isotopic Homogeneity. Application to U_3O_8 RMs for Gamma Spectrometry", Proc. 7th ESARDA Symposium, Liège, Belgium, May 1985, p. 277.

/5/ G. Müschenborn and W. De Bolle, "$^{235}U/^{238}U$ Isotope Ratio Measurements on Double Collector UF_6 Mass Spectrometers",Internal Report CBNM-R-MS-74-84.

/6/ R. Werz and G. Müschenborn,"Memory in UF_6 Mass Spectrometers", Internal Report CBNM-R-MS-4-80.

/7/ W. De Bolle, "Conversion of UF_6 in Uranyl Nitrate for Isotopic Measurements by Thermionic Mass Spectrometry", Internal Report CBNM-R-MS-25-83.

/8/ P. De Bièvre, R. Damen, L. Traas, W. De Bolle, A. Verbruggen, W. Lycke, M. Gallet, R. Werz, M. van den Berg, "Uncertainties and Certainties in Isotopic Measurements of Uranium: Keys to accuracy", Invited Plenary Lecture at the International Conference on Nuclear and Radiochemistry ICNR-86, Beijing China, September 1986

WHY SHOULD WE DO SOME MORE "ISOTOPE-SPECIFIC" THINKING IN MEASUREMENTS FOR SAFEGUARDS?

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Abstract

A number of reasons are given why more isotope-specific or isotope-related thinking should occur in measurements for Safeguards and Nuclear Material Management. It is isotopes which are fissile. Isotope-specific methods are available for direct quantitative determination of isotopes without a "detour" via elemental content with its problems in sample stoichiometry. Fissile isotopes in input solutions of reprocessing plants are already assayed by an isotope-specific method (IDMS*) and enable direct isotope-accountancy. Within the same amount of element, fissile isotopes (e.g. ^{235}U) can be substituted for non-fissile isotopes (e.g. ^{238}U) using isotope separation techniques of the near future leaving the total amount of element the same but changing the Safeguards character of the material drastically. Isotope-specific ratio of ratio measurement approaches, applied to IDMS have the potential of breaking through the 0.1% uncertainty barrier, at the same time making measurements easier. Large dynamic ratio measurement capability would enable the determination of the total amount of fissile isotope within a nuclear material batch (e.g. an input tank) directly against a small (ppm) but known added amount of a spike isotope (e.g. ^{233}U) without the necessity of separate calibration of the tank for mass or volume of solution. Large dynamic isotope ratio measurement capability could also make it meaningful to "tag" material batches by means of small amounts of a "tagging" isotope, resulting in a highly specific "identity card" and the possibility to monitor the "tag" by a measurement anytime.

*IDMS = Isotope Dilution Mass Spectrometry

1. Introduction

It is isotopes which are fissile for thermal neutrons : ^{233}U and ^{235}U, ^{239}Pu and ^{241}Pu. Consequently it is these isotopes which determine the economic and safeguards value of nuclear material and it is the amount of ^{233}U, ^{235}U, ^{239}Pu and ^{241}Pu which is of primary interest. However, in the early years of the nuclear era, analytical methods to determine the amount of a nuclear element (U, Pu) in a nuclear material (radioactive solution, oxide, ...) were borrowed from classical analytical chemistry and were, therefore, methods for elemental assay (U element, ...). A second step had to be added - isotopic analysis- which yielded the fissile isotope content($^{235}U/U$, ...) of the nuclear element (U, ...). The way of thinking in two steps - first element, then isotope - is so deeply entrenched in the minds that it is even used in cases where the direct assay of the fissile isotope by an isotope-specific method (e.g. Isotope Dilution Mass Spectrometry IDMS) in one step is possible : IDMS is universally used to assay element content followed by an isotopic analysis (two step approach) whereas it is basically a method to directly assay isotope content (one step approach). In addition : two steps necessarily carry more error possibilities and a larger uncertainty of the end result than one step, but even this simple consideration does not prevent analysts from taking the two step approach.

It is occasionally argued that isotopic analyses are too expensive. Consider, however, the value of the electricity generated by one ton of fuel rated at 30,000 megawatt days/ton. At $0.06/kWh this fuel will generate $50 400 000 in electrical sales. A 0.2 % uncertainty in fissile content represents a $100 800 uncertainty in revenue.

2. The lack of sensitivity of elemental assay for change in fissile isotope content

Elemental analysis/accountancy is very insensitive to variations in fissile isotope content. Consider the two extremes represented in Table I. It will be obvious that the ^{235}U (fissile) content has been reduced by 500 %. However, the atomic weight of the U has changed by only 0.06 %. The measured elemental concentration of two solutions of identical molar concentrations, but of wildly different isotopic assays, will only differ by 0.06 % even when measured by the most accurate elemental procedure available. To define the true economic and safeguards value of the fuel, one needs to multiply the elemental concentration by the ^{235}U abundance, $n(^{235}\mathrm{U})/n(\mathrm{U})$.

	Uranium (fresh fuel)	Uranium (after 80% burn-up)
mol % ^{234}U	0.030	0.015
mol % ^{235}U	5.500	1.100
mol % ^{236}U	0.053	0.600
mol % ^{238}U	94.417	98.285
Atomic Weight (Molar Mass)	237.883	238.005

Table I - Identical molar concentrations of fresh fuel and burn-up solutions are 0.06% different when recomputed in g·L^{-1}

A similar example for Pu is as follows :

	Plutonium (low burn-up)	Plutonium (medium burn-up)
mol % ^{238}Pu	0.01	0.35
mol % ^{239}Pu	98.00	72.32
mol % ^{240}Pu	1.90	24.50
mol % ^{241}Pu	0.08	2.58
mol % ^{242}Pu	0.01	0.25
% Fissile Isotopes	98.08	74.90
Atomic Weight (Molar Mass)	239.073	239.353

Table II - Identical molar concentrations of low burn-up and medium burn-up solutions are 0.12% different when recomputed in g·L^{-1}

The change in atomic weight and therefore in elemental concentration, is 0.12 % for two samples with the same molar concentration.

3. Exploiting Better the Isotope-Specific Character of Classical IDMS

In Isotope Dilution Mass Spectrometry (IDMS) an unknown amount of U is determined traditionally against a known added amount (spike) of ^{233}U by measuring the ratio $n(^{238}\mathrm{U})/n(^{233}\mathrm{U})$, where n is a number of atoms or of moles : see Fig. 1. The ^{238}U isotope is taken because it represents almost the total element amount (measured values for ^{234}U - ^{235}U - ^{236}U are added up to make up for the total element). A subsequent calculation by the ^{235}U abundance $n(^{235}\mathrm{U})/n(\mathrm{U})$ yields the ^{235}U fissile isotope content.

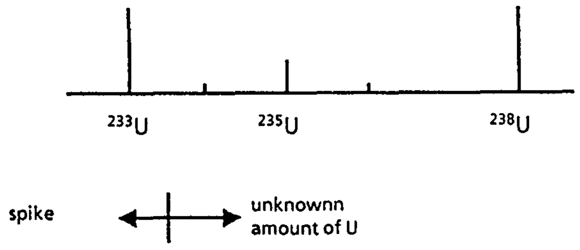


Fig. 1 Classical IDMS : *measured*: $\frac{n(^{238}U)}{n(^{233}U)}$

The uncertainty of the measurement has two main components : a contribution from the ^{238}U/^{233}U measurement and a contribution from the ^{235}U/U isotopic analysis. Recognising the isotope-specific character of the (IDMS) method, it is possible to measure directly the unknown ^{235}U amount against the known ^{233}U amount by a ratio measurement R = $n(^{235}\mathrm{U})/n(^{233}\mathrm{U})$. See Fig. 2. This would yield a measurement with one main uncertainty component, hence half the uncertainty of the previous approach. In addition much less ^{233}U is consumed in the process : spike ^{233}U should be added to match the ^{235}U content rather than the 50-200 times higher ^{238}U content, thus reducing consumption of ^{233}U by a factor of 50-200. Result : an isotope-specific method, properly applied, can define the fissile isotope content of the fuel in a direct, simple transparent measurement at half the uncertainty and much less spike consumption. A similar case can be made for Pu especially when using the very rare ^{244}Pu isotope.

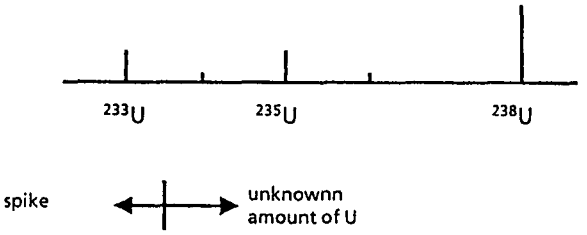


Fig. 2 Isotope-specific IDMS : *measured*: $\frac{n(^{235}U)}{n(^{233}U)}$

4. Transparency of the Isotope-Specific Measurement - Traceability to the Mole

Note that the measurement under point 3 above is carried out in an isotope mass spectrometer (IMS) as a simple ratio determination of ^{233}U and ^{235}U ion currents (Fig. 3).

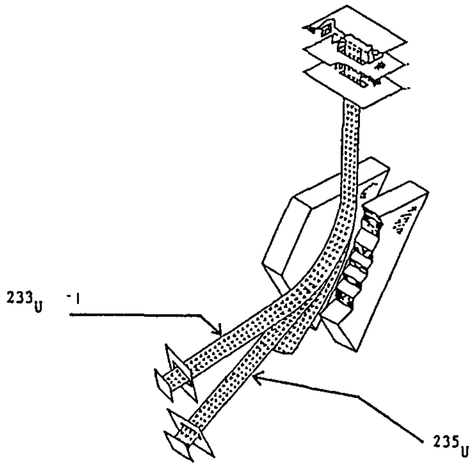


Fig. 3 - The ratio measurement $n(^{235}U)/n(^{233}U)$ (n = amount of substance expressed in numbers of atoms or moles)

It is fully analogous to the determination of the ratio of weights or masses by means of a balance (Fig. 4).

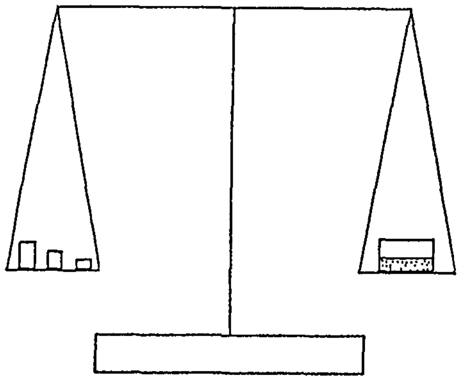


Fig. 4 The ratio measurement of masses (weights)

However, the balance compares amounts of substance in g or kg, whereas the IMS compares amounts of substance directly in numbers of atoms or of moles. <u>The determinations are directly traceable to the mole</u>, our international SI unit for amount of substance, in a simple, transparent way. At a time when the chemical measurement community starts to worry about the traceability of chemical measurements in order to ensure their comparability, time has also come for the Safeguards measurement community to demonstrate 'traceability" of its measurements. In both cases (chemical and Safeguards) traceability to our accepted international unit of measurements will have to be demonstrated if we want true compatibility of measurements.

5. Potential for Inherent Accuracy of an Isotope-Specific Methods such as IDMS

5.1 Once the sample and the added spike have been equilibrated, the newly established ratio is the same for any portion of the sample as it is for the entire material batch. Therefore, a knowledge of the chemical yield of any purification chemistry is not required. Or, simpler, the quantitativity of any sample treatment and handling is not anymore required as is a requisite in classical analytical chemistry.

5.2 Only isobars at the mass positions of the sample and spike isotopes in the mass spectrum, could create an error in the measured abundance ratio. But isobars can be detected easily for their presence/absence examining the neighbouring mass positions in the spectrum for the "isotopic fingerprint" related to the isobar.

5.3 The uncertainty terms associated with measuring the amount of other isotopes of the element do not now affect the uncertainty of the determination of the concentration of fissile isotope.

5.4 The measurement of the isotope ratio can be calibrated by means of Isotopic Reference Materials (IRMs) or even synthetic isotope mixtures, available from CBNM and certified to 0.1-0.01 % of value.

5.5 Considerations on neither stoichiometry nor chemical purity nor atomic weights must be made in an isotope-specific method such as IDMS, as must be done in classical elemental assay.

6. Evaluation of Process Losses

Plutonium exhibits more complex chemistry and presents a greater potential for selective fissile isotope process losses than uranium. The neutron density is not constant along the entire length of a fuel rod. For this reason, a higher percentage of the plutonium will occur in the centre of the assembly as isotopes of higher mass. Thus, in these regions, the amount of the fissile isotope 239 may be lower per gram of elemental Pu. Table II shows a typical isotopic composition for burn-up fuel. Only 74.9 % of the isotopes are fissile. In contrast, the Pu near the ends of the fuel rod may consist of more than 97 % fissile isotopes. It is also true that, because of differences in physico-chemical structure, the ends may have different solubilities than the centres and thus exhibit different recovery rates when subjected to the same separation chemistry. Because the process is optimized for elemental recovery, it may be more efficient in recovering more plentiful 74.9 % fissile plutonium. If the overall recovery efficiency of the process is 99 %, the elemental analysis will show these 99 %. If the 1 % elemental loss is entirely the "end" 97 % ^{239}Pu component of the fuel, the fissile isotope loss is

1 % ·97/74.9 = 1.3 %. The actual recovery of fissile isotopes is only 98.7 %. The waste stream from the first extraction would represent an attractive diversion path. The 0.3 % which represents selective isotopic loss would be a continuous source term that would be undetected by elemental analysis.

7. Diversion Scenarios

Recent advances in photon-induced isotope specific chemical reactions raise the possibility that a small portion of any isotope might be selectively precipitated, extracted or complexed on a continuous basis. A number of such systems have been described for uranium. Let us assume that fuel is reprocessed of the same isotopic composition shown in the 80 % burn-up column in Table I. By using one of the photon-induced isotopic reactions it is possible to remove 1.0 % of the ^{235}U isotope from the fuel on a continuing basis. The missing ^{235}U is then replaced by ^{238}U. For every 10 000 kg of uranium processed, the diversion would result in a loss of 1 kg of high purity ^{235}U. The elemental assay of the fuel remains the same and the fissile isotope loss goes undetected by elemental analysis of the product.

An isotopic accountancy clearly shows that all of the ^{238}U is still in inventory but that 1 kg of ^{235}U is missing : the accuracy for determining the unknown amount of ^{235}U by an isotope-specific method (IDMS) is better than 0.1 % in properly calibrated systems. Application of the same scenario to a solution phase in a fuel fabrication facility where the fissile isotope abundance is 5 %, the "losses" could be correspondingly greater. A similar diversion scenario possibility is given using a hypothetical enriching scheme for ^{239}Pu :

a an isotope separation process has been developed which is compatible with small-scale operation as a loop within a fuel reprocessing facility
b the process can produce a product that can enrich 83 % ^{239}Pu to 98 % ^{239}Pu; the remaining 2 % is ^{240}Pu.
c the system is being operated on a scale to remove 0.5 % of the available ^{239}Pu.
d the annual Pu production of the facility is 2000 kg.
e the mean isotopic assay of the input feed material :

	mol %
^{238}Pu	0.247
^{239}Pu	83.128
^{240}Pu	12.069
^{241}Pu	3.991
^{242}Pu	0.565

The effect of this scenario then could be :

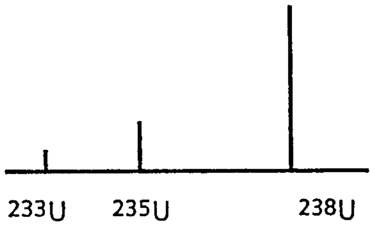
- at 0.5 % removal, 8.31 kg of ^{239}Pu is removed per year as well as 0.17 kg ^{240}Pu;
- this will affect the total Pu material balance by - 0.424 %;
- this same loss will shift the ^{239}Pu/^{240}Pu ratio by - 0.432 %.
- the ^{240}Pu loss will shift all of the other isotope ratios relative to ^{240}Pu by + 0.07 %.

This diversion can very well take place within present measurement capabilities as represented by the "Target Values" (0.5 % for random and 0.5 % for systematic error component in element assay of inputs and 0.3 % in ^{239}Pu/^{240}Pu isotope ratio measurements) /1/. The calculations here indicate that it can easily be detected with existing techniques if maintaining an isotopic material balance across the process and calculation of mean isotope ratios at regular intervals. Additionally, the degree of the consistency of the other isotope ratios provides a means of evaluating systematic errors in the isotopic measurements. This leads to the following conclusions :

1. The elemental assay of total Pu will probably not detect this type of diversion.
2. The diversion could take place within the present "Target Values for Uncertainty Components in Nuclear Material Measurements" in element assay of inputs /1/.
3. The shift in isotope ratios is within the 0.3 % presently indicated in the Target Values for isotopic measurements /1/. If the Operator and Inspector can each have 0.3 % uncertainty, then $\sqrt{0.3^2+0.3^2}$ = 0.42 % is allowed on the difference.
4. To detect this type of diversion it will be necessary to maintain material balances by isotope rather than by element.
5. The data for maintaining isotopic material balances is available. It includes elemental and isotope assay for input accountability tank batches and elemental and isotope assay for product batch.
6. Small errors in elemental assay of either input or output batches will not significantly alter the calculated ratios of isotopes in either materials balance. This fact reduces the effect of systematic errors due to tank calibrations and is the reason that isotopic material balances are superior for the detection of this type of diversion.
7. This same test will detect any systematic loss of one isotope that may be caused by failure to achieve isotopic equilibrium in the dissolution process.

8. Effects of Emerging Measurement Techniques : Direct Assay of Fissile U Isotope in an Input Tank

The modern commercial isotope mass spectrometer is capable of accurately measuring the ratio of amounts of two isotopes of an element. The major limitations to accuracy have been twofold : (1) the dynamic range of the ion detectors have been no better that 10^5 - 10^6 and (2) the abundance sensitivity (the ability to measure a small peak next to a large peak) has been limited to several ppm for adjacent masses. Recent advances that couple pulse counting and faraday cup detectors show promise of calibrateable dynamic ranges of 10^9 - 10^{10}: 10^{-11}A ($6 \cdot 10^9$ ions·s^{-1}) down to 10^{-19}A (1 ion·s^{-1}). A 1 ppm added "minor" isotope (e.g. ^{233}U or ^{244}Pu) would still measure at 10^{-17}A ($6 \cdot 10^3$ ions·s^{-1}). An integration time of 100 s would still yield < 0.2 %. Recent advances in ion optics on a commercial mass spectrometer have demonstrated abundance sensitivities of 2 x 10^8. The combination of these two technologies has made it possible, for the first time, to consider the possibility of measuring ratios of about 10^{-6} to an accuracy of 0.1 % or better. See Fig. 5.



$$n(^{233}U)/n(^{235}U) = 10^{-6}$$

$$n(^{235}U)/n(^{238}U) = 10^{-2}$$

Fig. 5 - Direct spiking of input tanks to assay fissile ^{235}U

All that has been lacking were suitable Reference Materials. Recently, a suite of uranium RMs with certified ratios in this range has been released by CBNM and work is underway at Los Alamos and the Central Bureau for Nuclear Measurements to verify the accuracy capabilities of the new technologies (see Table III).

Application of this technology can eliminate one of the largest error components associated with measurements of input amounts in fuel reprocessing facilities by spiking the whole tank with a suitable amount of ^{233}U, perform isotopic equilibration and measure fissile ^{235}U directly against this known ^{233}U.

Code number	Molar isotope ratios	
	^{233}U/^{235}U certified to ± 0.03 % of value	^{235}U/^{238}U certified to ± 0.000 20
CBNM-IRM		
072/ 1	1.000 33	0.991 03
072/ 2	0.699 67	0.991 68
072/ 3	0.499 85	0.992 12
072/ 4	0.299 87	0.992 56
072/ 5	0.100 01	0.992 99
072/ 6	0.050 091	0.993 10
072/ 7	0.019 994	0.993 17
072/ 8	0.010 165	0.993 19
072/ 9	0.005 000 0	0.993 20
072/ 10	0.002 001 2	0.993 21
072/ 11	0.000 968 92	0.993 21
072/ 12	0.000 500 88	0.993 21
072/ 13	0.000 101 82	0.993 21
072/ 14	0.000 019 996	0.993 21
072/ 15	0.000 001 999 5	0.993 21

Table III - CBNM Isotopic Reference Material Set to test linearity of mass spectrometers

Equation 1 shows the calculation presently required to measure that input by isotope dilution techniques.

$$n(^{235}U) = \frac{m_T}{m_S} \cdot R_{235/233} \cdot n(^{233}U) \qquad [1]$$

where :

$n(^{235}U)$ = total number of ^{235}U atoms in the input accountability tank

m_S = the mass of solution sample taken

m_T = the mass of solution in the input accountability tank

$R_{235/233}$ = the ratio numbers of ^{235}U atoms to ^{233}U atoms in the sample after spiking with ^{233}U

$n(^{233}U)$ = the known number of ^{233}U atoms, added to the measurement sample prior to purification chemistry

The term m_T/m_S represents the largest uncertainty because the problem of the calibration of an input tank has been there "forever" and is still unsolved as far as independent verification is concerned.

If one adds the ^{233}U spike directly to the tank rather than to a proportional sample from the tank (see Fig. 5), the equation for determining the fissile uranium in the tank is simplified to

$$N(^{235}U) = R_{235/233} \cdot N(^{233}U) \qquad [2]$$

or

$$N(^{235}U) = \frac{1}{R_{233/235}} \cdot n(^{233}U) \qquad [3]$$

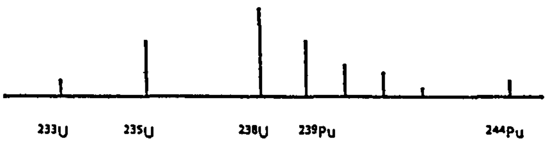
where $n(^{233}U)$ is the number of ^{233}U atoms added to the tank. A separate prior measurement will determine the traces of ^{233}U already present, permitting to correct for them.

The number of atoms of ^{235}U is expressed in terms of total atoms contained in the input tank rather than as atoms/gram of solution. The term m_T/m_S is eliminated from the equation. Only the uncertainty terms associated with measuring terms $R_{235/233}$ and $n(^{233}U)$ must be considered. Note also that after equilibration of the added spike, $R_{235/233}$ is the same for any portion of material withdrawn from the tank. This fact permits removal of very small aliquants of material suitable for direct chemical processing without any dilution or weighing. Because of the small amounts involved, it is likely that spiking and separations chemistry can be accomplished in the much cleaner environment of a minimum shielding facility or after a rough decontamination in a hot cell. At a U concentration of 250 $g \cdot L^{-1}$, the ^{235}U isotope concentration in the tank is about 2.5 $g \cdot L^{-1}$. Spiking with ^{233}U at the 1 ppm level relative to ^{235}U would only require 25 mg ^{233}U for a 10^4 L tank.

9. Effects of Emerging Measurement Techniques : Direct Assay of Fissile Pu Isotope(s) in an Input Tank

Let us now look at a possible measurement technique which could be used to determine the fissile plutonium content of the same batch of material. First prepare a mixed spike of ^{233}U and ^{244}Pu in which the ratio $n(^{244}Pu)/n(^{233}U) = R_{4/3}$ 1 $\mu g \cdot mL^{-1}$/2 $\mu g \cdot mL^{-1}$ is accurately determined to 0.1 %. Assume a tank solution at normal concentrations : $n(Pu)/n(U) \approx 2\ mg \cdot mL^{-1}/250\ mg \cdot mL^{-1}$ meaning $n(^{239}Pu)/n(^{235}U)$ 1.2 $mg \cdot mL^{-1}$/2.5 $mg \cdot mL^{-1}$ (see Fig. 6).

Spiking 1 mL of the tank input solution with 1 mL of the mixed spike, the relationship between the numbers n of atoms of isotopes in the blend can be written as (see also Fig. 6)



$$\frac{n(^{239}Pu)}{n(^{235}U)} \approx 0.5 \qquad \frac{n(^{233}U)}{n(^{235}U)} \approx 10^{-3} \qquad \frac{n(^{244}Pu)}{n(^{233}U)} \approx 0.5 \qquad \frac{n(^{239}Pu)}{n(^{244}Pu)} \approx 10^{+3}$$

Fig. 6 - Ratios in spiked input sample

$$\frac{n(^{233}U)}{n(^{235}U)} \cdot \frac{n(^{244}Pu)}{n(^{233}U)} \cdot \frac{n(^{239}Pu)}{n(^{244}Pu)} = R_{3/5} \cdot R_{4/3} \cdot R_{9/4} \qquad [4]$$

in our example $10^{-3} \cdot 0.5 \cdot 10^{3}$

or

$$\frac{n(^{239}Pu)}{n(^{235}U)} = R_{3/5} \cdot R_{4/3} \cdot R_{9/4} \qquad [5]$$

since $n(^{239}Pu)/n(^{235}U)$ in the spiked sample is the same as the ratio $n(^{239}Pu)_T/n(^{235}U)_T$ in the tank :

$$n(^{239}Pu)_T = n(^{235}U)_T \cdot R_{3/5} \cdot R_{4/3} \cdot R_{9/4} \qquad [6]$$

$n(^{235}U)_T$ has previously been determined from the direct tank spike measurement. R_{35} and R_{94} can be measured in the spiked sample. R_{43} has been determined during the preparation of the mixed spike. Therefore, $n(^{239}Pu)$ in the input tank has been defined. What is needed is measurement capability over a larger dynamic range on commercially available instruments of higher abundance sensitivity which can accurately measure large ratios (10^{-3} to $< \pm 0.1$ % for use in Eq. 4). Similar equations can be solved for all isotopes of plutonium :

$$n(^{i}Pu)_T = n(^{235}U)_T\, R_{3/5} \cdot R_{4/3} \cdot R_{i/4} \qquad [7]$$

Any traces ^{244}Pu already present can be determined in a separate measurement and corrected for. Summed up, they define the total number of atoms of plutonium in the input tank, yielding a detailed verification of all Pu isotopes independently.

The following points should be considered when evaluating the value of pursuing the proof testing of this potential new measurement technique for input analysis :

1. only one weighing operation is required (on the ^{233}U added to the tank). This weighing can be performed in the controlled conditions af the analytical laboratory.
2. aliquots for either the uranium isotope ratio determination R_{35} or the plutonium isotope ratio determinations R_{94} do not require additional weighings or volume measurements.
3. because the amounts of material withdrawn do not need to be quantitative, very small aliquots can be directly spiked with very small amounts of mixed spike thereby conserving the spike.
4. the isotope ratios to be measured are fixed at the moment of spiking thereby eliminating the error terms associated with evaporation and salting. These errors are of concern when subsampling is required prior to analysis.
5. no dilution errors can occur : all aliquoting is non-quantitative.
6. the error terms associated with the very difficult task of input accountability tank calibration are totally eliminated as

are the error terms associated with determining the precise amount of sample removed.

Simply summarised : all measurements are isotope ratio measurements which can be calculated to < ± 0.1 %.

Further to point 8, it may be suggested that the adding of ^{233}U in a small amount to the input accountability establishes a unique isotopic fingerprint for that batch. The degree to which that fingerprint is distorted will provide previously unavailable insight into the mixing processes which occur during process operations. Again it is subject to measurement capability with "higher abundance sensitivity" mass spectrometers. Such "tagging" technique provides a nuclear material batch (not only "inputs") with an unequivocal "isotopic identity card" which can be monitored by "isotope-specific" measurement at any time during the entire lifetime of the material batch. Other (enriched) isotopes could be used for "tagging" as well.

Conclusions

1. Since it is the fissile isotopes in nuclear fuel that define both the economic and strategic importance of the fuel, thinking about measurements for Safeguards and nuclear material management should concentrate primarily on isotopes.
2. Not only do elemental analyses alone not provide adequate information to define the fissile content of nuclear fuel when applied alone on materials such as depleted U and high burnup Pu, they would not detect "isotope substitution".
3. Because current practice requires both elemental and isotopic analyses, all information required for isotope-specific accountancy exists, but should be better exploited. In particular, calculation of measurement results and their uncertainties should centre on isotope contents, and handle element contents as a secondary product. A real reduction of uncertainty is thereby possible.
4. Isotope-specific accountancy provides a far better visibility of subtle diversion scenarios of fissile isotopes than does elemental accountancy.
5. Isotope dilution measurement techniques provide data for isotope-specific accountancy which is more directly traceable to our internal SI unit of substance (the mole).
6. Emerging measurement technologies provide the potential for eliminating major error terms associated with input accountability measurements in fuel reprocessing facilities, at the same time making the measurements much simpler under plant conditions and reducing the measurement uncertainties.
7. Direct spiking of input tanks does not require (ultra-)pure ^{233}U, nor does the spiking of an input smaple require (ultra-)pure ^{244}Pu. Isotopes already present can be corrected for after a separate measurement of them.
8. Manufacturers of isotope mass spectrometers should take inspiration from the needs and justifications for development as explained here, so as to make available on the commercial market the required instrumentation.
9. Nuclear Reference Material Institutes should actively prepare the required Spike and Isotopic Reference Materials enabling the performance of the above measurements and their calibration.

The answer to the question "Why should we do more Isotope-specific thinking in measurements for Safeguards?" appears to be :

because we can then conceive measurements for Safeguards which are

- simpler and more transparent
- have reduced uncertainty
- are less burdensome for the analyst

thus providing better basic data for isotope specific accountancy in Safeguards.

References

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INTERLABORATORY COMPARISON EXERCISE FOR THE DETERMINATION OF URANIUM BY POTENTIOMETRIC TITRATION
(FIRST PHASE)

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1. Introduction

Upon request of the ESARDA Working Group on "Low Enriched Uranium Conversion and Fuel Fabrication Plants" (ESARDA-LEU WG) the Central Bureau for Nuclear Measurements (CBNM) organized an interlaboratory comparison exercise for the determination of uranium by the potentiometric titration method. The special aim of this intercomparison, apart from giving the laboratories the opportunity to intercompare their results and their individual laboratory deviations from the certified values, is a general asessment of the precision and accuracy of this potentiometric titration method.

The exercise is to be executed in two phases. The first phase dealt with pure uranyl-nitrate solutions; the second phase deals with similar uranium solutions doped with impurities.

The whole exercise is a precision experiment. Methods of measurement are characterized by their precision and their accuracy. Accuracy is defined as the closeness of agreement between the results of a measurement and a reference value. The precision experiment is used as the test method to investigate the type of the variability of the data, and the difference between the results and the certified value(s).

The statistical treatment included:
- calculation of the intra-laboratory variance;
- calculation of the inter-laboratory variance;
- comparison of laboratory means;
- calculation of the repeatability;
- calculation of the reproducibility;
- comparison of the laboratory means with the certified value.

A similar statistical treatment was applied in 1983 for assessing the repeatability and reproducibility of gravimetric uranium determinations in UO_2 pellets /1/.

The following laboratories participated (in alphabetical order of company or institution).

BAM	Berlin	D
CNEN	Rio De Janeiro	BR
ECN	Petten	NL
EIR.	Wuerenlingen	CH
ENUSA	Salamanca	E
FBFC	Dessel	B
FBFC	Romans	F
FBFC	Pierrelatte	F
IAEA	Vienna	A
JRC	Ispra	I
JRC	Karlsruhe	D
NUKEM	Hanau	D
RBU	Hanau	D
SCK	Mol	B

2. Arrangement of the Interlaboratory Exercise

2.1 Sample Distribution.

For the distribution of the samples to be analysed CBNM prepared two solutions of uranyl-nitrate (called A and B) slightly different in concentration. Both solutions had a nominal uranium concentration of about 7.50 mg·g^{-1} solution, which had been certified by CBNM.

Samples of the solutions A and B were distributed in sealed glass ampoules, each containing about 60 ml. Each of the fourteen participating laboratories received simultaneously three ampoules of solution A and three ampoules of solution B. The certified values were not communicated to the participating laboratories before the report of the results.

2.2. Analyses and Submission of Results.

The laboratories were requested to perform the analyses on three different days (here coded with sequence numbers I, II, and III). The first day of analysis should be within two weeks after receipt of the ampoules; the second and third day should follow four and eight weeks later, respectively. The analysis was to be performed immediately after opening the corresponding ampoule.

For the comparison it was necessary to perform the uranium determinations always in pairs: on each of the three days of analysis one ampoule of solution A and one of solution B should be investigated. For each ampoule the analysis should be performed in duplo. Therefore two aliquots of about 20 g of solution had to be taken from each ampoule.

The laboratories were asked to give a detailed description of their procedure, as well as their calibrations results including information about the reference materials used. The results of the uranium determinations (expressed as mg·g^{-1} solution) and the description of the method were sent to CBNM, where they were coded before submission to ECN for the statistical treatment.

2.3. Preparation of the Reference Solutions.

CBNM has in stock a batch of a certified reference material, EC- NRM 110. It consists of sintered uranium dioxide pellets, containing about 1g of uranium each. Information about this material is given in the certification report /2/. Its uranium content has been certified calling on the application of the (100-X) route X being the total of the impurities determined via specific methods.

A known mass of this reference material was carefully dissolved in hot concentrated nitric acid. The solution was evaporated to low bulk and then diluted with 0.1 molar nitric acid until the required concentration (about 7.5 mg U per ml) was obtained. This solution was then homogenized and weighed.

The uranium concentration is expressed in mass of uranium per mass of solution thus resulting in two reference values (7.4911 ± 0.0015 $mg \cdot g^{-1}$ for solution A and 7.7415 ± 0.0015 $mg \cdot g^{-1}$ for solution B). The uncertainties of these values take into account the uncertainty of the reference material and the uncertainties of the different weighings on the basis of a 95% confidence interval.

3. Statistical Treatment

The statistical treatment of the original data set was performed in cooperation with the Netherlands Energy Research Foundation, Petten, Netherlands. The description of the procedure and the detailed results are laid down in an ECN internal report /10/, some additional information is given in an internal report of the Commission of the European Communities /11/.

3.1. Statistical approach.

Many reasons may lead to the variability of data. For this experiment involving many laboratories, factors such as differences in operators, equipment, calibration, environment and sample preparation on different days may give a large contribution to the variability. In practical situations two numerical measures of variability are recommended to take into account differences in the circumstances of new measurements, repeatability and reproducibility. Repeatability refers to the variability of results obtained at short intervals of time in one laboratory by one operator using the same apparatus. Reproducibility refers to the variability of results obtained in different laboratories, which implies different operators, equipment and/or at different times.

For the interpretation of the results obtained with this exercise the analysis of variance technique is a very powerful tool. It gives a good indication of the interlaboratory as well as of the intralaboratory variance and it investigates whether the average outcomes of the laboratories are equal taking into account the experimental uncertainties.

As mentioned earlier two solutions with slightly different uranium concentrations were used. This is known as a split-level experiment. At first, data for both solutions were treated separately as uniform-level experiment, and afterwards the split-level approach was considered.

In summary, the evaluation of the data set comprised the following steps:

- tabulation of data;
- test on homogeneity of the laboratory variances;
- search on the raw data to trace and eliminate wild results;
- analysis of variance;
- determination of repeatability and reproducibility;
- comparison with certified value.

3.2. Data Treatment.

3.2.1. Preliminary Overview. In the tables 1 and 2 the raw data are presented for solution A and B, respectively. The supplementary information as supplied by the laboratories made it clear that only laboratory 13 had incorporated a buoyancy correction in its results. Since this correction had been applied in the determination of the certified uranium concentration of solution A and B the results coming from the other laboratories were also corrected for this effect (0.105%).

Lab Code	Data Sequence Code			Lab Mean	Standard Deviation s (·10-3)	Variance s^2 (·10 6)	Coefficient of Variation v(%)
	I	II	III				
1	7.4901 7 4991	7 5001 7.4951	7.5021 7 5021	7.4981	4.69	22 00	0.063
2	7.4881 7.4901	7 4901 7.4911	7.4911 7.4911	7.4903	1.16	1 350	0.016
3	7.4956 7 4959	7 4936 7.4953	7.4945 7 4939	7 4948	0 955	0 912	0 013
4	7.4662 7.4971	7 4741 7 4662	7.4691 7 4851	7.4763	12 42	154 2	0 166
5	7.4960 7 5028	7.4938 7.4952	7.5001 7.4953	7.4972	3 47	12 05	0 046
6	7 5211 7 5061	7.5211 7 4981	7.5211 7 5031	7 5118	10 5	111 1	0.140
7	7.4931 7 4943	7 4942 7.4936	7.4910 7.4895	7.4926	1.95	3 78	0 026
8	7 4911 7.4911	7.4891 7.4941	7 4891 7.4891	7.4906	1.97	3.89	0.026
9	7.4115 7.4377	7.4739 7 4026	7 4757 7 4370	7.4397	30 5	929.7	0.410
10	7.5001 7 4981	7.4911 7.4941	7 4931 7.4931	7 4949	3.43	11.75	0.046
11	7.5021 7.4921	7.4961 7.5021	7.5061 7.5011	7.4998	4 76	22 65	0.063
12	7.6919 7 5920	7 6919 7.5920	8.2913 7 8917	7.7918	268 1	71856 1	3.44
13	7.4899 7 4911	7 4926 7.4936	7.4829 7.4849	7.4892	4.32	18 65	0.058
14	7.4801 7.4921	7.4891 7 4951	7.4991 7.5031	7 4931	8 07	65 19	0.108
				overall solution A			
mean	7.4999	7.5000	7.5345	7.5114			
S(·10-3)	47 13	46.61	167 1	103 8			
S^2(·10 6)	2221.6	2172.1	27919 6	10780 1			
v(%)	0 628	0.621	2 22	1 38			

Table 1: Data for solution A.

Lab Code	Data Sequence Code			Lab Mean	Standard Deviation s (·10^{-3})	Variance s^2 (·10^{-6})	Coefficient of Variation v(%)
	I	II	III				
	7 7449	7 7558	7.7598				
1				7 7553	6 79	46 13	0 088
	7 7648	7.7519	7 7548				
	7.7399	7.7409	7 7419				
2				7 7407	1.16	1 355	0 015
	7.7389	7 7409	7 7419				
	7.7494	7.7485	7 7465				
3				7 7482	1 61	2 29	0 020
	7 7502	7 7481	7 7465				
	7.7099	7 7289	7.6989				
4				7.7176	13 94	194.3	0 181
	7.7169	7 7129	7 7379				
	7.7377	7 7421	7.7493				
5				7 7417	6 41	41 08	0 083
	7.7405	7 7324	7.7483				
	7.7359	7 7489	7.7469				
6				7.7489	8 71	75.84	0 112
	7.7628	7.7519	7 7469				
	7.7469	7.7466	7.7434				
7				7 7457	2 42	5 87	0 031
	7 7487	7 7464	7 7422				
	7 7449	7.7459	7 7469				
8				7 7452	1 36	1.85	0.018
	7.7459	7.7429	7 7449				
	7.6937	7.7124	7.8807				
9				7.7597	79 00	6241.6	1.02
	7.6816	7.7686	7 8213				
	7 7489	7 7419	7.7449				
10				7.7459	4.00	15.99	0 052
	7.7479	7.7409	7 7509				
	7.7658	7.7608	7 8438				
11				7 7815	49 4	2437.6	0.634
	7 8428	7.7439	7 7319				
	7.7918	7 9916	7.8917				
12				8 0083	299 1	89487.4	3 74
	7 7918	7.9916	8 5910				
	7.7491	7.7461	7.7443				
13				7 7458	2 32	5.41	0.030
	7 7467	7 7422	7 7461				
	7.7479	7.7538	7 7509				
14				7 7474	6 94	48 16	0 090
	7 7399	7.7379	7 7538				
				overall solution B			
mean	7.7474	7 7613	7.7910	7.7666			
S(·10^{-3})	29 79	66.10	162.7	103 2			
S^2(·10^{-6})	887 59	1368 8	26475 7	10657 8			
v(%)	0.385	0.852	2 09	1 33			

Table 2: Data for solution B.

To get a first impression, some characteristic data were calculated such as mean, standard deviation, variance and coefficient of variation. The average results of the measurements found by each laboratory are illustrated in the graphs of figure 1 and figure 2 for both solutions. A general inspection shows that the data contain several deviating values in each of the solutions. Some values among the original results (for example those from laboratory 12) deviate so much from comparable entries in the same tables, that special attention was given to the search of outlying data, at first on a graphical basis, to get a rough, preliminary insight.

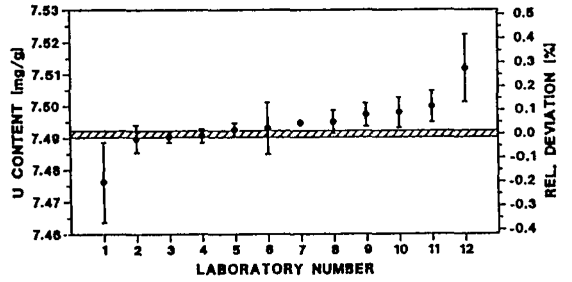

Fig. 1. Data for solution A (buoyancy corrected, coding different from text).

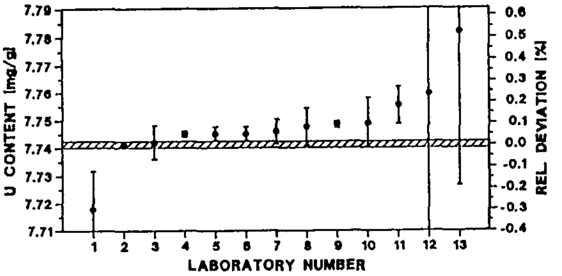

Fig. 2. Data for solution B (buoyancy corrected, coding different from text).

This was performed by means of a Youden diagram /3, 4/, where the results of the measurements for two series (A and B) constitute the co-ordinates of points in a plot. One pair of results yields a single point in the diagram, furthermore horizontal and vertical lines denoting the median values are presented based on all results in a series.

In an ideal situation the points are expected to constitute an elliptic cluster around the point with median value co-ordinates. Because the two solutions are nearly equal in uranium concentration, it is possible to have an indication of the precision of the data in the figures by means of a circle (centered on the intersection of the median lines). A radius of about 2.5 to 3.0 times the standard deviation gives the smallest region that can be expected when a normal distribution approximately applies.

The graphical presentations show that at least one laboratory (coded 12) yields deviating results (figures 3a, 3b, 3c). Suspected data from other laboratories such as 9 and 11 look also incompatible with other entries. In these cases numerical outlier and straggler tests have been applied. Since it is necessary to handle deviating results with care, and since various outlier tests have different levels of discrimination, a combination of several tests was chosen.

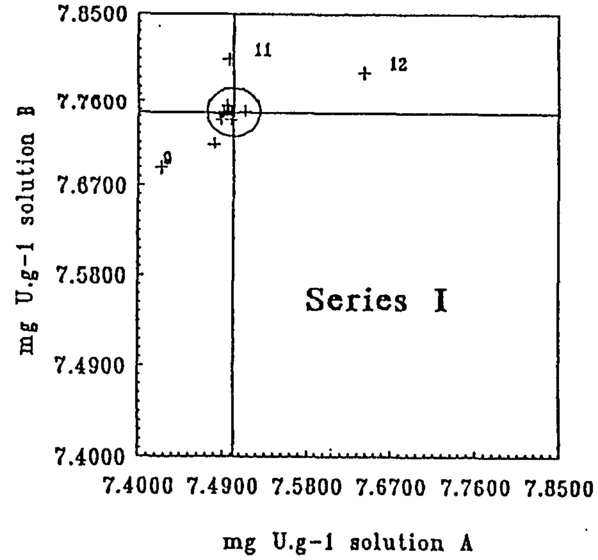

Fig. 3a. Youden diagram solution A versus B (average data laboratories series I).

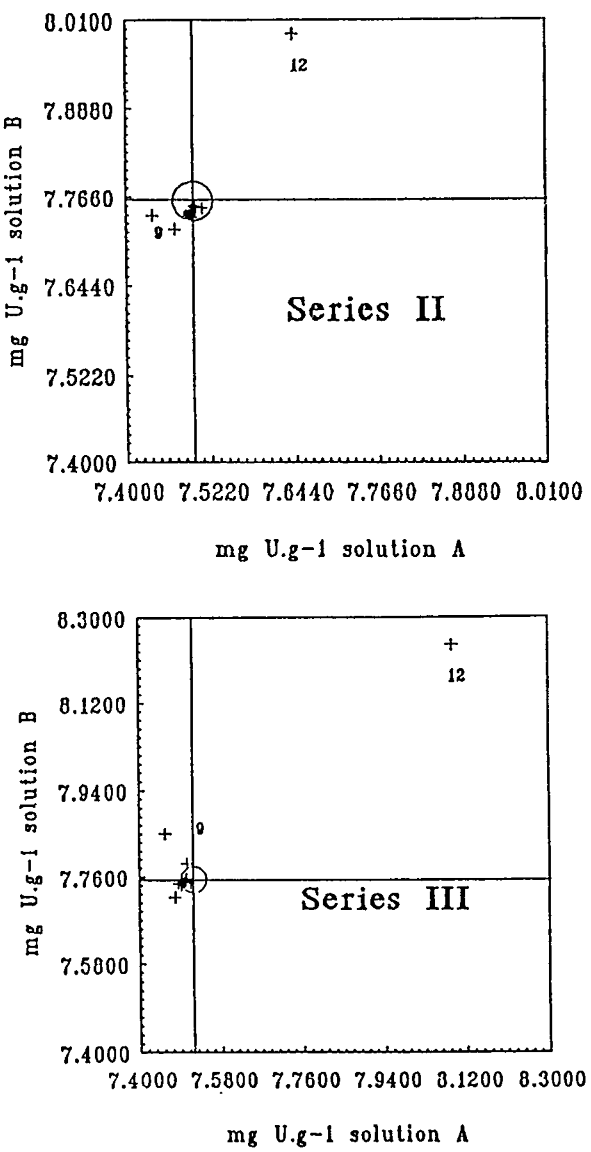

Fig. 3b. and c. Youden diagram solution A versus B (average data laboratories series II and III).

Experience had shown that between the laboratories large differences exist in the intra-laboratory variances (see tables 1 and 2). Therefore, Cochran's maximum variance test seemed very useful /6/, investigating the significance of the difference between one large variance value and the other remaining variance values in a set of data. It considers only the highest value in a set of ranges and is therefore a one-sided outlier test. Small values of range may be strongly influenced by the degree of rounding of the original results and are for that reason not very reliable. Due to the present lack for recommendations of statistical tests designed for simultaneous examination of the presence of several outliers, the procedure of repeated application of the single outlier test was applied.

The next step in the analysing of the data set was the application of the Dixon test /7/. A summary of the most important results of these tests is presented in table 3 (using laboratory and sequence codes).

Sol.	Cochran Test		Dixon Test	
	Outlier	Straggler	Outlier	Straggler
A	9II; 9III; 12I, 12II; 12III; 6II	-	4II; 9I; 9II; 12I; 12II; 12III	6I; 6II
B	4III; 9II; 9III; 11I; 11III; 12III	6I	4I; 4III; 9I; 9III; 11I; 11III; 12I; 12II; 12III	4II

Table 3: Results of Cochran and Dixon test.

Examination of these results makes it clear that statistical outliers are present for the laboratories coded 9 and 12 at nearly all levels. Therefore it was concluded that it was justified to discard all the data coming from laboratories 9 and 12.

After this deletion still some statistic outliers remain on the results of solution A and B at single levels for the laboratories coded 6, 4 and 11. Due to the repeated application of the tests and the fact that these outliers occur only at single levels it was not thought to be justified to discard these data at this stage of the analysis. Also all stragglers were kept in the further analysis due to the fact that repetition of the tests makes decisions of removal questionable.

3.2.2. Analysis of Variance. The analysis of variance method investigates the significance of differences between the laboratory outcomes. For more details of the calculations see reference /10/ and /11/.

The results for solution A are presented in table 4. The large differences in the sample variances between the original and cleaned data sets confirm earlier conclusions concerning the presence of the outliers for the laboratories 9 and 12. The test on main effects (labs) was done leading to the conclusion that the expected laboratory means are not equal.

	all data N=84	Deleted labs	
		9 and 12 N=72	9, 12, 4 and 6 N=60
Source of variation	Sample Variance ($\cdot 10^{-4}$)		
between labs	406.6812	4.0490	0.7558
between days within labs	96.8378	0.2795	0.2602
residual	22.6002	0.4074	0.0970
total	107.8014	0.9284	0.2528
mean	7.5114	7.4941	7.4941

Table 4: Analysis of variance (sol A).

The results for solution B are presented in table 5. The test on the presence of inter-laboratory effects leads to the same conclusions as for solution A: the laboratory mean values are not equal.

	all data N=84	Deleted labs	
		9 and 12 N=72	9, 12, 4 and 6 N=60
Source of variation	Sample Variance (·10-4)		
between labs	301.2114	12.0322	0.9776
between days within labs	83.5630	1.3576	0.1901
residual	61.6781	3.0892	0.2800
total	106.5782	3.8894	0.3559
mean	7.7666	7.7470	7.7465

Table 5: Analysis of variance sol. B).

In accordance with the approach for chosing the concentrations for this precision experiment the split-level analysis is suitable. The results are presented in table 6.

Analysis of variance for solution A and B (random effects ; laboratories 9 and 12 deleted ; N = 72)			
source of variation	sum of squares (·10 ⁴)	degrees of freedom	sample variance (·10 ⁴)
between labs	68.4480	12-1 = 11	6.2225
between days within labs	10.3290	12·(3-1) = 24	0.4304
residual	31.6738	72-12·3 = 36	0.8798
total	110.4508	72-1 = 71	1.5556

Table 6: Analysis of variance (split level).

It is shown that earlier conclusions concerning main effects for solution A and B separately, are still valid as well as the conclusion that at least one laboratory mean value deviates significantly from the remaining mean values. In neither of the three parts of variance analysis a time effect was proved to be present.

The next step was a further investigation of the laboratory effect, i.e. of the question which laboratories show mean results differing significantly from other laboratory means. Therefore, the test of Link-Wallace was applied /8/, excluding a priori the laboratories 9 and 12. The conclusion was found justifiable that for solution A the laboratories 4 and 6 give significantly deviating means, and for solution B the laboratories 4 and 11.

The repeatibility r and the reproducibility R results are shown in table 7.

	Solution	r	R	m
Uniform Level	A B	0.0169 0 0438	0.0279 0.0566	7.4941 7.7470
Split Level	both A and B	0.0237	0.0360	7.6205

Table 7: Repeatibility and reproducibility results.

3.2.3. Distribution of Differences. The evaluation so far was based on the analysis of variance technique. A possible drawback of the straight-forward application of conventional definitions for averages as well as for standard deviations is that extreme data can influence the results of these calculations seriously. For this reason, and in particular concerning the interlaboratory standard deviation, a comparison with the "Distribution of Differences" method (DoD) /9/ between the results was of interest.

The interlaboratory standard deviation ("s_{DoD}") was calculated for the data sets of solution A and B, as well as for the smaller data set with only average laboratory results for each solution and the results are shown in figures 4 and 5. To investigate the effect of the outlying laboratory 12 the plots are shown for the two situations with and without this laboratory.

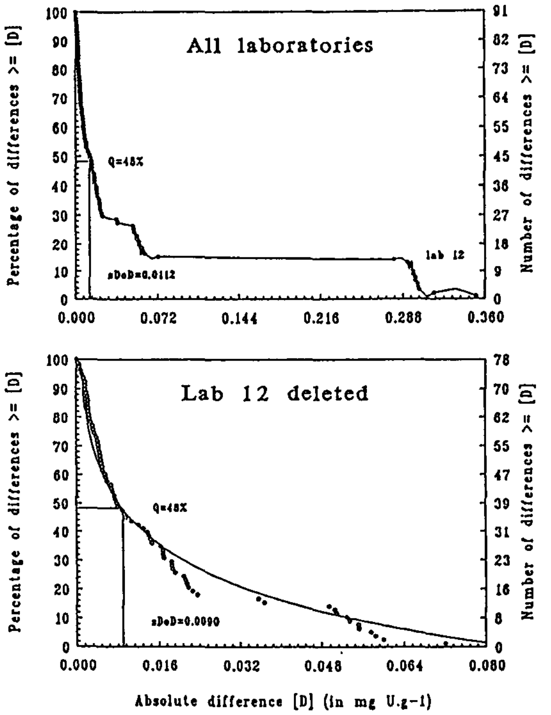

Fig. 4 Distribution of differences with fit using average data for solution A (48 % value indicates the estimated s_{DoD} of the interlab standard deviation).

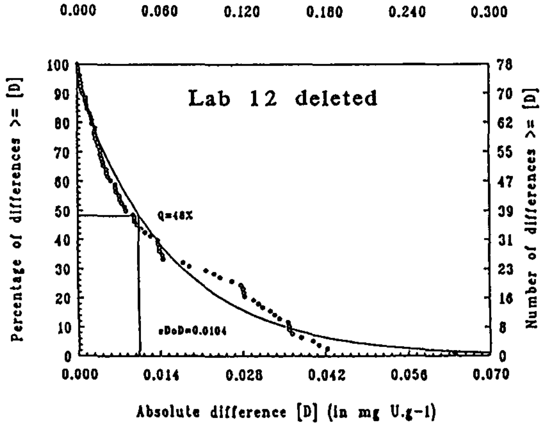

Fig. 5 Distribution of differences with fit using average data for solution B (48 % value indicated the estimated s_{DoD} of the interlab standard deviation).

The numerical value for S_{DoD} remains the same for the two situations. The comparison of the values for s_{DoD} and s_{tot} is given in table 8.

Quantity Calculated	Situation	Sol. A	Sol. B
s_{DoD}	All data involved Average lab data	0 0112 0.0135	0.0143 0.0134
Conventional S_{tot}	without outliers	0.0096	0.0197

Table 8: Comparison of s_{DoD} and s_{tot}.

It is noteworthy that for solution B the conventional standard deviation s_{tot} is appreciably higher than s_{DoD}. This indicates that in case of solution B still extreme values are present in the data set. The results show that all standard deviations have more or less the same order of magnitude.

4. Discussion

4.1. General.

The method most often used was the potentiometric titration by Davies and Gray. There were variations of the working procedure, for instance according to the modification by J. Cherry or the use of the method calling upon addition of vanadium as described by A.E. Eberle et.al. Other variations (concentration and volume of reagents, intervals of time between the successive steps of the chemical reactions, approach towards the end-point) were as well present. Some laboratories have used automatic titrators, some others did not mention the type of equipment.

All the reference materials used, except in the case of one laboratory (metal chips), seem to be valuable reference materials. Nevertheless some of them have better confidence limits attached to their certified value than others because of a more elaborate characterization. The handling of the reference material by the laboratory might influence the quality of the reference solution, for some of these materials need a preliminary treatment like calcination or cutting of a large piece followed by a chemical cleaning of the surface. The non adherance to strict rules when dissolving the material (avoiding loss of material when heating solutions, contaminations, etc.) may also influence the accuracy of the calibration.

4.2. Comparison with Certified Values.

In tables 9 and 10 a comparison is presented between the average values found by each laboratory and the certified values for both solutions (A and B). As shown in these tables the average absolute deviation from the certified value /d/ as well as its standard deviation o/d/ were also calculated with and without the data of the outlying laboratories 9 and 12. These tables show that the deviations of a lot of laboratory mean values with respect to the certified values are larger than the standard deviation stated for certification.

Lab Code	Standard		mean (in mg U g⁻¹)	d_i deviation from certified val (7.4911) (in mg U·g⁻¹)	$\frac{d_i}{\delta_R}$ *
	Code	Material			
1	NBS 960	U metal	7.4981	+0 0070	0.710
2	-	UO_2	7 4903	-0 0008	-0 0812
3	EC 110	UO_2	7.4948	+0 0037	0 375
4	NBL 97	U_3O_8	7.4763	-0 0148	-1 50
5	EC 110	UO_2	7 4972	+0 0061	0 619
6	NBS 950a	U_3O_8	7.5118	+0.0207	2 10
7	NBS 960	U metal	7.4926	+0 0015	0.152
8	NBS 960	U metal	7.4906	-0.0005	-0.0507
9	CEA F1 Anemone	U metal	7.4397**	-0 0514**	-5.21
10	NBS 960	U metal	7 4949	+0 0038	0.385
11	NBS 960	U metal	7.4998	+0 0087	0.883
12	NBS 950b	U_3O_8	7 7918**	+0 3007**	30 5
13	EC 101	U metal	7.4892	-0 0019	-0 193
14	NBS 960	U metal	7.4931	+0 0020	0 203
$\|d\|$ all laboratories $\sigma_{\|d\|}$ all laboratories				0 03026 $\frac{\|d\|}{\sigma_{\|d\|}}$ = 0 383 0 07898	
$\|d\|$ without labs 9 and 12 $\sigma_{\|d\|}$ without labs 9 and 12				0 005958 $\frac{\|d\|}{\sigma_{\|d\|}}$ = 0.962 0.006196	

* δ_R = 0 009857
** outliers

Table 9: Comparison with certified value for solution A.

Lab Code	Standard		mean (in mg U·g⁻¹)	d_i deviation from certified val (7 7415) (in mg U g⁻¹)	$\frac{d_i}{\delta_R}$ *
	Code	Material			
1	NBS 960	U metal	7.7553	+0 0138	0 690
2	-	UO_2	7.7407	-0.0008	-0 0400
3	EC 110	UO_2	7.7482	+0 0067	0 335
4	NBL 97	U_3O_8	7.7176	-0 0239	-1 20
5	EC 101	UO_2	7.7417	+0.0002	0 010
6	NBS 950a	U_3O_8	7 7489	+0 0074	0 370
7	NBS 960	U metal	7.7457	+0 0042	0 210
8	NBS 960	U metal	7 7452	+0 0037	0.185
9	CEA F1 Anemone	U metal	7.7597**	+0 0182**	0 910
10	NBS 960	U metal	7.7459	+0.0044	0 220
11	NBS 960	U metal	7.7815**	+0 0400**	2.00
12	NBS 950b	U_3O_8	8.0083**	+0.2668**	13 3
13	EC 101	U metal	7 7458	+0 0043	0.215
14	NBS 960	U metal	7.7474	+0 0059	0 295
$\|d\|$ all laboratories $\sigma_{\|d\|}$ all laboratories				0 02859 $\frac{\|d\|}{\sigma_{\|d\|}}$ = 0 412 0.06941	
$\|d\|$ without labs 9 and 12 $\sigma_{\|d\|}$ without labs 9 and 12				0.009608 $\frac{\|d\|}{\sigma_{\|d\|}}$ = 0 835 0.001151	

Table 10: Comparison with certified value for solution B.

For calibration purposes of the titration method used, verification by comparison with an appropriate internationally recognized uranium reference material is necessary. Part of the calibration procedure is the dissolution of reference material. Considering a possible dissolution effect the results were clustered according to the type of reference material used (table 11a). Based on these data the general idea arose that the use of uranium oxide as reference material causes roughly a two or three times higher standard deviation.

Also the laboratories using a NBS 960 or an EC standard were investigated (table 11b). The laboratories using the EC reference materials show for both solutions slightly smaller deviations from the certified values than the laboratories using the NBS reference material. Nevertheless this difference can not be considered as significant enough to justify a conclusion.

	Material		Mean	s_{tot}
Solution A	U metal	inclusive outliers	7.4873	0.0195
		exclusive outlier (lab 9)	7.4940	0.0038
	U oxide	inclusive outliers	7.5437	0.122
		exclusive outlier (lab 12)	7.4941	0.0128
Solution B	U metal	Inclusive outliers	7.7520	0.0124
		exclusive outlier (lab 11)	7.7483	0.0060
	U oxide	inclusive outliers	7.7927	0.121
		exclusive outlier (lab 12)	7.7389	0.0146

Table 11a: Results arranged by reference material.

	Standard	Deviation from cert. value	Mean	s_{tot}	No. Labs.
Solution A	EC 110; 101	+0.0026	7.4937	0.00410	3
	NBS 960	+0.0038	7.4949	0.00350	6
Solution B	EC 110;101	+0.0037	7.7452	0.00328	3
	NBS 960	+0.0120	7.7535	0.0124	6
	NBS 960 excl. Lab. 11	+0.0064	7.7479	0.0042	5

Table 11b: Results arranged by reference material origin.

4.3. Statistical Evaluation.

The original data set consisting of 84 observations originating from 14 laboratories comprises for nearly all determinations on solution A as well as solution B statistical outliers. It was found justified to discard all the data coming from laboratories 9 and 12. This conclusion is demonstrated in the summary of results given in table 12.

		Uniform level		Split Level
		Sol. A	Sol. B	Sol. A and B
original data set	m	7.5114	7.7666	
	s^2	0.0108	0.0107	
	s	0.1038	0.1032	
	v (in %)	1.38	1.33	
cleaned data set (without labs 9 and 12)	m	7.4941	7.7470	7.6205
	s^2	0.000093	0.000389	0 000156
	s	0.00964	0.01972	0.01247
	v (in %)	0.129	0.255	0.164
Repeatability	r	0.0169	0.0438	0.0237
Reproducibility	R	0.0279	0.0566	0.0360

Table 12: Summary of results.

The overall variances for the complete data sets of each solution separately are roughly 10 times larger than those of the clean data sets (without the outliers). These sets do not show significant deviations from the normal distribution of Laplace-Gauss.Furthermore, the existence of a time effect is not proved.

The following overall conclusions can be drawn :

- The best estimate for the repeatability standard deviation is 0.0060 (i.e. relatively 0.080 %) for solution A, 0.0155 (i.e. 0.20%) for solution B, and for the split-level approach 0.0084 (i.e. 0.110%).
- The best estimate for the inter-laboratory standard deviation is 0.0079 (i.e. relatively 0.105%) for solution A, 0.0127 (i.e. 0.164%) for solution B, and for the split level approach 0.0096 (i.e. 0.126%).
- The best estimate for the reproducibility standard deviation is 0.0099 (i.e. relatively 0.132%) for solution A, 0.0200 (i.e. 0.258%) for solution B, and for the split-level approach 0.0127 (i.e. 0.167%).

It may further be concluded that a relative standard deviation of about 0.25% describes very well the total reproducibility variability in the interlaboratory comparison. An experienced laboratory will have a within-laboratory standard deviation as low as 0.08%.

The conclusions above concern the cleaned data sets (without the results from the laboratories 9 and 12), and are drawn confirm the recommendations in the report ISO 5725 /5/.

The test of Link-Wallace was applied to find indications about laboratories with significant deviations. A further reduction of the data sets by excluding the results of the laboratories 4(sol A and B), 6(sol A) and 11(sol B) was not allowed, because outliers occur not on all levels. Such an action would not affect the overall conclusions but the variances would be reduced importantly.

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RESULTS OF REIMEP '89
UO_2 POWDER

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Abstract

The interest in the safeguards of fissile material focuses on a limited number of compounds which play key roles in the nuclear fuel cycle. Amongst these materials uranium dioxide powder is of considerable importance as it is the base material for pellet production. In LWR's uranium dioxide with an initial ^{235}U content of about 3 mass % is used, whereas natural or depleted material is applied for the breeding zone in FBR's.

The 89/90 round of REIMEP covered uranium materials with ^{235}U abundances in the range of natural or depleted material. UO_2 powder was distributed to 20 laboratories for destructive analysis. Two laboratories participated with non-destructive methods.

The participating laboratories were asked to determine the uranium content and the isotopic composition of the material. The results reported by the participants are presented as graphs thus giving a picture of the 'state-of-the-practice'.

1. Introduction

In the frame of the REIMEP programme in 1989 a UO_2 powder was distributed. Through this CBNM intends to provide nuclear analytical laboratories with a regular opportunity

- for an external check on the quality of their measurements
- to establish the state-of-the-practice of assaying common nuclear materials for isotope and element content
- to present a true picture of the real life interlaboratory spread of given measurements on given materials.

This paper reports on the first UO_2 powder round in REIMEP.

All participating laboratories reported values obtained by destructive analysis. The choice of the method was up to the participants.

2. Participation List

1. Martin Marietta Energy Systems
   Piketon,Ohio, USA,Mr. Akers
2. Martin Marietta Energy Systems
   Paducah, Kentucky, USA, Mr. Beverly
3. Los Alamos National Laboratory
   Los Alamos, NM, USA, Mr. Abernathey
4. WAK
   Karlsruhe, Germany, Mr. Stojanik
5. ENEA
   Saluggia, Italy, Mr. Pagliai
6. NMCC
   Tokai-Mura, Japan, Mr. Nomura
7. ENUSA
   Salamanca, Spain, Mr. Marin
8. BNF
   Springfields, UK, Mr. Hayes
9. New Brunswick Laboratory
   Argonne, Ill., USA, Mr. Lewis
10. Inst. for Energy Techn.
    Kjeller, Norway, Mr. Stijfhoorn
11. Belgonucléaire
    Dessel, Belgium, Mr. Eeckhout
12. University of Helsinki
    Helsinki, Finland, Ms. Keinonen
13. ECN
    Petten, Netherlands, Mr. Woittiez
14. FBFC
    Dessel, Belgium, Mr. Pauwels
15. IAEA/SAL
    Seibersdorf, Austria, Mr. Deron
16. TUI
    Karlsruhe, Germany, Mr. Koch
17. JRC
    Ispra, Italy, Mr. Guzzi
18. BAM
    Berlin, Germany, Mr. Tamberg
19. CEN
    Grenoble, France, Ms. Andreani
20. SCK
    Mol, Belgium, Mr. De Regge
21. ECN
    Petten, Netherlands, Mr. Aaldijk
22. AERE
    Harwell, UK, Mr. Brown

The numbers do not correspond to the laboratory codes mentioned in the graphs.

3. Preparation, Verification of Homogeneity and Certification of Element Content

3.1 Preparation

A batch of UO_2 powder was obtained from a commercial nuclear fuel fabrication facility. Portions of about 10 g of material were filled in pre-weighed bottles. After several days of storage under controlled conditions (relative humidity, temperature, pressure) the bottles were closed and weighed again.

3.2 Verification of homogeneity and certification of element content

The uranium mass fraction was obtained through direct uranium measurements by means of potentiometric titration calibrated against EC-NRM 101 (uranium metal) and controlled potential coulometry calibrated against EC-NRM 110 (uranium oxide).

The potentiometric titration was applied to three bottles of UO_2, the respective results were within each others uncertainty. Controlled potential coulometry was applied to two bottles, here again the individual results were within each others uncertainty.

The uranium element content was certified as the weighted mean of uranium contents found respectively by two methods. The following value was obtained :

859.91 ± 0.36 $g \cdot kg^{-1}$.

A verification of this value was performed by the so-called (100-X) route, i.e. the determination of the total impurity content expressed as oxide and subtracted from 100 percent. This was applied independently to five bottles yielding a mean value of

859.97 ± 0.12 $g \cdot kg^{-1}$.

4. Certification of Isotopic Composition

4.1 Determination of the $^{235}U/^{238}U$ ratio

In order to minimise the uncertainty on the $^{235}U/^{238}U$ ratio, this figure was determined by UF_6 gas mass spectrometry. The conversion of the uranium oxide to UF_6 was done in the following way.

The UO_2 powder was dissolved in excess HNO_3, the solution was then evaporated to dryness. The residue was heated to 350°C overnight. The resulting $UO_3 \cdot xH_2O$ was heated to 250°C in vacuum for 35 minutes. The resulting UO_3 was treated in vacuum at 250°C for 35 minutes in order to remove the water. The waterfree UO_3 was then mixed with CoF_3 and the mixture degassed in vacuum at 200°C for 30 minutes. Subsequent heating to 425°C for 70 minutes and condensation of the reaction product UF_6 at -196°C. The UF_6 was purified at -78°C and then homogenized at 150°C during 60 minutes.

For the determination of the $^{235}U/^{238}U$ molar isotope ratio of the sample, the double standard method was applied. The two isotope reference materials (IRM) are chosen such that:

$$\frac{^{235}U/^{238}U\ (sample)}{^{235}U/^{238}U\,(IRM_1)} > 1 > \frac{^{235}U/^{238}U\ (sample)}{^{235}U/^{238}U\,(IRM_2)}$$

The $^{235}U/^{238}U$ molar isotope ratio was determined as :

0.007 283 4 ± 0.000 002 9

4.2 Determination of the Isotopic Composition

The minor abundant isotopes ^{234}U and ^{236}U as well as the major compmonents ^{235}U and ^{238}U were measured by thermal ionisation mass spectrometry.

For this purpose several fractions of the powder were dissolved in nitric acid, evaporated to dryness and redissolved in 1 molar nitric acid. The uranium concentration was adjuested to about 5 $mg \cdot g^{-1}$. The measurements were performed on a Finnigan MAT 260 instrument. Parallel measurements on a Finnigan MAT 262 equipped with a decelerating system for enhanced abundance sensitivity and an ion counter were made in order to determine the minor abundant isotopes more accurately.

The measurements were calibrated against synthetic isotope mixtures: the isotope reference materials EC 183 to EC 186. The uncertainties on the reference materials were taken into account for the final accuracy statement.

4.3 Calculation of the isotopic composition

Using the calibrated $^{234}U/^{235}U$ and $^{236}U/^{235}U$ ratios determined by thermal ionisation mass spectrometry and the result of the gas mass spectrometric determination of $^{235}U/^{238}U$, the reference values for the isotopic composition of the sample were calculated.

The accompanying uncertainties were used for the calculation of the final uncertainty statement.

The following values of the isotopic composition were certified :

^{234}U	0.005 13 ± 0.000 06
^{235}U	0.713 97 ± 0.000 29
^{236}U <	0.000 10 ± 0.000 00
^{238}U	99.280 90 ± 0.000 29

5. Results

Basing on the results reported by the participants, graphs were worked out which give a picture of the state of the practice. These graphs are shown on the following pages.

In addition to the graphs we report also the interlaboratory spread as calculated by the DoD (Distribution of Differences) method /1/ and the interlaboratory mean. The median value of the reported results is also mentioned.

6. Discussion

The analysis of uranium dioxide powder requires handling, weighing and dissolving of powder material.

The graph displaying the results of the element assay shows that the capability for this determination is for the majority of laboratories under good control. The values indicated in brackets could not be corrected for weight change during transport, thus the values appear somewhat too low.

The determination of ^{235}U and ^{238}U abundances showed that some of the laboratories seem to have problems with the $^{235}U/^{238}U$ ratio measurement. These laboratories reported too high ^{235}U and too low ^{238}U abundances as compared to the reference values.

It seems that not all the laboratories do estimate their accuracy realistically.

The standard deviations on the interlaboratory means were determined using the DoD (Distribution of Differences) method /1/.

The DoD software was worked out by Kernforschungszentrum Karlsruhe.

The external quality control programme REIMEP is under sponsorship of the Euratom Safeguards Authorities, Commission of the European Communities, DG XVII, DCS.

References

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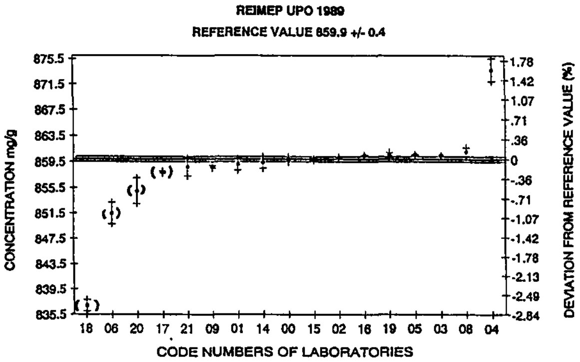
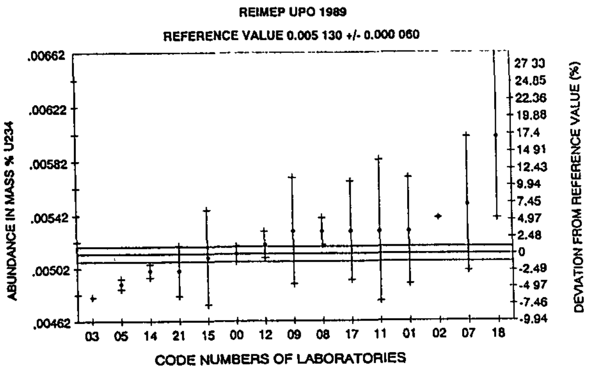
Uranium Element Assay

Reference Value: 859.91 ± 0.4 mg·g^{-1}
Interlab. Mean : 858.48 ± 2.2 mg·g^{-1}
Median Value : 859.8 mg·g^{-1}



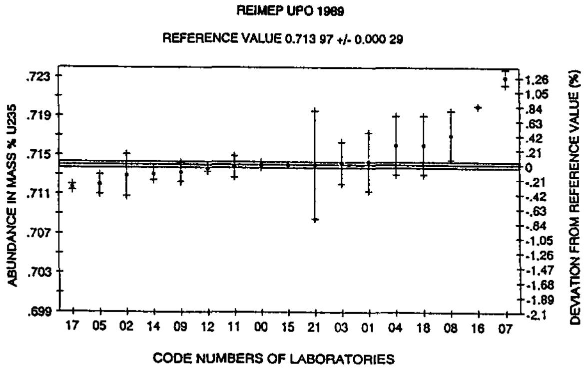
^{234}U **Abundance Determination**

Reference Value: 0.005 130 ± 0.000 060 mass %
Interlab. Mean : 0.005 24 ± 0.000 20 mass %
Median Value : 0.005 3 mass %



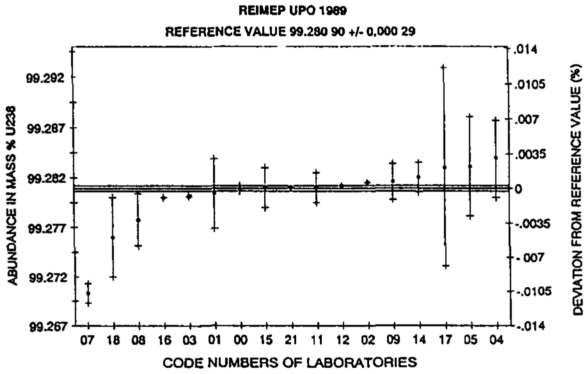
^{235}U **Abundance Determination**

Reference Value: 0.713 97 ± 0.000 29 mass %
Interlab. Mean : 0.713 9 ± 0.002 4 mass %
Median Value : 0.714 0 mass %



^{238}U **Abundance Determination**

Reference Value: 99.280 90 ± 0.000 29 mass %
Interlab. Mean : 99.280 0 ± 0.001 8 mass %
Median Value : 99.280 7 mass %



RESULTS OF REIMEP '89
UO_2 PELLET

K. Mayer, A. Alonso, P. De Bièvre, W. Lycke,
W. De Bolle, M. Gallet, F. Hendrickx

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Joint Research Centre
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Abstract

The interest in the safeguards of fissile material focuses on a limited number of compounds which play key roles in the nuclear fuel cycle. Amongst these materials uranium dioxide pellets are of considerable importance as they enter the reactors in order to generate energy. In LWR's pellets with an initial ^{235}U content of about 3 mass % are used, whereas natural or depleted material is applied for the breeding zone in FBR's.

The 89/90 round of REIMEP covered uranium materials with ^{235}U abundances in the range of natural or depleted material. UO_2 pellets were distributed to 21 laboratories for analysis.

The participating laboratories were asked to determine the uranium content and the isotopic composition of the material. The results reported by the participants are presented as graphs thus giving a picture of the 'state-of-the-practice'.

1. Introduction

In the frame of the REIMEP programme in 1989 a uranium dioxide material was distributed. Through this CBNM intends to provide nuclear analytical laboratories with a regular opportunity

- for an external check on the quality of their measurements
- to establish the state-of-the-practice of assaying common nuclear materials for isotope and element content
- to present a true picture of the real life interlaboratory spread of given measurements on given materials.

This paper reports on the first UO_2 pellet round in REIMEP. Although a number of laboratories received both samples for destructive analysis (DA) and non-destructive analysis (NDA) only one laboratory reported a NDA value.

2. Participation List

1. Martin Marietta Energy Systems
   Piketon,Ohio, USA, Mr. Akers
2. Martin Marietta Energy Systems
   Paducah, Kentucky, USA, Mr. Beverly
3. New Brunswick Laboratory
   Argonne, Ill., USA, Mr. Lewis
4. Los Alamos National Laboratory
   Los Alamos, NM, USA, Mr. Abernathey
5. NMCC
   Tokai-Mura, Japan, Mr. Nomura
6. Inst. for Energy Techn.
   Kjeller, Norway, Mr. Stijfhoorn
7. IAEA
   Seibersdorf, Austria, Mr. Deron
8. BNF
   Capenhurst, UK, Mr. Ogden
9. BNF
   Salwick, UK, Mr. Hayes
10. ENEA
    Saluggia, Italy, Mr. Pagliai
11. FBFC
    Dessel, Belgium, Mr. Pauwels
12. Belgonucléaire
    Dessel, Belgium, Mr. Eeckhout
13. WAK
    Karlsruhe, Germany, Mr. Berg
14. ENUSA
    Salamanca, Spain, Mr. Marin
15. JRC
    Ispra, Italy, Mr. Guzzi
16. SCK
    Mol, Belgium, Mr. De Regge
17. ECN
    Petten, Netherlands, Mr. Woittiez
18. TUI
    Karlsruhe, Germany, Mr. Koch
19. BAM
    Berlin, Germany, Mr. Tamberg
20. AERE
    Harwell, UK, Mr. Brown
21. CEN
    Grenoble, France, Ms. Andreani

The numbers do not correspond to the laboratory codes mentioned in the graphs.

3. Preparation, Verification of Homogeneity and Certification of Element Content

The material used for this REIMEP round was the CBNM Reference Material No. 106. Preparation, homogeneity tests and determination of uranium mass fraction are described extensively in the certification report, a copy of which is available from CBNM/1/.
The certified value of the element content is :
0.881 43 ± 0.000 24 $g \cdot g^{-1}$ of material

4. Certification of Isotopic Composition

4.1 Determination of the $^{235}U/^{238}U$ ratio

In order to minimise the uncertainty on the $^{235}U/^{238}U$ ratio, this figure was determined by UF_6 gas mass spectrometry.

The conversion of the uranium oxide to UF6 was done in the following way.

A pellet was dissolved in an excess of

HNO_3, the solution was then evaporated to dryness and the residue heated to 350°C overnight.

The resulting $UO_3 \cdot xH_2O$ was heated to 250°C in vacuum for 35 minutes. The waterfree UO_3 was then mixed with an excess of CoF_3, the mixture degassed in vacuum at 200°C for 30 minutes. Subsequent heating to 425°C for 70 minutes and condensation of the reaction product UF_6 at -196°C. The UF_6 was purified at -78°C and then homogenized at 150°C during 60 minutes.

For the determination of the $^{235}U/^{238}U$ molar isotope ratio of the sample, the double standard method was applied. The two isotope reference materials (IRM) are chosen such that:

$$\frac{^{235}U/^{238}U\ (sample)}{^{235}U/^{238}U\,(IRM_1)} > 1 > \frac{^{235}U/^{238}U\ (sample)}{^{235}U/^{238}U\,(IRM_2)}$$

4.2 Isotopic Composition

The isotopic composition was determined by thermal ionisation mass spectrometry.

For this purpose pellets were dissolved, the solution then adjusted to 1 molar nitric acid and a uranium concentration of about 5 mg/g. The measurements were performed on a Finnigan MAT 260 instrument. Parallel measurements on a Finnigan MAT 262 mass spectrometer equipped with a decelerating system for improved abundance sensitivity and with an ion counter were made in order to determine the minor abundant isotopes more accurately.

The measurements were calibrated against synthetic isotope mixtures : the isotope reference materials EC 183 to EC 186. The uncertainties on the reference materials were taken into account for the final accuracy statement.

As the accuracy on the $^{235}U/^{238}U$ molar isotope ratio achievable by UF_6 (gas) mass spectrometry is far better than that of thermionic mass spectrometry the isotopic composition of the sample was calculated by taking the $^{235}U/^{238}U$ ratio as determined by gas mass spectrometry and the $^{234}U/^{235}U$, $^{236}U/^{235}U$ ratios from the thermal ionisation mass spectrometry.

The $^{235}U/^{238}U$ molar isotope ratio was certified to be :

0.007 308 7 ± 0.000 002 9

The following values of the isotopic composition (in mass %) were certified :

^{234}U	0.005 51 ± 0.000 50
^{235}U	0.716 43 ± 0.000 29
^{236}U <	0.000 40 ± 0.000 00
^{238}U	99.278 06 ± 0.000 57

5. Results

Basing on the results reported by the participants, graphs were worked out which give a picture of the state of the practice. These graphs are shown on the following pages.

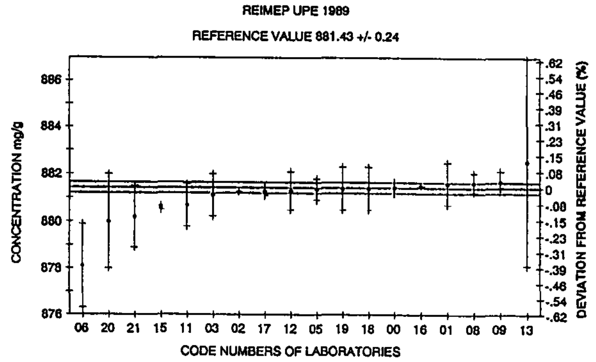
The uncertainties on the reference values were established on a 2s basis.

Uranium Element Assay

Reference Value: 881.43 ± 0.24 $mg \cdot g^{-1}$

Interlab. Mean : 881.06 ± 0.70 $mg \cdot g^{-1}$

Median Value : 881.3 $mg \cdot g^{-1}$

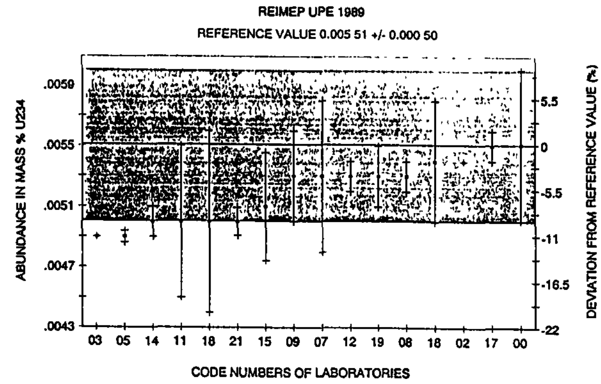


^{234}U Abundance Determination

Reference Value: 0.005 51 ± 0.000 50 mass %

Interlab. Mean : 0.005 19 ± 0.000 20 mass %

Median Value : 0.005 3 mass %

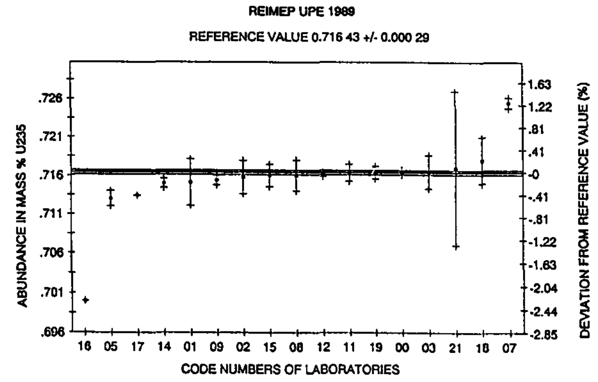


^{235}U Abundance Determination

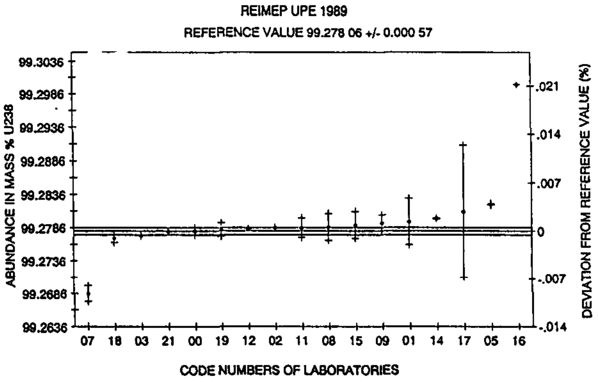
Reference Value: 0.716 43 ± 0.000 29 mass %

Interlab. Mean : 0.715 4 ± 0.001 6 mass %

Median Value : 0.716 mass %



^{238}U Abundance Determination
Reference Value: 99.278 06 ± 0.000 57 mass %
Interlab. Mean : 99.279 6 ± 0.001 7 mass %
Median Value : 99.278 6 mass %



6. Discussion

The analysis of a uranium dioxide pellet requires weighing and dissolution steps.

The determination of the element content did not pose problems to the majority of the laboratories. A tendency to lower than the certified value is observed for those laboratories with deviating values.

A similar trend is observed for the ^{235}U abundance values. Some of the laboratories reported too low ^{235}U abundances, while their ^{238}U abundance values are too high compared to the reference value.

The reference value for the ^{234}U abundance is the highest of all values displayed. However, the majority of the laboratories reported values that lie within the range of uncertainty of the reference value.

The standard deviations on the interlaboratory means were determined using the DoD (Distribution of Differences) method /2/. The DoD software was worked out by Kernforschungszentrum Karlsruhe.

The external quality control programme REIMEP is under sponsorship of the Euratom Safeguards Authorities, Commission of the European Communities, DG XVII, DCS.

References

/1/ Y. Le Duigou, V. Verdingh; "Certification of a Uranium Dioxide Reference Material for in-plant Uranium Determinations - CBNM Reference Material No. 106", Communication No. 4146

/2/ W. Beyrich et al. ; "The DoD Method of Measurement Data Evaluation", International Symposium on Nuclear Material Safeguards, IAEA, Vienna, Austria, Proc. IAEA-SM-231/8 (1978)

RESULTS OF REIMEP '89
URANYLNITRATE SOLUTION

K. Mayer, A. Alonso, P. De Bièvre,
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V. Verdingh†, Y. Le Duigou

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Joint Research Centre
Central Bureau for Nuclear Measurements, GEEL,Belgium

Abstract

The interest in the safeguards of fissile material focuses on a limited number of compounds which play key roles in the nuclear fuel cycle. Uranylnitrate solution is a material which appears in several stages of the nuclear fuel cycle. Wherever uranium is to be determined by destructive methods nitric acid is the solvent of choice. Therefore the measurement capability of nuclear analytical laboratories of this material is of interest.

The 89/90 round of REIMEP covered uranium materials with ^{235}U abundances in the range of natural or depleted material. Uranylnitrate solution was distributed to 16 laboratories for destructive analysis.

The participating laboratories were asked to determine the uranium content and the isotopic composition of the material. The results reported by the participants are presented as graphs thus giving a picture of the 'state-of-the-practice'.

1. Introduction

In the frame of the REIMEP programme in 1989 a uranylnitrate solution was distributed. Through this CBNM intends to provide nuclear analytical laboratories with a regular opportunity

- for an external check on the quality of their measurements
- to establish the state-of-the-practice of assaying common nuclear materials for isotope and element content
- to present a true picture of the real life interlaboratory spread of given measurements on given materials.

This paper reports on the results of the first uranylnitrate solution round in REIMEP. All laboratories reported values obtained by destructive analysis. The choice of the method was up to the participants.

2. Participation List

1. Savannah River Plant
   Mr. Satkowski
2. Martin Marietta Energy Systems
   Piketon,Ohio, USA, Mr. Akers
3. Martin Marietta Energy Systems
   Paducah, Kentucky, USA, Mr. Beverly
4. ECN
   Petten, Netherlands, Mr. Woittiez
5. Belgonucliaire
   Dessel, Belgium, Mr. Eeckhout
6. New Brunswick Laboratory
   Argonne, Ill., USA, Mr. Lewis
7. BNF
   Capenhurst, UK, Mr. Ogden
8. BNF
   Springfields, UK, Mr. Hayes
9. WAK
   Karlsruhe, Germany, Mr. Stojanik
10. Los Alamos National Laboratory
    Los Alamos, NM, USA, Mr. Abernathey
11. SCK/CEN
    Mol, Belgium, Mr. De Regge
12. JRC
    Ispra, Italy, Mr. Mousty
13. TUI
    Karlsruhe, Germany, Mr. Koch
14. BAM
    Berlin, Germany, Mr. Tamberg
15. IAEA
    Seibersdorf, Austria, Mr. Deron
16. CEN
    Grenoble, France, Ms. Andreani

The numbers do not correspond to the laboratory codes mentioned in the graphs.

3. Preparation of the Solution

The solution was prepared by dissolving quantitatively a known mass of a reference material. The concentration of the solution has therefore been calculated on weight basis.

The reference material used for this purpose was the EC reference material No. 110. It consists of sintered uranium dioxide pellets. Detailed information on this material is available in the certification report /1/.

A known mass of this reference material 35 g) was carefully dissolved in hot concentrated nitric acid. The solution was then evaporated and subsequently diluted with 0.1 molar nitric acid until the required concentration was obtained. The solution was then homogeneised and weighed. In place of the apparent mass used in conventional weighing procedures, the true mass of solution is calculated by appling the buoyancy correction.

Thus, the reference value was established on a gravimetrical basis. The uranium concentration is expressed in mass of uranium per g of solution :

$$7.491\,1 \pm 0.001\,5 \text{ mg}\cdot\text{g}^{-1}.$$

The uncertainty attached to the reference value is established on a 2s basis for all uncertainty for all uncertainty components.

4. Certification of Isotopic Composition

4.1 Determination of the $^{235}U/^{238}U$ ratio

In order to minimise the uncertainty on the $^{235}U/^{238}U$ ratio, this figure was determined byUF_6 gas mass spectrometry. The conversion to UF_6 was done in the following way. A pellet (EC 110)was dissolved in excess HNO_3, the solution was then evaportaed to dryness. The residue was heated to 350°C overnight. The resulting $UO_3 \times H_2O$ was treated in vacuum to 250°C for 35 minutes in order to remove the water. The waterfree UO_3 was then mixed with an excess of CoF_3 and the mixture degassed in vacuum at 200°C for 30 minutes. Subsequent heating to 425°C for 70 minutes and condensation of the reaction product UF_6 at -196°C. The UF_6 was purified at -78°C and then homogenized at 150°C during 60 minutes.

For the determination of the $^{235}U/^{238}U$ molar isotope ratio of the sample, the double standard method was applied. The two isotope reference materials (IRM) are chosen such that:

$$\frac{^{235}U/^{238}U\ (sample)}{^{235}U/^{238}U\,(IRM_1)} > 1 > \frac{^{235}U/^{238}U\ (sample)}{^{235}U/^{238}U\,(IRM_2)}$$

The atomic isotope ratio was determined to: $^{235}U/^{238}U$ = 0.003 204 5 ± 0.000 002 0

4.2 Determination of the isotopic composition

The minor abundant isotopes ^{234}U and ^{236}U as well as ^{235}U were determined by thermal ionisation mass spectrometry.

For this purpose one of the ampoules was opened, the solution evaporated to dryness and redissolved in 1 molar nitric acid. The uranium concentration was adjusted to about 5 $mg \cdot g^{-1}$. The measurements were performed on a Finnigan MAT 260 instrument. Parallel measurements on a Finnigan MAT 262 equipped with a decelerating system for enhanced abundance sensitivity and with an ion counter were made in order to determine the minor abundant isotopes more accurately.

The measurements were calibrated against synthetic isotope mixtures : the isotope reference materials EC 183 to EC 186. The uncertainties on the reference materials were taken into account for the final accuracy statement.

4.3. Calibration of the isotopic composition

Using the calibrated $^{234}U/^{235}U$ and $^{236}U/^{235}U$ ratios as determined by thermionic mass spectrometry and the results of the gas mass spectrometric determination of $^{235}U/^{238}U$ the reference values for the isotopic composition of the sample were calculated.

The accompanying uncertainties were used for the calculation of the final uncertainty statement.

The $^{235}U/^{238}U$ ratio was certified as :

0.003 204 5 ± 0.000 002 0

The following values of the isotopic composition were certified :

^{234}U	0.001 96	± 0.000 27 mass %
^{235}U	0.315 36	± 0.000 20 mass %
^{236}U	0.012 58	± 0.000 28 mass %
^{238}U	99.670 10	± 0.000 43 mass %

5. Results

Basing on the results reported by the participants, graphs were worked out which give a picture of the state of the practice. These graphs are shown on the following.

The uncertainties on the reference values were established on a 2s basis.

Discussion

The analysis of a uranylnitrate solution is the easiest case amongst the materials distributed as no dissolution of the material is required.

Nevertheless the interlaboratory spread in the element concentration determination is about 0.4 %.

The determination of ^{235}U and ^{238}U abundances showed that some of the laboratories seem to have problems with the $^{235}U/^{238}U$ ratio measurement. These laboratories reported too high ^{235}U and too low ^{238}U abundances as compared to the reference values.

It seems that not all the laboratories do estimate their accuracy realistically.

The standard deviations on the interlaboratory means were determined using the DoD (Distribution of Differences) method /2/. The DoD software was worked out by Kernforschungszentrum Karlsruhe.

The external quality control programme REIMEP is under sponsorship of the Euratom Safeguards Authorities, Commission of the European Communities, DG XVII, DCS.

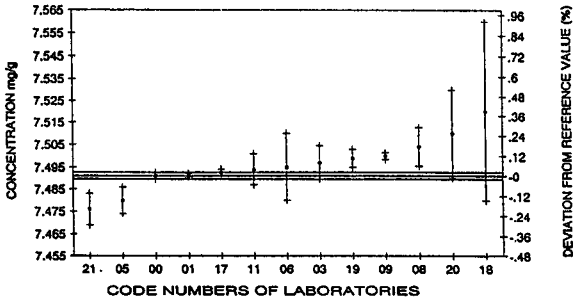
References

/1/ Y. Le Duigou (Editor); "Certification of a Uranium Dioxide Reference Material for Chemical Analyses", Report EUR 8753 EN, (1984)

/2/ W. Beyrich et al. ; "The DoD Method of Measurement Data Evaluation", International Symposium on Nuclear Material Safeguards, IAEA, Vienna, Austria, Proc. IAEA-SM-231/8 (1978)

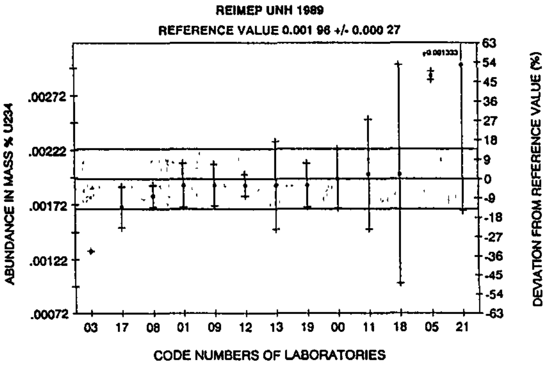
Uranium Element Assay

Reference Value:	**7.491 1 ± 0.001 6**	**$mg \cdot g^{-1}$**
Interlab. Mean :	**7.496 1 ± 0.011**	**$mg \cdot g^{-1}$**
Median Value :	**7.495 0**	**$mg \cdot g^{-1}$**



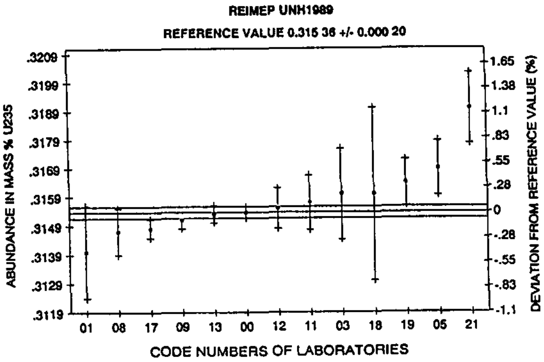
^{234}U Abundance Determination

Reference Value: 0.001 96 ± 0.000 27 mass %
Interlab. Mean : 0.002 01 ± 0.000 20 mass %
Median Value : 0.001 9 mass %



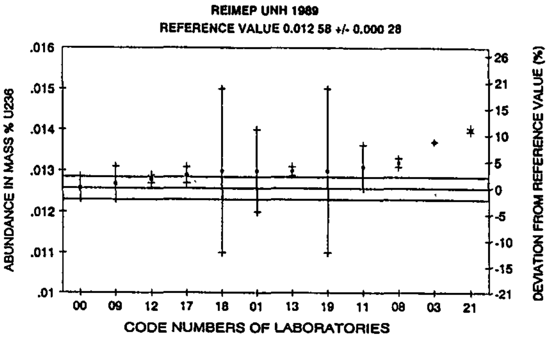
^{235}U Abundance Determination

Reference Value: 0.315 36 ± 0.000 20 mass %
Interlab. Mean : 0.315 8 ± 0.001 0 mass %
Median Value : 0.315 50 mass %



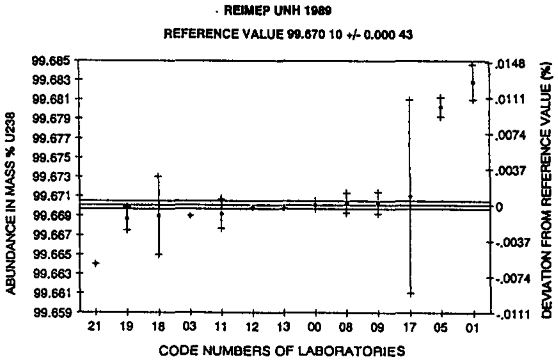
^{236}U Abundance Determination

Reference Value: 0.012 58 ± 0.000 28 mass %
Interlab. Mean : 0.013 1 ± 0.003 0 mass %
Median Value : 0.013 00 mass %



^{238}U Abundance Determination

Reference Value: 99.670 10 ± 0.000 43 mass %
Interlab. Mean : 99.671 1 ± 0.001 6 mass %
Median Value : 99.669 8 mass %



FLUORESCENT DETERMINATION OF NEPTUNIUM IN PLUTONIUM

V.M.Alexandruk, A.S.Babaev, T.A.Dem'yanova, A.V.Stepanov
Khlopin Radium Institute, Leningrad, USSR

Abstract

This paper describes a new procedure for direct determination of neptunium in plutonium using laser indused time resolved fluorescence method. The procedure based on measurement of fluorescence intensity of neptunium followed its concentration in effective layer of pellet of calcium fluoride. Detection limit of determination of neptunium is $2\cdot10^{-12}$ g. At the level of neptunium content in plutonium more than 5 ppm relative standard deviation is equal 0.08-0.12. For carrying out of single measurement it is necessary neither more nor less 5 mkg plutonium.

1. Introduction

The determination of neptunium in the solution of spent fuel particularly in plutonium is usually performed by alpha-spectrometric or spectrophotometric methods. These methods require to conduct a time-consuming process for the separation of neptunium from accompanying alpha-emitting nuclides or elements which show similar photometric reactions, especially from uranium and plutonium. Rather high detection limit of neptunium by means of these methods (0.05-0.1 mkg) necessitate to use comparatively large amounts of highly radioactive samples.

One of the most sensitive ways for neptunium determination is the fluorescent method based on CaF_2:Np crystallophosphor intensity measurement /1/. This crystallophosphor under ultraviolet irradiation exhibits fluorescence in the region of 630-720 nm and has characteristic structure of the spectrum with maxima at 637, 651 and 663 nm (Fig. 1), the two latter being more intensive. The fluorescence of CaF_2:Np in visible region is observed only on cooling crystallophosphor below 120 K. Neptunium detection limit is 0.1 ng for 0.1 g mass of cristallophosphor.

High sensitively of fluorescent method allows to determine neptunium in a wide variety of objects, including spent fuel solution with rather high content of neptunium. However further lowering of neptunium detection limit is possible. This enables to decrease considerably the amount of sample needed for analysis, what is great importance in the case of highly radioactive samples.

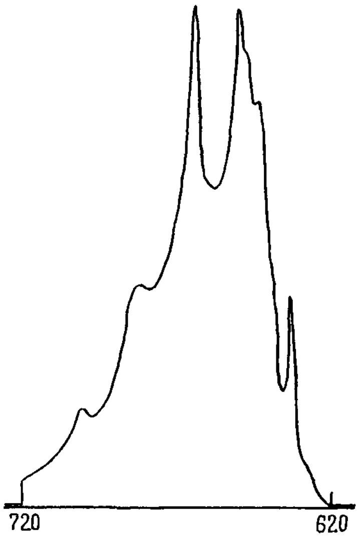


Fig. 1. Fluorescence spectrum of Np

The present work is concerned with the development of a procedure for the determination of neptunium in plutonium solutions.

2. Apparatus and Reagents

The work was conducted by using a fluorescent homemade spectrometer (Fig. 2) on the basis of MDR-23 monochromater with reciprocal linear dispersion of 1.2 nm/mm. Fluorescence was induced by means of nitrogen laser ILGI-503 having light pulse duration of 10 ns and pulse frequency of 100 Hz. Emission pulse energy was 50 mkJ. Time resolved technique was used with counting delay of analytical pulses relative to laser pulse at a certain value of time gate. Delay time and gate width could be controlled within 1-10000 mks. The spectrometer permitted to record spectrum and measure fluorescence lifetime. The measurements were performed when cooling the samples to 77 K.

All chemicals excepting plutonium were high pure grade. Solutions were prepared in double distilled water. For preparation of standard solutions 0.3 g plutonium dioxide were dessolved in nitric and hidrofluoric acids. Plutonium nitrate was cleaned from trace of neptunium with use

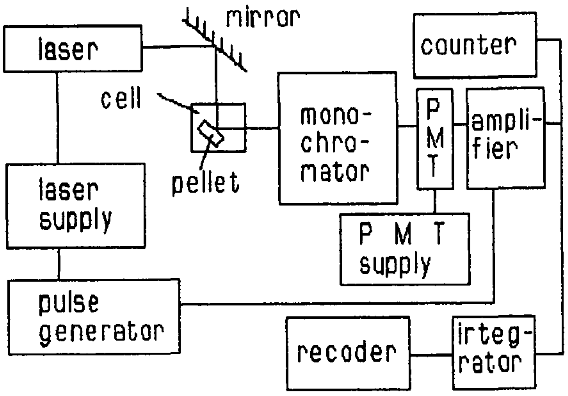


Fig. 2. Block-scheme of fluorescence spectrometer.

of chromatographic separation. Standard solutions of plutonium (IV) were prepared by suitable dilutions. Concentration of plutonium was determined by liquid-scintillation technique with $S_r=0.3$ %. Ratio Np/Pu in this solution was not higher than 0.1 ppm. Standard solutions of neptunium were prepared by suitable dilutions of reference stock solution.

3. Results and Discussion

The mentioned-above limit for neptunium by fluorescent method was obtained in homogeneous distribution of neptunium in CaF_2:Np. However in this case in processes of fluorescence exitation and emission merely a small part of cristallophosphor sample is involved while the value of this contribution depends on beam diameter of laser emission (2 mm) and depth of effective fluorescence layer which constitutes commonly for crystallophosphor powders 0.08-0.1 mm /2/.

One can show that the detection limit of neptunium contained in this volume showed be about 10^{-12} g. The achievement of such detection limit demands to change the gomogenous distribution of neptunium in the volume CaF_2 on its concentration in surface layer of crystallophosphor. The possibilities of ion-exchange concentration of neptunium in surface layer of calcium fluoride pellets were investigated. The pellets were manufactured from CaF_2 powder by using a hand press. Pellet diameter was 8 mm, thickness about 1 mm.

The sorption of Np(IV) and Np(V) from nitric and hydrochlorical acids solutions on CaF_2 powder was studied under static conditions. The investigation of neptunium sorption was conducted by means of synthetized CaF_2 specimens and commercial high-pure CaF_2 specimen.

No appriciable sorption of Np(V) from nitric or hydrochloric acid solutions of 0.01-0.5 M was observed. On the contrary Np(IV) was sorbed on CaF_2 powder from 0.05-5 M HCl solutions with distribution coefficient near to 100.

All the investigated speciments of calcium fluoride developed the similar ion-exchange properties, therefore the commercial high-pure calcium fluoride was used for further experiments.

The preparation of crystallophosphor consisted in applying the neptunium-containing solution on the surface of CaF_2 pellet with subsequent drying under infrared lamp and calcining in a muffle furnace. Then the pellet was placed in spectrometer cuvette department and cooled by liquid nitrogen. The measurement of intensity and fluorescence was carried out at 651 nm. Widths of monochromater slits were 0.5 mm.

Optimum press effort for preparation the pellets was 3 tonns. Under the stresses less than 1 tonne the pellets proved to be mechanically unstable and under the stress more than 6 tonnes they began to laminate.

The volume of solution once applied on a pallet should not exceed 0.03 ml. So, larger solution volumes were applied by 0.02 ml parts with subsequent drying.

When applying of 0.02 ml solution, the pellet is completely wetted and neptunium is uniformly distributed over the whole surface, as revealed by laser scanning of the pellets.

It was shown earlier /1/ that when preparation of crystallophosphor powder the fluorescence intensity does not depend on the oxidation state of neptunium in solution. Therefore, the difference in behaviour of Np(IV) and Np(V) may be assumed to be caused by their different ion-exchange properties in relation to CaF_2.

The fluorescence intensity of Np(IV) is independent of HCl concentration in the range of 0.05-5 M. With increasing solution volume, specific intensity expressed counts$\cdot s^{-1} \cdot ng^{-1}$ remains constant up to V=0.1 ml. Np(IV) is consentrated in surface layer of CaF_2 pellet. Its distribution in depth obeys the exponential law and some 90% of neptunium are accumulated in the layer to depth less than 0.07 mm for the solution volume from 0.02 to 0.1 ml.

The fluorescence intensity of Np(V) is constant within 0.01-0.1 M HNO_3. At the concentration of HNO_3 above 0.5 M the decrease in fluorescence intensity is observed due to increasing Np distribution depth in pellet.

The fluorescence intensity of Np(V) drops with increasing the solution volume applied to. This results from high mobility of Np(V). However, at the constant volume of applied solution the fluorescence intensity depends linearly on the amount of neptunium applied to the pellet.

Just as for Np(IV) solution, neptunium (V) is distributed in the surface layer to depth no more than 0.1 nm when applying 0.02 ml solution of Np(V) in 0.01-0.1 M HNO_3. The distribution follows the exponential law as well.

The linear dependence of fluorescence intensity is valid up to 10 ng both for Np(IV) and Np(V). Further tests were conducted with Np(V) solutions in order to eliminate the stage of Np(V)-Np(IV) reduction in the course of analysis.

The fluorescence intensity has a maximum on calcining the the pellet at 800 °C for 20 minutes (Fig. 3). When calcining at higher temperature, the fluorescence intensity decreases due to neptunium diffusion into pellet. After heating at 1200 °C the fluorescence of Np is observed on the opposite side of pellet.

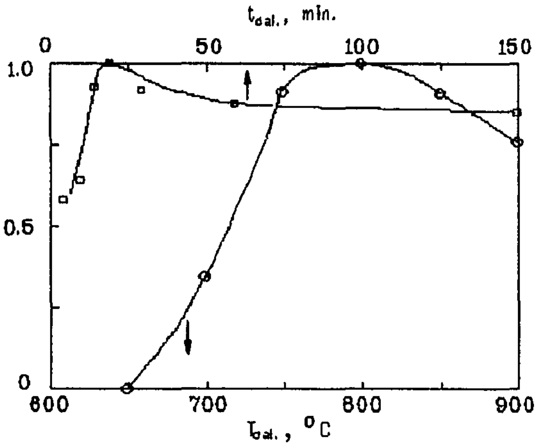


Fig. 3. Influence of temperature and duration of calcination on Np fluorescence intensity

Fluorescence spectrum of neptunium on pellets is similar to that of crystallophosphor powder (Fig. 1). Np fluorescence lifetime is 450 mks, background fluorescence lifetime - 12 mks. On the basis of these data, the delay time of 100 mks and gate-width of 1000 mks were established.

The crystallophosphor based on CaF_2 pellets retains its fluorescent properties when prolonged storage in air and also on repetitive cooling in liquid nitrogen. Stable properties enable to use the same reference specimens during a long period of time.

The intensity of neptunium determination was $1 \cdot 10^5$ count·ng^{-1} for pulse counting time of 10 s. Neptunium detection limit is equal to $2 \cdot 10^{-12}$ g (~0.05 mBk). This value is somewhat higher than one could be expected, what is caused by a greater value of pellet diameter as compared to the diameter of laser beam. The attained limit for neptunium detection by fluorescent method is rather low and compares favourably with the detectability by neutron activation ($5 \cdot 10^{-13}$ g Np) [3]. However for performing the latter a high-flux reactor is necessary.

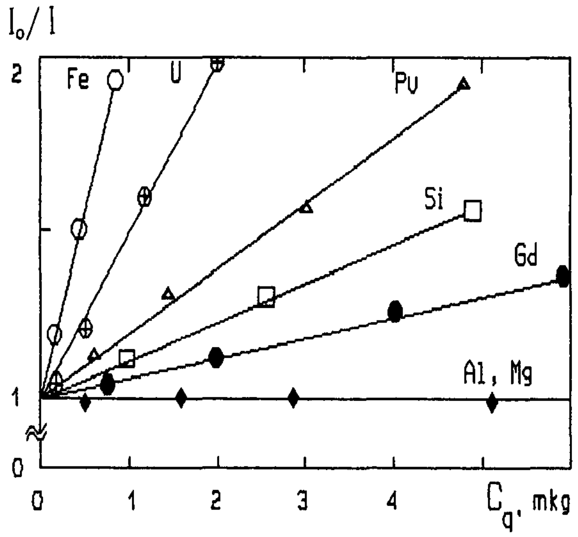


Fig. 4. Influence of some quenchers on Np fluorescence intensity

The dependence of fluorescence intensity on concentration of several elements, including plutonium, is presented on Fig. 4. For the most cations under investigation, the linear dependence is observed between the ratio of I_0/I and the concentration of quencher.

The mass of plutonium at which the fluorescence intensity halved comes to 5 mkg. For Fe(III) this mass is 1 mkg. Alkaline and earth-alkaline elements do not affect the fluorescence intensity of neptunium up to 5 mkg per a pellet.

The most cations produce no spectral disturbances, expect for uranium and aluminium. Uranium generates intensive emission in calcium fluoride with maximum at 530 nm, which rise to a significant increase of background in 651 region. The permissible content of uranium, without any increase of background in 651 nm region, constitutes 0.2 mkg per pellet.

In the presence of aluminium calcium fluoride exhibits fluorescence peak with a maximum at 694 nm. The maximum permissible content of aluminium which does not require to take into account the background changes is 0.1 mkg.

The determination of neptunium in plutonium

Table 1. Comparison of the data on determination of neptunium in plutonium

N	Np/Pu, %		
	fluoresc.	X-ray fluor.	alpha-spectr.
1	10.8 ± 1.2	-	9.7 ± 0.5
2	6.9 ± 0.5	-	7.2 ± 0.4
3	0.81 ± 0.04	-	0.94 ± 0.04
4	0.69 ± 0.07	-	0.70 ± 0.08
5	$(2.9\pm0.2)\cdot10^{-2}$	$(2.9\pm0.2)\cdot10-^{2}$	-
6	$(5.2\pm0.5)\cdot10^{-3}$	$(4.6\pm0.6)\cdot10^{-3}$	-
7	$(2.9\pm0.3)\cdot10^{-2}$	$(2.8\pm0.2)\cdot10^{-2}$	-
8	$(1.6\pm0.2)\cdot10^{-3}$	$(1.4\pm0.6)\cdot10^{-3}$	$(1.4\pm0.1)\cdot10^{-3}$
9	$(0.70\pm0.05)\cdot10^{-2}$	-	-

solutions could be carried out according to method of standard additions. In this case the effect of all quenchers is taken into consideration. However if the content of other quenchers does not exceed some per cents with respect to plutonium, the determination could be performed according to graduated plot constructed for pure neptunium solutions. Correction for fluorescence quenching by plutonium itself might be introdused in accordance with Pu dependence shown on Fig. 4.

On the base of both the detection limit of neptunium in pure solution and the value of fluorescence quenching by plutonium, it is possible to estimate the detection limit of neptunium in plutonium. This value comes to 1-5 ppm depending on mass of the latter on pellet. For 5 mkg Pu mass and content of $5\cdot10^{-4}$ % Np, the value S_r was 0.12, for $5\cdot10^{-3}$ % and more - 0.08. In any case, plutonium mass on pellet should not exceed 5 mkg. On neptunium content above $5\cdot10^{-2}$ %, the amount of plutonium applied to pellet should be respectively decreased because of high pulse-counting rate. Experimental value of detection limit determined as doubled value of background intensity was close to the data mentioned-above.

It was shown that F^-, Cl^-, and $C_2O_4^{2-}$ at concentration up to 0.1 M have no influence on Np fluorecsence intensity. In the presence of I^--ion, fluorescence is observed in 694 nm region similarly to that in the presence of Al.

The Table 1 presence the comparison of the data on the determination of neptunium in plutonium by fluorescent method and the results of alpha-spectrometry and x-ray fluorescence analysis. When determining neptunium by two latter methods, its seperation from plutonium was conducted by means of partition-chromatography. The yield of neptunium was determined from ^{239}Np.

Good agreement is observed within the error limits for the compared methods.

When determinating neptunium in plutonium below $5\cdot10^{-4}$ %, the separation of these elements is requied. We used prosedure of exstraction-chromatographic separarion /4/ with trimethyloctyl ammonium (TMOA) applied to porous teflon. Neptunium was eluated by 0.03 % oxalic acid. Neptunium yield was determined by ^{239}Np.

Using 1-2 mg Pu, neptunium up to 10^{-6} % could be determined in such a manner.

4. Conclusion

This work has proved that consentration of neptunium in effective layer of pellet of calcium fluoride allowed to decrease the significantly detection limit of fluorescent determination of neptunium in plutonium in compare with gomogenous distribution of Np in volume of cristallophosphor. It is possible to carry out direct determination of neptunium of in plutonium in spite of high quenching action of latter using simple dilution of initial solution. High sensitivity of the method allowed to use for measurements two order less quantity of plutonium in compeare with other methods. For determination of 5 ppm or less of neptunium in plutonium 5 mkg it is necessary to carry out chemical separation of these elements. Nevetheless in this case the quantity of plutonium used for analysis will be smaller as compared with other methods.

Technique of concentration in surface layer of crystallophosphor may be also applay for decrease of detection limit of other elements. For example detection limit of uranium in this case can be reduce down to 10^{-14} g.

5. References:

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/2/ N.P.Efryushina, G.K.Novikova, E.A.Zgikchareva, N.S.Poluektov. Effective Lengths of Active Layer of Crystallophosphors. Journal of Applied Spectroscopy (Sov. Un.), vol. 29 (1978) p. 744.

/3/ S.May, Ch.Engelmann, G.Pinte. Neutron Activation Determination of ^{237}Np in Irradiated Experimental Fuels and in Waste Solutions and Distribution Studies in Sea-waters and Submarine and Flora of Disposal Areas. Journal of Radioanalytical and Nuclear Chemistry, Vol. 113 (1987) p.343.

/4/ V.A.Mikhailov. Analytical Chemistry of Neptunium. Moscow: "Nauka", 1971, p. 131.

OPTIMAL SAMPLE TO TRACER RATIO FOR ISOTOPE DILUTION MASS SPECTROMETRY: THE POLYISOTOPIC CASE

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Abstract

The Isotope Dilution Mass Spectrometry (IDMS) measurement technique provides a means for determining the unknown amount of various isotopes of an element in a sample solution of known mass. The sample solution is mixed with an auxiliary solution, or *"tracer"* containing a known amount of the same element having the same isotopes but of different relative abundances or isotopic composition and the induced change in the isotopic composition measured by isotope mass spectrometry. The technique involves the measurement of the abundance ratio of each isotope to a (same) *"reference"* isotope in the sample solution, in the tracer solution, and in the blend of the sample and tracer solution. These isotope ratio measurements, the known element amount in the tracer, and the known mass of sample solution are used to calculate the unknown amount of one isotope in the sample solution. Subsequently the unknown amount of element is determined.

The purpose of this paper is to examine the optimization of the ratio of the estimated unknown amount of element in the sample solution to the known amount of element in the tracer solution in order to minimize the relative uncertainty in the determination of the unknown amount of element.

1. Introduction

The Isotope Dilution Mass Spectrometry (IDMS) measurement technique provides a means for determining unknown amounts or concentrations of elements in solutions by finding the unknown amount of one or more of the various isotopes in a sample. To accomplish this, one uses an auxiliary solution, called *"tracer"* containing a known amount of the same element with some or all the same isotopes as the sample but of a different isotopic composition.

Three sets of measurements obtained using isotope mass spectrometry are needed. These are the abundance ratios of the various isotopes relative to one reference isotope in (i) the original sample; (ii) the tracer and (iii) the blend of the sample and the tracer solutions. From the three sets of ratio measurements, the known masses of the tracer and sample solutions, and the known amount of element in the tracer, one calculates the unknown amount of element in the solution. All measurements in this procedure are performed with known uncertainties. Note that any amount of tracer in the IDMS scheme provides a value for the amount of sample; the answer, however, might not be optimal as far as the associated uncertainty is concerned. provides a value for the unknown amount of sample; the answer, however, might not be optimal as far as the associated uncertainty is concerned. Consequently, one is interested in deriving the optimal blend ratio, i.e., the one with the smallest relative uncertainty. The main steps of this optimization process are as follows.

Following De Bièvre/1/ we use $R_i(x)$, $R_i(y)$, (where $i = 1, 2, \ldots, n$ and n is the number of isotopes), to denote the number of isotope ratios in the sample and in the tracer. Let N_i (x) denote the number of atoms of the ith sample isotope. Then

$$R_i(x) = N_i(x)/N_1(x),$$

where isotope 1 is the arbitrarily chosen reference isotope. It is best to choose a major abundance isotope in the sample as the reference. Likewise for the tracer

$$R_i(y) = N_i(y)/N_1(y).$$

The blend of the sample and the tracer is uniquely described by the ratio

$$R_b = (N_2(x) + N_2(y)) / (N_1(x) + N_1(y)),$$

where isotope *2* is any isotope other than the one used for the reference. It is better, however to use the most abundant isotope in the tracer. These two isotopes then become the only ones that must be measured in the blend.

The problem as posed in /1/ is to minimize the relative uncertainty of the quantity

$$q = \Sigma N_i(x)/\Sigma N_i(y) \ ,$$

where *q* is the ratio of the amount of element in the sample relative to the amount of element in the tracer.

2. Optimization Model

In isotope ratios *q* can be expressed as follows /1/

$$q = \Sigma R_i(x)/\Sigma R_i(y) \ \frac{R_2(y) - R_b}{R_b - R_2(x)} \ .$$

The novel features of this paper are that *q* is expressed as a function of R_b, the optimum is determined in terms of R_b, and the contribution to the uncertainty of all the isotopes in a poly-isotopic mixture is considered. Computationally, the problem treated in this fashion is advantageous for

$$\min\big(R_2(x),\ R_2(y) < R_b < \max(R_2(x),\ R_2(y)\big).$$

Next, the $Var(q)/q^2$ must be determined. Here, we apply the propagation of error technique based on the first order *Taylor* approximation of the multivariate function

$$F\big(R_1(x),\ \ldots.\ R_n(x),\ R_1(y),\ \ldots\ R_n(y);\ R_b\big) \ .$$

The function to be minimized in terms of R_b is $Var(q)/q^2$:

$$\frac{Var\ q}{q^2} = \left[\frac{R_2\ (x)}{R_b - R_2\ (x)} + \frac{R_2\ (x)}{\Sigma\ R_i\ (x)}\right]^2 \frac{Var\ R_2\ (x)}{R_2^2(x)}$$

$$+ \frac{\sum\limits_{i=3}^{n} R_i^2\ (x)\ Var\ R(i)\ (x)/R_i^2(x)}{(\Sigma\ R_i\ (x))^2}$$

$$+ \left[\frac{R_2\ (y)}{R_2\ (y) - R_b} - \frac{R_2\ (y)}{\Sigma\ R_i\ (y)}\right]^2 \frac{Var\ R_2\ (y)}{R_2^2\ (y)}$$

$$+ \frac{\sum\limits_{i=3}^{n} R_i^2\ (y)\ Var\ R_i\ (y)/R_i^2\ (y)}{(\Sigma\ R_i\ (y))^2}$$

$$+ R_b^2\left[\frac{R_2\ (x) - R_2\ (y)}{(R_b - R_2\ (x))\ (R_2(y) - R_b)}\right]^2 \frac{Var\ R_b}{R_b^2} \ .$$

Note that the second and the fourth terms do not contain R_b. Using simplified notations the problem consists of minimizing the positive function *f(t)* defined as follows:

$$f(t) = x_2^2\left(\frac{1}{t - x_2} + \frac{1}{\Sigma\ x_i}\right)^2 C_x + y_2^2\left(\frac{1}{y_2 - t} - \frac{1}{\Sigma\ y_i}\right)^2 C_y$$

$$+ t^2\ \frac{(x_2 - y_2)^2}{(t - x_2)^2\ (y_2 - t)^2}\ C_b$$

with:

$$\begin{aligned} &(i)\ \ t = R_b \\ &\quad x_i = R_i(x);\ y_i = R_i(y) \quad (i=1,\ \ldots,\ n) \\ &\quad C_x = Var\ R_2(x)/R_2^2(x); \\ &\quad C_y = Var\ R_2(y)/R_2^2(y); \\ &\quad C_b = Var\ R_b/R_b^2 \\ &(ii)\ \ \min(x_2,\ y_2) < t < \max(x_2,\ y_2) \ . \end{aligned}$$

The first derivative of *f(t)* can be written as the ratio of a fourth order to a sixth order polynomial

$$f'(t) = \frac{p^{(4)}(t)}{p^{(6)}(t)}, \quad where$$

$$p^{(4)}(t) = -\ C_x x_2^2\ (y_2 - t)^3 \left(1 + \frac{(t - x_2)}{\Sigma\ x_i}\right)$$

$$+ C_y\ y_2^2\ (t - x_2)^3 \left(1 - \frac{(y_2 - t)}{\Sigma\ y_i}\right)$$

$$+ C_b\ t\ (x_2 - y_2)^2\ (t^2 - x_2 y_2)$$

The function to be minimized in terms of R_b is $Var(q)/q^2$:

$$\frac{Var\ q}{q^2} = \left[\frac{R_2\ (x)}{R_b - R_2\ (x)} + \frac{R_2\ (x)}{\Sigma\ R_i\ (x)}\right]^2 \frac{Var\ R_2\ (x)}{R_2^2(x)}$$

$$+ \frac{\sum\limits_{i=3}^{n} R_i^2\ (x)\ Var\ R(i)\ (x)/R_i^2(x)}{(\Sigma\ R_i\ (x))^2}$$

$$+ \left[\frac{R_2\,(y)}{R_2\,(y) - R_b} - \frac{R_2\,(y)}{\Sigma\, R_i\,(y)}\right]^2 \frac{Var\ R_2\,(y)}{R_2^2\,(y)}$$

$$+ \frac{\sum_{i=3}^{n} R_i^2\,(y)\ Var\ R_i\,(y)/R_i^2\,(y)}{\left(\Sigma\, R_i\,(y)\right)^2}$$

$$+ R_b^2\left[\frac{R_2\,(x) - R_2\,(y)}{\left(R_b - R_2\,(x)\right)\left(R_2(y) - R_b\right)}\right]^2 \frac{Var\ R_b}{R_b^2}$$

Note that the second and the fourth terms do not contain R_b. Using simplified notations the problem consists of minimizing the positive function $f(t)$ defined as follows:

$$f(t) = x_2^2\left(\frac{1}{t - x_2} + \frac{1}{\Sigma\, x_i}\right)^2 C_x + y_2^2\left(\frac{1}{y_2 - t} - \frac{1}{\Sigma\, y_i}\right)^2 C_y$$
$$+ t^2\,\frac{\left(x_2 - y_2\right)^2}{\left(t - x_2\right)^2\left(y_2 - t\right)^2}\, C_b$$

with:

$$\begin{aligned} (i)\quad t &= R_b \\ x_i &= R_i(x);\ y_i = R_i(y)\quad (i=1, \ldots, n) \\ C_x &= Var\ R_2(x)/R_2^2(x); \\ C_y &= Var\ R_2(y)/R_2^2(y); \\ C_b &= Var\ R_b/R_b^2 \\ (ii)\quad \min(x_2,\ y_2) &< t < \max(x_2,\ y_2)\ . \end{aligned}$$

The first derivative of $f(t)$ can be written as the ratio of a fourth order to a sixth order polynomial

$$f'(t) = \frac{p^{(4)}(t)}{p^{(6)}(t)}, \quad where$$

$$p^{(4)}(t) = -\,C_x x_2^2\left(y_2 - t\right)^3\left(1 + \frac{\left(t - x_2\right)}{\Sigma\, x_i}\right)$$
$$+ C_y\, y_2^2\left(t - x_2\right)^3\left(1 - \frac{\left(y_2 - t\right)}{\Sigma\, y_i}\right)$$
$$+ C_b\, t\left(x_2 - y_2\right)^2\left(t^2 - x_2 y_2\right)$$

```
Data given :     ==> R.() vector components: (U238/U238, U235/U238, U234/U238, U236/U238)
____________

      Rx()             Rel. error (%)
      ------------------------------

      1.000000          0.000000
      0.135800          0.220000
      0.001100          1.000000
      0.000175          1.000000

      Ry()             Rel. error (%)
      ------------------------------

      1.000000          0.000000
     17.269300          0.100000
      0.184400          0.500000
      0.077408          1.000000

      Rb()             Rel. error (%)
      ------------------------------

                        0.150000


Numerical results :
___________________

RbOpt     : calculated optimal isotopic ratio of the blend of sample & spike;      ===> 1.700215
OptQ      : optimal mass ratio of sample to spike;                                 ===> .6106591
Rel. err. : relative error of mass ratio of sample to spike (for short);           ===> .1860134
Dsl       : sample quantity (mg);                                                  ===> 5
Opt. Dsp  : optimal spike quantity (mg);                                           ===> 8.187875

____________________________________________

            Spike             Rel. error
            (mg)
____________________________________________

           1.00000            0.30350
           2.00000            0.22757
           3.00000            0.20505
           4.00000            0.19519
           5.00000            0.19024
           6.00000            0.18767
           7.00000            0.18643
           8.00000            0.18602
           9.00000            0.18616
          10.00000            0.18668
```

Figure 1

$$p^{(6)}(t) = \frac{1}{\left[(t - x_2)\,(y_2 - t)\right]^3} \; .$$

From *(ii)* it follows that the denominator of $f'(t)$ is not zero on the open interval (x_2, y_2), and is > 0. Thus the problem consists of determining the zeros of the numerator of $f'(t)$. The analysis of the function shows that $f'(t)$ could have only one zero of multiplicity of order 2 or 4 which indicates that the function $f(t)$ has only one minimum in the interval (x_2, y_2).

3. Computer Code and Example

A computer code in Microsoft Basic has been developed to calculate the optimum blend ratio R_b for given sample and tracer isotope compositions and relative uncertainties. Figures 1 and 2 present sample outputs of two examples. To facilitate the proper selection of tracer amount a table of tracer amounts together with the corresponding relative error of sample to tracer ratio is generated. Typically the curve of relative error of sample to tracer ratio vs. spike amount is rather flat close to its minimum value, therefore a near optimal blend ratio can be achieved with less amount of tracer. The curves of Figure 3 show this phenomenon.

4. Conclusion/Comments

A simple algorithm has been designed to calculate the minimum value of $Var(q)/q^2$ in terms of R_b for the polyisotopic case.

The form of $Var(q)/q^2$ makes the optimum value calculation easy, since

$$\min\left(R_2(x),\, R_2(y)\right) < R_b < \max\left(R_2(x),\, R_2(y)\right) .$$

```
Data given :     ==> R.() vector components: (Pu240/Pu240, Pu239/Pu240, Pu241/Pu240, Pu238/Pu240, Pu242/Pu240)
__________

    Rx()              Rel. error (%)
    ------------------------------

    1.000000            0.000000
    3.608000            0.100000
    0.105030            0.200000
    0.006166            1.500000
    0.033270            0.500000

    Ry()              Rel. error (%)
    ------------------------------

    1.000000            0.000000
   21.194700            0.200000
    0.226210            0.200000
    0.001435            2.000000
    0.001844            2.000000

    Rb()              Rel. error (%)
    ------------------------------

                        0.150000

Numerical results :
_________________

RbOpt      : calculated optimal isotopic ratio of the blend of sample & spike;      ===> 8.425359
OptQ       : optimal mass ratio of sample to spike;                                 ===> .5617742
Rel. err.  : relative error of mass ratio of sample to spike (for short);           ===> .4168476
Dsl        : sample quantity (mg);                                                  ===> 4
Opt. Dsp   : optimal spike quantity (mg);                                           ===> 7.120298

_____________________________________________________

          Spike                  Rel. error
          (mg)
_____________________________________________________

         1.00000                 0.93874
         2.00000                 0.60143
         3.00000                 0.49729
         4.00000                 0.45175
         5.00000                 0.42985
         6.00000                 0.41990
         7.00000                 0.41688
         8.00000                 0.41826
         9.00000                 0.42261
        10.00000                 0.42910
```

Figure 2

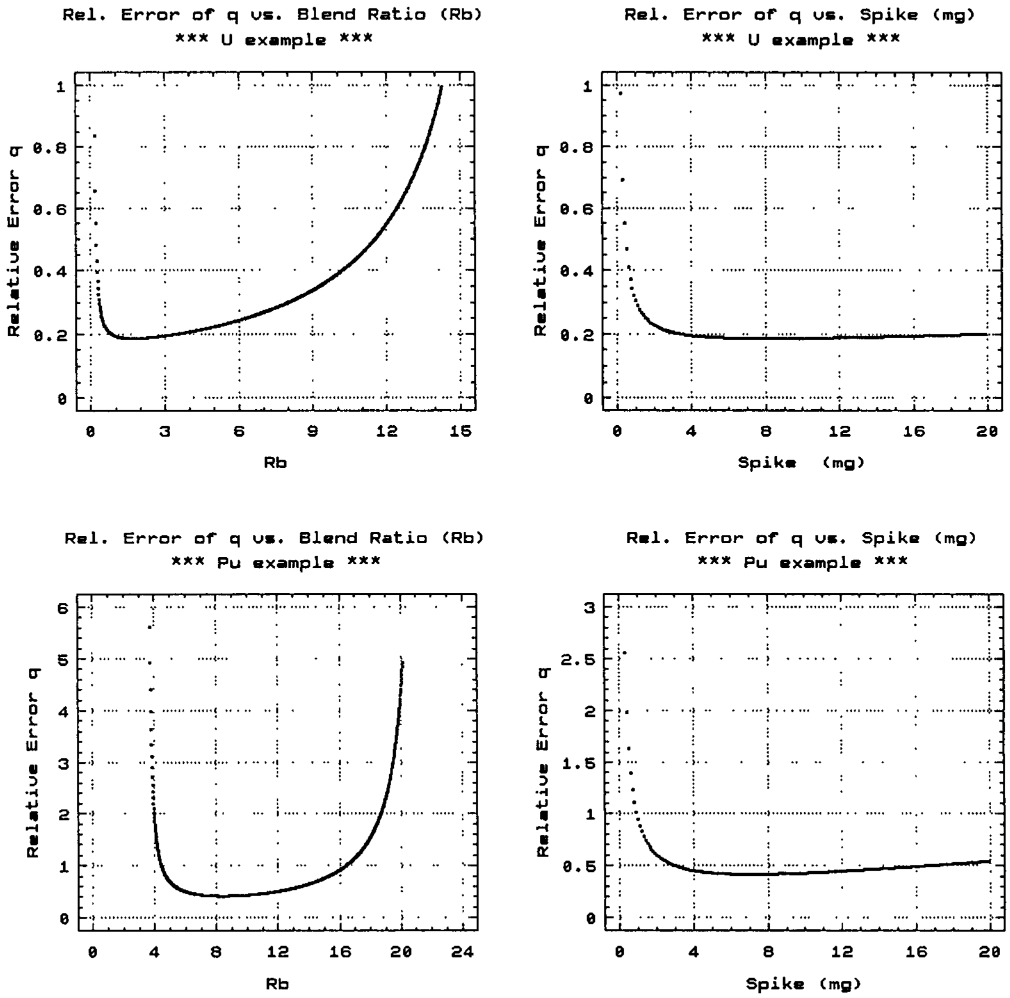

Figure 3

It has been demonstrated that there is a unique minimum value for $Var(q)/q^2$ attained.

The calculation of the optimal mix of sample and spike which achieves $R_{b,opt}$ must be achieved through an iterative procedure.

To prepare the optimal mix one has to know the amounts to be mixed, but one of the components of the blend is what is to be determined either in terms of atoms or mass or solution concentration. A good approximate knowledge of the sample amount guarantees a good approximation of the optimal blend ratio$R_{b,opt}$. (Note that lack of knowledge of the sample amount does not preclude its determination from $R(x)$, $R(y)$ and R_b and the spike amount. In such a case IDMS simply does not produce the smallest possible relative uncertainty).

5. Reference

/1/ P. J. De Bièvre, G. H. Debus, "*Precision Mass Spectrometric Isotope Dilution Analysis*", Nuclear Instruments and Methods, 32 (1965), 224-228

SYNTHETIC ^{239}PU/^{242}Pu ISOTOPE MIXTURES AS BASIS FOR ABSOLUTE Pu ISOTOPIC MEASUREMENTS AT CBNM

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Abstract

In view of the characterisation of reference materials for isotopic measurements of Plutonium, a series of 20 synthetic isotope mixtures have been made. Highly enriched ^{239}Pu and ^{242}Pu materials were mixed gravimetrically as Pu oxides in ratios ranging from 10^{-1} to 10^{+1}.

Taking into account the different uncertainty sources : chemical impurities in the starting materials, isotopic composition of the starting materials, hygroscopicity, stoichiometry and weighing, the total uncertainty for any of the mixtures is ≤ 0.02 %. This means that the correction factor for isotope fractionation of two neighbouring masses (e.g. ^{239}Pu/^{240}Pu) can be determined to ± 0.01 % for all practical purposes.
These mixtures are stored at CBNM in units of - 1 mg and will form the basis of "absolute" Pu isotopic measurements at CBNM for the next 20 years.

1. Introduction

Synthetic mixtures of Pu isotopes serve for the calibration of mass spectrometers, used in the characterisation of isotopic reference materials.

2. Preparation of the mixtures

Principle of making the mixtures (Fig. 1)

The mixtures were made gravimetrically (PuO_2 form).

Laboratory Layout

A complete new facility was installed. In order to avoid cross contamination between the enriched isotopes, the chemical operations were kept strictly separated. Figure 2 shows schematically the laboratory layout.

A microbalance with a total weighing range of 4.1 g and a standard deviation < 1 µg was selected and installed in a very stable metal glovebox.

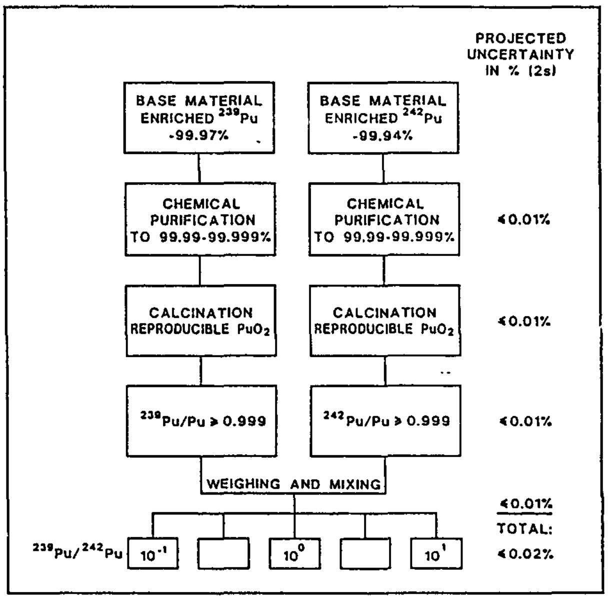

Fig. 1

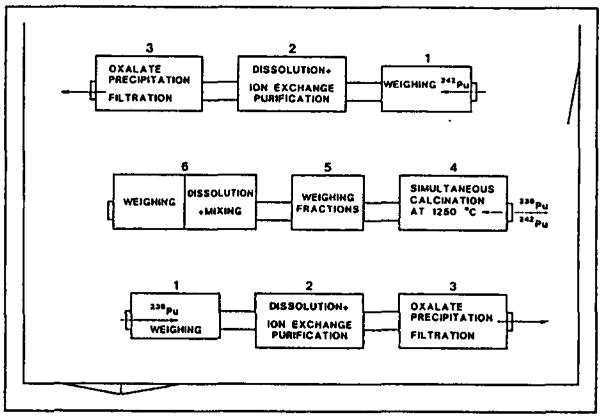

Fig. 2

The successive steps in the procedure

The base materials for the mixtures were highly enriched ^{239}Pu and ^{242}Pu (Table 1). in oxide form. These materials were taken in solution, chemically purified by means of ion exchange on anion resin, precipitated in oxalate form and calcinated to PuO_2. The calcination was done heating Pu oxalate in steps up to 1250°C and calcining for 2 hours. Under these conditions, the stoichiometry of the PuO_2 is known to be 0.01 %. On this figure we put an uncertainty of ± 0.01 %.

Experiments on the water uptake of several PuO_2 samples, calcinated at 1250°C, showed a reproducibility such that we decided only to keep the humidity content of the air stable during the weighing procedure.

	ENRICHED ^{239}Pu		ENRICHED ^{242}Pu	
	ATOM %	UNCERTAINTY (on 2s basis)	ATOM %	UNCERTAINTY (on 2s basis)
^{238}Pu	0.000 30	± 0.000 20	0.003 50	± 0.001 00
^{239}Pu	99.966 56	± 0.000 77	0.004 79	± 0.000 10
^{240}Pu	0.026 06	± 0.000 64	0.019 39	± 0.000 40
^{241}Pu	0.005 87	± 0.000 37	0.025 94	± 0.000 54
^{242}Pu	0.001 11	± 0.000 03	99.946 03	± 0.001 20
^{244}Pu	0.000 10	± 0.000 01	0.000 35	± 0.000 01

Table 1

Determination of the chemical impurities

The purity of the Pu oxides was measured by means of Spark Source analysis. Table 2 gives the impurities in ppm.

CALIBRATED*	^{239}Pu	^{242}Pu
Li	0.062	0.023
B	5.5	2.2
Na	0.17	0.30
Al	3.3	8.0
Si	15.0	3.8
P	0.35	0.30
Ca	1.2	1.1
Ti	0.26	0.018
V	0.020	<0.010
Cr	1.1	0.32
Mn	0.21	0.021
Fe	2.5	1.9
Co	0.036	<0.010
Ni	1.0	<0.070
Cu	0.33	0.55
Zn	1.5	0 51
In	0.095	0.090
Sn	1.8	0.77
Ba	<0.015	..0.40
W	0.86	<0.030
Pb	0.23	0.29
Th	11.0	5.5
	≰ 47	≰ 26
UNCALIBRATED	^{239}Pu	^{242}Pu
S	14.0	9.7
Cl	9.2	7.9
K	12.0	8.0
Ce	< 0.10	0.40
Np	14.0	0.53
	≰ 49	≰ 26
	≰ TOTAL 96	≰ TOTAL 52

*CALIBRATED: THE MEASUREMENTS HAVE BEEN CALIBRATED WITH IMPURITY STANDARDS FOR THE ELEMENTS CONCERNED. ALL IMPURITIES ARE MEASURED RELATIVE TO THE ^{239}Pu ISOTOPE, USED AS " INTERNAL STANDARD ".

- IN THE CALCULATION OF THE RATIO VALUES FOR THE SYNTHETIC MIXTURES, TOTAL IMPURITY LEVELS OF 75 ± 50 $\mu g \cdot g^{-1}$ HAVE BEEN USED (THESE ARE HIGHLY CORRELATED).

Table 2

3. Total uncertainty on the $^{239}Pu/^{242}Pu$ ratio

The total uncertainty on the ratio is the sum of different contributions :

- the uncertainty on the isotopic composition of the starting materials
- the uncertainty on the purity grade of the starting materials. In consideration are to be taken chemical impurities as well as the water uptake on the materials
- the uncertainty on the stoichiometry
- the uncertainty on the weighings

Table 3 is an example of the mixture with $^{239}Pu/^{242}Pu$ ratio = 0.235 75.

USING THE MIXTURE WITH $^{239}Pu/^{242}Pu$ = 0.23575 AS AN EXAMPLE

CHARACTERISTICS OF ENRICHED ISOTOPE MATERIAL	p p m. (RELATIVE TO VALUE)
ISOTOPIC COMPOSITION	27.6
ELEMENTAL PURITY	31.6*
IDENTITY OF STOICHIOMETRY	14.8*
MASSES (WEIGHING)	50 8
TOTAL	67.5

*TAKING INTO ACCOUNT THE HIGH CORRELATION.

Table 3

4. Verification Measurements

Each of the mixtures has been measured several times on a thermal ionisation mass spectrometer. The average value of the K factors

$$K = \frac{calculated\ value}{measured\ value}$$

are represented in fig. 3.

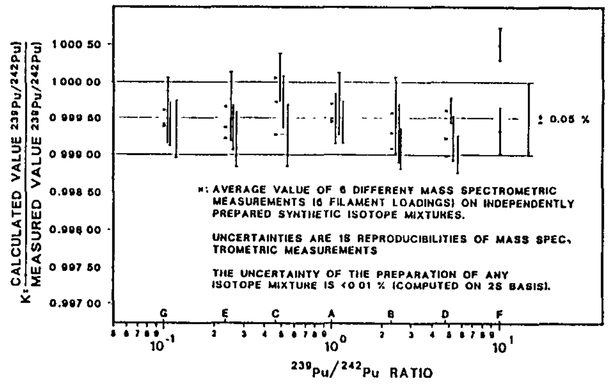

Fig. 3

In consequence of these measurements one of the mixtures (F) has been rejected.

5. Conclusions

Synthetic ^{239}Pu/^{242}Pu mixtures have been prepared with an uncertainty of ≤ 0.02 % on the ratio ^{239}Pu/^{242}Pu. From 19 different mixtures with ^{239}Pu/^{242}Pu in the range 0.1 to 10, units of 1 mg Pu as dried Pu nitrate were prepared (see Table 4).

Table 4

Pu SYNTHETIC ISOTOPE MIXTURES AS Pu NITRATE (DRIED FORM)

^{239}Pu /^{242}Pu	UNITS 1 mg	^{239}Pu /^{242}Pu	UNITS 1 mg
0.101358 11	490	1.025140 83	86
0.101884 11	482	1.013450 82	96
0.099926 11	488	1.000730 82	95
0.232119 16	472	2.31008 16	291
0.235754 16	450	2.28691 16	290
0.229387 16	479	2.25369 16	286
0.466521 33	279	4.65674 31	486
0.476282 33	271	4.54568 30	498
0.477061 33	275	4.78777 32	511
		10.0574 11	481

SOLID SPIKES FROM CBNM FOR INPUT ANALYSIS

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Abstract

Two types of ^{235}U/^{239}Pu solid spikes for the assay of reprocessing plant input solutions are under study at CBNM. Dried U/Pu nitrate spikes have been prepared from metallic reference materials and uranium and plutonium concentrations and isotopic compositions have been certified (CBNM IRM-1027 a and b). Metallic U/Pu spikes are prepared by quantitative crucible-less alloying with quality control carried out by IDMS and by gamma counting on solid and dissolved samples. Consideration of IDMS error propagation shows that, for typical LWR input solutions, the optimum Pu/U+Pu ratio for CBNM solid spikes is 0.025 to 0.1.

1. Introduction

Solid spikes for isotope dilution mass spectrometry (IDMS) analyses offer a simple and reliable procedure for the uranium and plutonium assay of reprocessing plant input solutions /1-3/. The solid spikes used are of ^{235}U/^{239}Pu composition, avoiding the use of the more exotic ^{233}U and ^{242}Pu isotopes. They are relatively large in size (~ 100 mg) and spiking can be carried out on undiluted input samples eliminating the need for quantitative dilution and simplifying the spiking process in the hot cell. The large size of the samples and spikes also means that they are relatively insensitive to chemical contamination. The dried nitrate version of the solid spike, the Large Size Dried Spike (LSD), has been successfully used by the IAEA at inspections in Japan /4-5/. This paper reports on the preparation and certification of LSD U/Pu nitrate spikes and describes the status of the preparation and characterization of U/Pu metallic spikes. The procedure used to optimise spike composition and size is also discussed.

2. Large Size Dried Spikes

Two batches of LSD spike, CBNM IRM-1027a and CBNM IRM-1027b have been prepared on a semi-commercial scale. The preparation procedure is described in Fig. 1.

Base material

For the preparation of both batches the base materials given in Table 1 have been used.

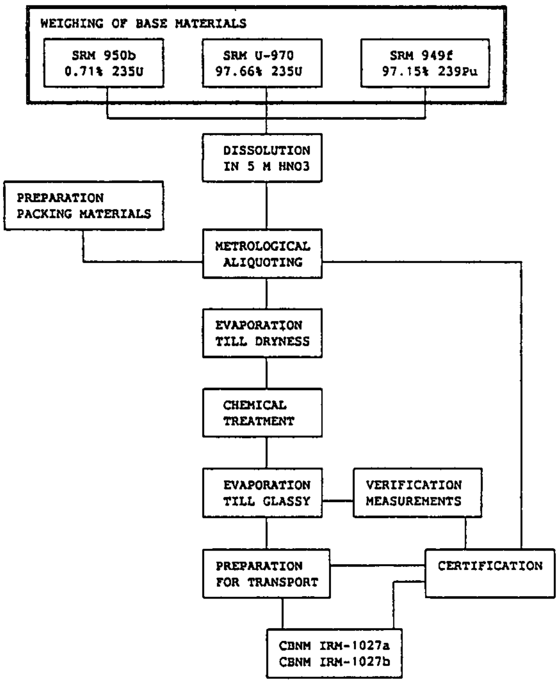

Fig. 1 - Preparation route of LSD spikes

		mol %	mass %	uncertainty
NBS 950b				
	234U :	0.005 71	0.005 61	0.000 07
	235U :	0.719 80	0.710 77	0.000 29
	236U :	0.001 20	0.001 19	0.000 01
	238U :	99.273 29	99.282 42	0.000 30
NBS SRM U-970				
	234U :	1.665 3	1.658 2	0.001 5
	235U :	97.662 7	97.662 5	0.002 5
	236U :	0.149 1	0.149 7	0.000 5
	238U :	0.522 9	0.529 6	0.000 5
NBS SRM 949f				
	238Pu :	0.003 7	0.003 7	
	239Pu :	97.148 7	97.136 8	
	240Pu :	2.802 5	2.813 9	
	241Pu :	0.039 1	0.039 5	
	241Pu :	0.006 0	0.006 1	

Table 1 - Base materials used to prepare LSD spikes. (Uncertainties are 2s)

Chemical preparation

Before weighing any of the uranium batches, both materials were ignited in an open furnace at 800°C for one hour and cooled in a dessicator just prior to use in order to ensure the stoichiometry of U_3O_8.

The fraction of NBS SRM 950b was weighed directly into a flask. The fraction of SRM U-970 was weighed in a recipient and subsequently transferred into the same flask. One unit of NBS SRM-949f plutonium reference material was added to the flask, after verification of the certified mass. All operations were performed in a specially equipped weighing glove-box.

Nitric acid was added to obtain final uranium and plutonium concentrations of about 19.4 mg U/g and 0.6 mg Pu/g solution.

Aliquoting

Each solution was aliquoted by transfer into 50 ml syringes in order to avoid evaporation losses. Metrological aliquots of 2.5 g or 5 g were subsequently weighed into borosilicate glass 10 ml penicillin vials (approximately 250 units per batch). Weighings were carried out on a Mettler AE-163 analytical balance, by weighing the syringe before and after transfer of the aliquot into the recipient.
After dispensing the whole batch, the content of each unit was evaporated until dryness on a hot plate. After addition of a THF solution, each unit was heated until dryness at 60°C. A glassy material was formed which adhered very well to the walls of the vial. The vials were then closed with a fluoelastomere (isoversinic) stopper and aluminium tear-off seal. The outside wall of each vial was checked for radioactive contamination. After sealing in a plastic bag CBNM IRM-1027 is ready for transport. This procedure was used to prepare two batches :

CBNM IRM-1027a :	144 units containing about 47.5 mg uranium and 1.6 mg plutonium 96 units containing about 97 mg uranium and 3 mg plutonium
CBNM IRM-1027b :	160 units containing about 47.5 mg uranium and 1.6 mg plutonium 68 units containing about 97 mg uranium and 3 mg plutonium

Provisional certification

Preliminary certification values are based on the metrological data of the weighings and the certified values of the base materials.

Weighings:
SRM 950b : (15.734 5 ± 0.001 0) g material (U308)
SRM U-970 : (3.813 67 ± 0.000 10) g material (U308)

Calculated amount uranium: (16.565 7 ± 0.003 1) g uranium
SRM 949f (tube #164) : (0.511 52 ± 0.000 05) g material (Pu)
Total amount of solution : (854.175 ± 0.010) g nitric acid 5M

Isotopic composition (uranium):

	mole %	mass %	uncertainties (2s)
234U	0.332 3	0.327 5	0.000 3
235U	19.797 6	19.597 7	0.002 2
236U	0.030 3	0.030 1	0.000 1
238U	79.839 8	80.044 6	0.002 2

Isotopic composition (plutonium):

	mole %	mass %
238Pu	0.003 7	0.003 7
239Pu	97.148 7	97.136 9
240Pu	2.802 5	2.813 9
241Pu	0.039 1	0.039 4
242Pu	0.006 0	0.006 1

Certified concentration:
(19.393 8 ± 0.003 6) mg U/g solution
(0.598 44 ± 0.000 31) mg Pu/g solution

Table 2 - Certification data for CBNM IRM-1027a

Weighings:
SRM 950b : (13.981 2 ± 0.001 0) g material (U308)
SRM U-970 : (3.403 72 ± 0.000 10) g material (U308)

Calculated amount uranium: (14.732 5 ± 0.002 7) g uranium
SRM 949f (tube #164) : (0.454 68 ± 0.000 05) g material (Pu)
Total amount of solution : (757.290 ± 0.010) g nitric acid 5M

Isotopic composition (uranium):

	mole %	mass %	uncertainties (2s)
234U	0.333 4	0.328 6	0.000 3
235U	19.865 4	19.665 0	0.002 2
236U	0.030 4	0.030 2	0.000 1
238U	79.770 7	79.976 1	0.002 2

Isotopic composition (plutonium):

	mole %	mass %
238Pu	0.003 7	0.003 7
239Pu	97.148 7	97.136 9
240Pu	2.802 5	2.813 9
241Pu	0.039 1	0.039 4
242Pu	0.006 0	0.006 1

Certified concentration:
(19.454 2 ± 0.003 6) mg U/g solution
(0.599 99 ± 0.000 31) mg Pu/g solution

Table 3 - Certification data for CBNM IRM-1027b

Verification measurements

Verification measurements of the metrologically established certified value for element assay of uranium and plutonium will be performed by IDMS.

3. Metal Spikes

Metal spikes are easily handled during the spiking operation in the hot cell and offer the advantage that the excess uranium reduces all the plutonium to Pu^{4+} in solution obviating the need for a redox cycle.

Uranium-plutonium alloys are made by crucible-less induction melting /6/ usually with a 10 to 15 g sample supported on a water-cooled copper pedestal. There is no significant contamination of the melt and, with an atmosphere of 5 x 10^4 Pa argon, evaporation losses are small. This method can be described as "quantitative alloying" with the alloy composition determined by the mass of the components, and the uncertainty on the composition calculated from the mass loss, should this be measurable. During melting, vigorous electromagnetic stirring occurs, and combined with relatively rapid cooling and solidifation, ensures good homogeneity in the button. Complete solubility of plutonium in the uranium matrix means that even micro-inhomogeneity should not occur.

The uranium-plutonium alloys so far produced (0.1-5 % plutonium) are brittle and the spikes have been prepared by breaking the alloy buttons under a press into pieces of about 100 mg. These are weighed and sealed individually into glass ampoules.

Spikes are characterised by IDMS and the homogeneity of past series of metallic spikes (RSD of plutonium content) has been determined as 0.17-0.5 %. The uncertainty statement is limited by the measurement method. Metal spike homogeneity has also been investigated by gamma spectrometry on both solid and dissolved samples /7/. In order to reduce the effect of the variation in self-absorption arising from the irregular shapes of the individual spikes in solid form, the specific count rate ratio $R_1 = Pu_1/U_1$ was calculated from the 185.72 keV U count rate and the 129.28 KeV Pu count rate. However, because the change in self-absorption for different spike masses depends on gamma-ray energy it is necessary to make a further correction using the specific count-rate ratio $R_U = U_1/U_2$, where U_2 is the specific count rate for 143.76 KeV gamma rays i.e. this uses the ^{235}U in the spike as an internal standard. A corrected R_1 value can then be calculated :

$$R_{IC} = R_1\left[1 + \frac{R_{Ua} - R_U}{R_U} \cdot \frac{\Delta E_1}{\Delta E_3}\right]$$

where

$\Delta E_1 = E_\gamma U_1 - E_\gamma Pu_1 = 56.44$ KeV

$\Delta E_3 = E_\gamma U_1 - E_\gamma U_2 = 41.96$ KeV

and R_{Ua} is the R_U value for a spike of average mass.

This method has been demonstrated using spikes of U-3% Pu alloy with ^{235}U enrichment of 93 %. The RSD of R_{1C} (indicating Pu content) achieved with solid samples was 0.5 % and with dissolved samples was 0.2 %, which was comparable with the RSD achieved by IDMS carried out on the same solutions. A series of gamma counting results for liquid samples are given in Fig. 2, showing that the correction factor for the R_1 ratio eliminates the influence of spike mass.

Current work is concentrating on the development of U-Pu-X metallic spikes that can be fabricated into foil or wire form by appropriate cold work and annealing. The advantage of this type of improved spike is better homogeneity following the mechanical working and spikes of more regular shape; either discs or wires. This reduces random errors in gamma counting arising from irregular spike shapes, and allows more efficient quality control of the solid spikes.

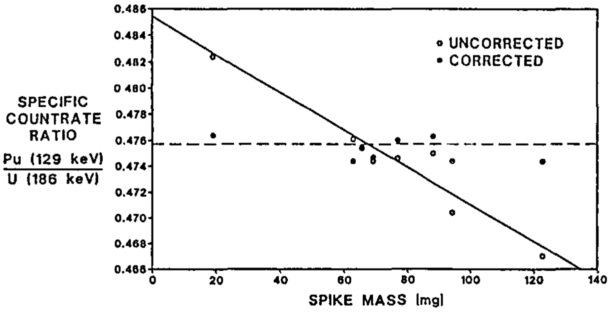

Fig. 2 - Correction of count rate ratio R_1 for spike mass using U-235 as internal standard

4. Spike Optimization

The precision of a uranium or plutonium assay by IDMS depends not only on the quality of the spike, but it is also limited by the precision ε with which isotope ratios can be measured and by the propagation of uncertainties in the subsequent calculation of concentration. The effect of using spikes of unsuitable size or isotopic composition can be dramatic and it is important to optimize the spike to suit the input solution to be assayed. The uncertainty propagation for the case of bi-isotopic samples and spikes is discussed in ref. 8 and is considered here for $^{240}Pu/^{239}Pu$ and $^{235}U/^{238}U$ ratio measurements on spiked input solutions. The error magnification factor (emf), when multiplied by ε, gives the expected relative precision $\sigma(q)/q$ of a measurement of element concentration in the sample; $q = N_T/N_S$, where N_T the number of atoms of that element in the sample and N_S is the number in the spike.

Figure 3 shows the influence on emf of uranium and plutonium content in the spike. Two curves for each are shown representing the maximum and minimum concentrations from LWR input solutions. In this example the isotopic composition of the sample corresponds to a fuel of 30 000 MWd/t burn-up and 3.3 % initial enrichment. The lowest attainable emf in this case is about 2. The optimum uranium and plutonium content in the spike are directly proportional to the corresponding concentration in the sample. In Fig. 4 the effect of the isotopic composition of the sample is investigated, assuming typical average values of uranium and plutonium concentrations in the sample. The emf minima, representing optimum spike compositions, lie between 1.5 and 3 for the extremes of isotopic composition taken.

These curves indicate that the optimum composition of the CBNM solid spikes(^{239}Pu/Pu

= 0.97, ^{235}U/U = 0.20) is Pu/Pu+U = 0.025 - 0.1. However, the selection of a single spike composition remains a compromise and the IDMS error magnification factor could be at least a factor of two above the lowest achievable value in unfavourable cases.

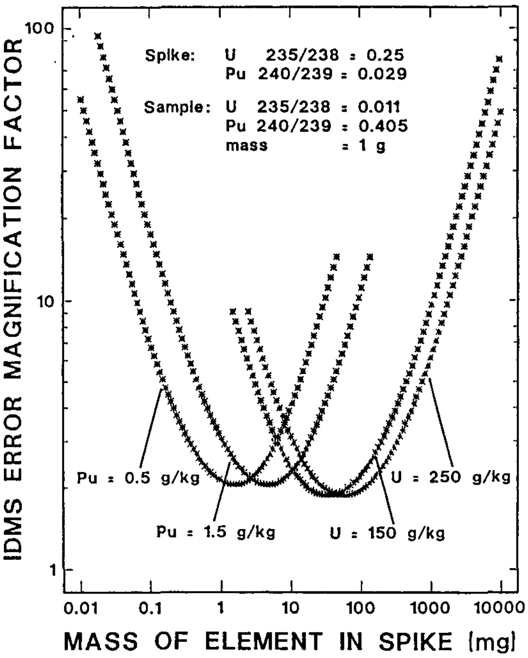

Fig. 3 - IDMS error magnification curves for input concentrations : U = 150-250 g/kg, Pu = 0.5 - 1.5 g/kg

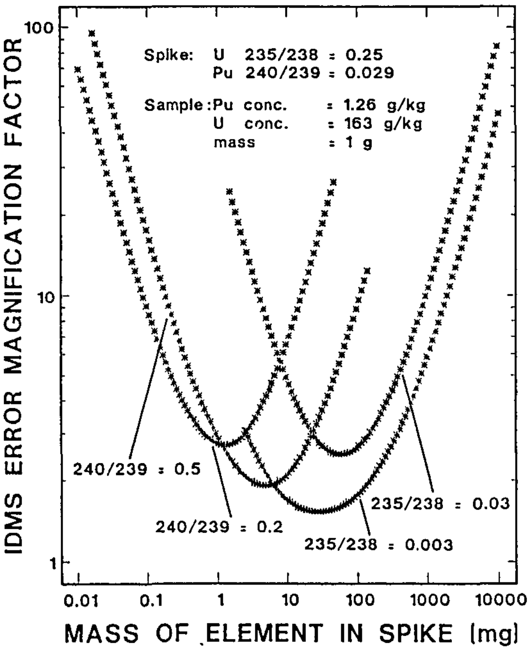

Fig. 4 - IDMS error magnification curves for input isotopic compositions :
U 235/238 = 0.003 - 0.03,
Pu 240/239 = 0.2 - 0.5

5. Conclusions

CBNM solid spikes prepared using 97 % enriched ^{239}Pu and 20 % enriched ^{235}U fall within the optimum composition range (Pu/U+Pu = 0.025 - 0.1) for LWR input solutions. A batch of large size dried U/Pu nitrate samples has been prepared and certified as reference materials CBNM IRM-1027 a and b. Metallic U/Pu spikes have potentially excellent homogeneity and can be improved to allow routine quality control by gamma counting of solid samples.

Acknowledgements

The authors would like to thank R. Eykens for assistance with the gamma spectrometry and F. Peetermans for carrying out the alloying.

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MOX PELLET REFERENCE MATERIAL

présenté par Jean-Pierre PEROLAT

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Abstract

A first batch of MOX pellets certified in plutonium and uranium has been prepared and characterised in France to meet the needs of laboratories which are engaged upon destructive analysis for safeguards purposes especially in fuel fabrication plants. The pellets sintering has been obtained in a special fabrication to achieve an homogeneity better than 0,1 %. The plutonium and uranium characterisation by chemical analysis has been carried out by two laboratories using at least two different methods.

1. Introduction

Les combustibles d'oxyde mixte $(U,Pu)O_2$ permettent de recycler le plutonium dans les réacteurs à eau légère. Alors que la production de combustible pour réacteurs à neutrons rapides stagne, ces combustibles sont appelés à un grand développement. Leur fabrication, le contrôle des garanties, nationales ou internationales font appel à des matériaux de référence, en particulier dans le domaine de l'analyse destructive.

La CETAMA engagée depuis de nombreuses années dans la réalisation de matériaux de référence /1/ a entrepris la réalisation de pastilles frittées $(Pu, U)O_2$ du type MOX certifiées en uranium et plutonium. Cette réalisation, coordonnée par le Secrétariat de la CETAMA a été effectuée au sein d'un de ses groupes de travail. L'objectif visé dans un premier temps est de fournir aux laboratoires d'analyse destructive des références représentatives pour ce qui concerne la teneur en plutonium des lots de pastilles présents et futurs.

2. Produits de départ

Pour la réalisation de ce lot, les poudres d'oxyde d'uranium et d'oxyde de plutonium ont été choisies pour leur bonne aptitude au frittage. En particulier l'oxyde d'uranium UO_2 en provenance de l'atelier du CE/CADARACHE a été obtenu par le procédé voie humide ADU, dont les poudres qui en sont issues présentent une excellente aptitude au frittage. L'oxyde de plutonium provient du retraitement de combustible de réacteurs à eau légère. Un produit "frais" à faible teneur en américium a été choisi.

Les compositions isotopiques qui ne sont pas certifiées, ont cependant été mesurées avec une justesse suffisante pour la détermination des masses atomiques moyennes. La teneur en américium, le rapport O/M ont également été déterminés.

3. Fabrication des pastilles

Le procédé adopté est celui du cobroyage direct, dit procédé "COCA". Utilisé au CE/CADARACHE pour la fabrication des combustibles de réacteurs à neutrons rapides, il conduit, sur une base industrielle, à l'obtention d'une homogénéité interpastilles satisfaisante /2/. Un matériau de référence (appelé "VENDEMIAIRE") certifié aux valeurs suivantes en 1983 :

- U : (74,83 ± 0,04) %
- Pu : (13,025 ± 0,30) %

fabriqué par ce même procédé présentait une hétérogénéité interpastilles de ± 0,025 %.

Compte tenu des caractéristiques des combustibles actuels ou en projet, la teneur en plutonium souhaitée pour cette nouvelle fabrication a été fixée à 6 %, sans toutefois vouloir obtenir une valeur précise.

En raison de l'objectif visé et de difficultés éventuelles dues à la faible teneur en plutonium, ce travail a été confié à un Laboratoire de Recherche et Développement, afin de mieux choisir et contrôler les différents paramètres du procédé.

En particulier le cycle de frittage, effectué sous atmosphère réductrice, d'une durée de 40 heures a comporté un palier de 4 heures à 1750°.

Des contrôles ont été effectués au cours des différentes étapes de la fabrication, notamment :

- la granulométrie des poudres initiales et des mélanges (UO_2, PuO_2) après broyage,
- la métrologie des comprimés crus.

Après frittage, les caractéristiques suivantes ont été examinées :

- métrologie des pastilles,
- aptitude à la dissolution,
- structure métallographique,
- homogénéité de la solution solide $(U, Pu)O_2$ à l'échelle macroscopique par autoradiographie et à l'échelle microscopique par analyse à la microsonde. Comme on peut le voir sur la figure 1 le résultat est satisfaisant ; les profils de concentration en U et Pu obtenus sur une traversée diamétrale de la pastille indiquent des répartitions assez homogènes, avec toutefois quelques surconcentrations locales, de faibles diamètres (~20 µm) et d'une teneur évaluée à 15% .

4. Contrôle de l'homogénéité

Ce contrôle a été effectué sur 12 pastilles prélevées au hasard. Chaque pastille a été dissoute et

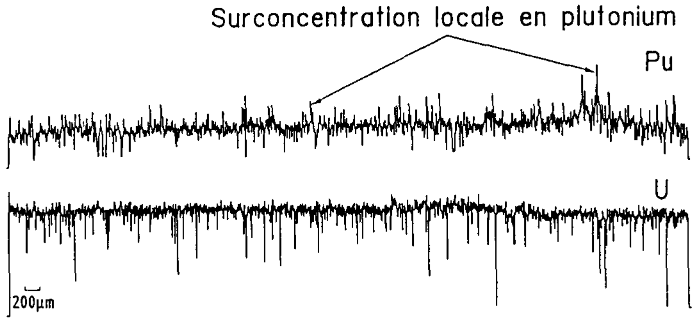

Fig. I - Profils de concentration sur section diamétrale

trois déterminations de la teneur en plutonium ont été effectuées.

La méthode utilisée est celle dite à "l'oxyde argentique" /3/. Au total 36 dosages ont été effectués dans un ordre aléatoire de façon à écarter l'influence d'une dérive éventuelle au cours du temps, la campagne d'analyse ayant duré environ un mois et demi.

L'analyse de variance a un facteur contrôlé /4/ effectué sur les 36 résultats conduit à un écart type d'homogénéité non significatif et estimé à une valeur nettement inférieure à 0,1 %.

5 Mesures de certification

Pour ce premier lot, on ne certifie que les teneurs en plutonium et en uranium, en faisant appel à deux laboratoires, les méthodes employées étant les suivantes :

- Plutonium : coulométrie,
  titri-gravimétrie /5/
  méthode à "l'oxyde argentique" /3/
- Uranium : dilution isotopique, spectrométrie de masse,
  méthode au titane,
  Davies et Gray.

Les impuretés interférant dans ces méthodes font l'objet d'analyses spécifiques.

6. Resultats

Les valeurs provisoires, au stade actuel, toutes les mesures n'étant pas terminées sont les suivantes :

- Pu : (5,7335 ± 0,0048) %
- U : (82,18 ± 0,06) %

7. Conclusions

Les résultats obtenus sur ce premier lot sont satisfaisants. Un second lot de taille plus importante, environ 800 pastilles, est prévu, de mêmes caractéristiques sauf pour ce qui concerne le plutonium, qui sera très riche en ^{239}Pu (~ 91,5%). Ce choix présente les avantages d'éviter une teneur en ^{241}Am importante au bout de quelques années, et donc d'assurer une pérennité du matériau de référence sur plus de 10 ans. Par contre, sur ce point n'étant pas représentatif des fabrications, il n'est pas appproprié aux analyses non destructives, à l'exception éventuellement de la fluorescence - γ - X_K.

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CERTIFICATION OF A PLUTONIUM DIOXIDE REFERENCE MATERIAL FOR ELEMENTAL ANALYSES
(EC-NRM 210)

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Abstract

A new EC plutonium reference material is made available in the form of 5 g samples of plutonium dioxide powder. The plutonium content (880.26 ± 0.44) $g \cdot kg^{-1}$ has been derived from plutonium measurements performed by three different laboratories applying each a different oxydo-reductive method. The results of the plutonium measurement, the statistical evaluation of the uncertainty of the plutonium content together with information on the impurities present in the material are given in the contribution.

1. Introduction

The reference materials essential to guarantee the elemental analyses of uranium and plutonium were among the high priority needs demonstrated by an inquiry in the European Community (EC) /1, 2/.

The first EC material with a certified plutonium content was a well characterized reference plutonium metal (EC-NRM 201) having a plutonium mass fraction known with an uncertainty of 0.5 $g.kg^{-1}$ /3/.

On the basis of the information collected during the inquiry on the needs, CBNM, in collaboration with national experts, came to the conclusion that the production of a second certified reference material in the form of a dioxide seemed necessary. It was felt that this second material, although less stringently characterized, would be very useful to those laboratories that perform a large number of control analyses.

Today the need of this material has become more imperative because it is intended to replace the EC-NRM 201, the stock of which is exhausted, and thus to be the unique material with an EC certified plutonium content.

2. Base material

A batch of plutonium dioxide powder was produced in the BNF plc nuclear plant by thermal decomposition of purified plutonium oxalate at a temperature of about 550°C followed by an ignition at 1250°C /4/. The powder was subdivided and filled into polythene container, each containing about 5 g PuO_2.

3. Characterization of the material

3.1 Working scheme

Bearing in mind the considerable efforts required to obtain reliable results for a characterization via the substraction of the total impurities (100-X method) the plutonium working group experts agreed to perform direct plutonium measurements for the characterization of the material and to take the figures on impurities for a rough verification.

The members of the Working Group, discussing a scheme for the plutonium measurements, agreed that:

- the number of measuring laboratories should be at least three;
- the applied methods should call on different oxido-reduction systems and should have different responses to the impurities present in the material;
- the participating laboratories should be dispersed through-out the different EC member countries.

The following scheme was adopted: the work accomplished by each particular laboratory was discussed by the experts on the basis of a report on the analysis of the material and the results were combined to obtain the certified plutonium value for the mass fraction, and the connected uncertainty.

3.2 Homogeneity of material

The approach choosen was to derive a figure characterizing the inhomogeneity from the between samples variance in one particular lab. The results of lab 3 were the most appropriate because it shows the best precision in replicating the measurements on 12 different samples. As a consequence, the inhomogeneity of the material is characterized by a standard deviation of 0.013 weight percent plutonium.

3.3 Americium content

The americium content at the date of Dec. 8, 1978 (period of preparation of the material) was 10 $mg.kg^{-1}$. On May 30, 1980 it was measured by gamma-spectrometry and found by two different laboratories to be equal to 205 and 238 $mg.kg^{-1}$, respectively. The americium content calculated from the decay of Pu-241 [for the same date] was 228 $mg.kg^{-1}$. When discussing these results it was agreed to include in the certificate, a recommendation for calculating the americium content by decay from the isotopic composition at the date of preparation of the material. The americium at the date of

(30.05.1988) was calculated as being 1170 mg.kg^{-1} metal to plutonium metal. The decay data used for the calculation of the americium content are given in /5/.

3.4 Uranium content

The uranium content at the date of Dec. 8, 1978 was found to be less than 1 mg.kg^{-1}. It was measured as being respectively 83 and 85 mg.kg-1 by two different laboratories on May 30, 1980. As for the americium it will be recommended on the certificate to calculate the uranium content by decay of the plutonium isotopes. The uranium content at the date of certification (30.05.1988) was calculated as being 325 mg.kg^{-1} relative to plutonium dioxide.

3.5 Iron content

The iron content has been measured by different laboratories using different methods. The mean of all sample measurements is 34 mg.kg^{-1} iron relative to PuO_2. The mean of the spectrophotometric measurements taken separately is also 34 mg.kg^{-1}.

3.6 Impurity measurements

The concentration ranges of the major and minor impurities in the material have been determined mostly using spark source mass spectrometric, spectophotometric and emission spectographic methods.

After combining the lowest limits of the major and minor impurities (expressed as oxides), the detection limits of the non-volatile impurities as well as the uranium and americium contents (around the time of May 30. 1988), a lowest limit of the total impurities of about 1690 mg.kg^{-1} has been estimated.

A similar calculation, taking into account the highest (instead of the lowest) limits of the major and minor impurity leads to a figure of about 1790 mg.kg^{-1} as the highest limit of the total impurities.

Using a stoichiometric factor of 0.881975 (oxygen to metal ratio measurements have shown that the values obtained on the calcined material are within ± 0.001 atoms oxygen per metal atom of stoichiometric PuO_2) to calculate the plutonium content of the oxide it can be estimated that the plutonium content of the material is between 88.039 (88.04) and 88.048 (88.05) weight percent.

3.7 Isotopic composition of the plutonium

The isotopic composition of the material at the date of Dec. 8, 1978 was the following

Pu-238	0.013 atom percent
239	93.329
240	6.298
241	0.319
242	0.041

The atomic weight calculated for this date is 239.121. It was agreed to report on the certificate an atomic weight of 239.12.

3.8 Loss on heating at 1250°C

To be mentioned briefly the figures reported by different laboratories are: 0.18, 0.05, 0.20 and 0.04 %.

3.9 Plutonium content

Scheme of the analytical campaign. Three laboratories (CEA, Fontenay aux Roses, BNFL, Sellafield and JRC, Geel) took part in the determination of the plutonium content of the material.

It was agreed that the number of samples to be analyzed by each laboratory would be up to twelve. Three results were expected from each dissolution thus leading to thirty six measurements per laboratory.

Four bottles containing five grammes of plutonium dioxide each were put at the disposal of each of the laboratories, thus allowing them to perform at least three samplings per bottle. It was also agreed that the three participating laboratories should use the EC Nuclear Reference Material 201 (piece of plutonium metal) to determine a calibration factor which should be used to calculate the titration values obtained on the PuO_2 samples.

In the certification report /6/ of the plutonium RM the plutonium content and its confidence intervals are expressed as

$$99.976 \pm 2\sqrt{(0.0115)^2 + \frac{1}{n}(0.0225)^2}$$

for a confidence level of 95 percent, n being the number of samples of reference material used. The first component is the uncertainty inherent to the plutonium determination in the reference material. The second component is inherent to the inhomogeneity of the reference material and is of random nature.

As each of the three collaborating laboratories have used two samples of reference material for their calibration this component becomes $(0.0225)/\sqrt{2}$.

Working procedures. A summary of each working procedure /7 to 10/ is given in the following. The calibration factor (its mean being expressed in the relevant unit according to the working procedures followed by the lab) is calculated from a given number of repetitions and accompanied by a statement on the uncertainty of this factor. This statement includes both the uncertainty of the reference material as stated above and the uncertainty of the calibration measurements themselves; it corresponds to a confidence interval for a confidence level of about 95 percent. This uncertainty will, later on, be converted into an uncertainty figure attached to the plutonium content.

Laboratory 1

Method: The sample of plutonium oxide heated in a platinium crucible at 1250 °C during two hours and accurately weighed is dissolved in a mixture of hydrochloric and hydrofluoric acids into a ptfe dish (duration: about half a day). The solution is transferred to a polythene bottle. A weighed fraction of the solution is evaporated to near dryness in the presence of a small amount of sulphuric acid, and it is then diluted with a mixture of 0.5 M sulphuric/1M nitric acids. To this solution is added excess titanium (III) chloride solution to reduce plutonium to the trivalent state. The mixture is then allowed to stand until the excess titanium (III) ions have been oxidised by the nitric acid whilst the plutonium remains as plutonium (III). The progress of these reactions is followed potentiometrically by means of platinum and calomel electrodes. The plutonium (III) is then oxidised quantitatively to plutonium (IV) by means of a cerium (IV) sulphate solution which is added by weight burette until a small excess is present. The excess cerium is titrated with a iron (II) ammonium sulphate solution, and the equivalence point is measured potentiometrically.

Calibration: The titrations were completed with titrations prepared from the EC-NRM 201 reference material. These titrations were carried out at the beginning, middle and end of each run. Twelve fractions of plutonium EC-NRM 201 reference material solution were taken for calibration. The mean calibration factor with its confidence interval at a 95 percent confidence level is (11.1405 ± 0.0054) mg Pu/g cerium sulphate.

Laboratory 2

Method: The sample of plutonium dioxide heated in a platinum crucible at 1250°C during two hours and accurately weighed is dissolved (duration: three to five days) under reflux either in 11 M HNO_3 / 0.05 M HF (first two samples) or in the same mixture completed with a perchloric acid addition (a further seven samples). The solution is evaporated to near dryness and is then diluted with 1 M nitric acid. To a weighed aliquot of this solution is added (300 ± 10) mg of silver (II) oxide to oxidize the plutonium to the hexavalent state, the excess of silver (II) oxide being destroyed by heating the solution. The plutonium (VI) is reduced to the quadrivalent state by a known amount of iron (II) solution. The excess of iron (II) is determined using a cerium (IV) solution as titrant. The equivalent point is measured spectrophotometrically (510 nm) by recording the change of colour of iron (II) orthophenanthroline (the amount added being constant for each measurement).

Calibration: The oxidizing power of the cerium sulfate solution used as titrant has been determined using the plutonium EC-NRM 201 reference material. Seven fractions of this plutonium reference solution have been taken for the calibration. The mean calibration factor with its confidence interval at a 95 percent confidence level is $(0.94672 \pm 0.0058).10^{-5}$ equivalent per gramme.

Laboratory 3

Method: The sample of plutonium oxide heated in a platinum crucible at 1250°C during two hours and accurately weighed is dissolved under reflux (duration: one day) in a mixture of hydrochloric and hydrofluoric acid (11M HCl/0.05M HF). The solution is evaporated in the presence of sulfuric acid (three times) and is diluted with 0.5 M sulfuric acid. A weighed aliquot of this solution is transferred into the coulometric cell. The plutonium is reduced at + 0.230 V via the saturated calomel electrode until a constant background current of about 4 µA is obtained. The solution is then oxidized at + 0.710 Volt until a similar background current is obtained. The first step is carried out in 180 to 220 minutes and the second step in 70-75 minutes. After being corrected for the background effect the read-out is converted into an plutonium amount.

Calibration: A chemical calibration factor has been determined by titrating eight aliquots of EC-NRM 201 reference plutonium solution. The mean calibration factor with its confidence interval at a 95 percent conference level is (40 345 ± 14) counts/mg Pu. The electrical calibration factor is 99.966 counts per coulomb and corresponds to 40.341 counts per mg Pu. The difference between the two factors (about 0.01 percent) is not significant. As a calibration via the EC-NRM 201 reference material was requested from each laboratory the results obtained by coulometry had to be reported on the basis of the chemical calibration. Keeping in mind the difference between the two calibration factors it seemed relevant to take half of it into account (arbitrarely assuming it as a random error) so as to include that half in the overall uncertainty of the calibration. By doing so only a slight increase of the overall uncertainty of the calibration is obtained. This additional uncertainty is included in the uncertainty figure quoted above.

4. Statistical evaluation of the results

4.1 Statistical tests

A number of statistical tests have been applied to the results reported by the three collaborating laboratories /11/. All tests have been applied at a probability level of 95 percent.

- Outlier test on all the replicates of a particular laboratory, (Dixon test).
- Test of equality of variances, (Cochran test).
- The within-samples variance which characterizes the spread of a replicate measurement on a particular sample.

4.2 Estimation of the uncertainty of the plutonium content in a particular laboratory

- Estimation of the uncertainty of the experimental results (scheme of evaluation): for both the calibration factor (F) and the plutonium content of oxide (C) the mean-value (M_F or M_C) and its uncertainty have been derived from the following approach:
  $M = \bar{m} + 2\ \sigma_M$ where:
  $\bar{m}$ is the mean of the n determinations of a particular laboratory, σ being the corresponding standard deviation,

σ_{M_F} being equal to $\sqrt{\sigma^2_{RM} + \frac{1}{n}\sigma^2_F}$ and

σ_{M_C} being equal to $\sqrt{\frac{1}{n}\sigma^2_C}$

σ_F or σ_C is deduced from the estimated standard deviation s by applying the relation $\sigma = s.t/2$ where t is the Student factor corresponding to (n-1) degrees of freedom for a 95 % probability level.

- Estimation of the random uncertainty of the calibration factor:

  For the calibration factor (F) the random uncertainties attached to the plutonium content value are given in table 1.

Laboratory	σ_{M_F}
1	0.021
2	0.027
3	0.016

Table 1

- Estimation of the random uncertainty of the plutonium measurements:

  The random uncertainties of the plutonium content measurements of the oxide (C) are given in table 2.

Laboratory	σ_{M_C}
1	0.023
2	0.029
3	0.008

Table 2

σ_{M_C} is expressed as a figure attached to the plutonium content value.

- Estimation of the random uncertainty of the plutonium content:

  The random uncertainty of the plutonium mean value in a particular laboratory is obtained by combining according to the law of error propagation, the uncertainties σ_{M_F} and σ_{M_C}. These combined uncertainties σ_M are given in table 3 together with the plutonium content found.

Laboratory	Pu content M in wt %	Random uncertainty of the Pu content σ_M
1	88.041	0.031
2	87.965	0.040
3	88.036	0.018

Table 3

A test of comparison of the means using the figures given in table 3 (calculating $u_p = \frac{x_1 - x_2}{\sqrt{\sigma^2_{x_1} + \sigma^2_{x_2}}}$) shows that the means are sufficiently homogeneous for the treatment reported in section 4.3.

4.3 Evaluation of the plutonium content and overall uncertainty

The most probable value of the mean plutonium content is given by the weighed mean of the plutonium contents found respectively by the three participating laboratories. The statistical formula used to calculate this weighed mean (M) is the following:

$$M = \frac{\frac{M_1}{\sigma_1^2} + \frac{M_2}{\sigma_2^2} + \frac{M_3}{\sigma_3^2}}{\frac{1}{\sigma_1^2} + \frac{1}{\sigma_2^2} + \frac{1}{\sigma_3^2}}$$

The random uncertainty attached to M is derived from

$$\sigma_M^2 = \frac{1}{\frac{1}{\sigma_1^2} + \frac{1}{\sigma_2^2} + \frac{1}{\sigma_3^2}}$$

Using the set of figures given in table 3, following values are obtained:

$M = 88.026$

$\sigma = 0.015$

When taking into account the uncertainty (first component, see section 3.9) inherent to the reference material the overall uncertainty of the plutonium content is characterized by a standard deviation s given by $s^2 = (0.015)^2 + (0.0102)^2 = (0.018)^2$.

As the samples are unique (about 1 g oxide) at the time of the sampling in a particular laboratory the uncertainty on this unique sample will then be characterized by a variance σ_u^2 resulting from a total of two variances σ_1^2 and σ_2^2. The variance σ_1^2 is attached to the plutonium mean-content of the material. The variance σ_2^2 is attached to the heterogeneity of the material:

$\sigma_u^2 = \sigma_1^2 + \sigma_2^2$

This heterogeneity is characterized by a variance which has been derived from an analysis of variance applied to the results of laboratory 3 which shows the lowest within-samples variance (see section 3.2). The resulting variance is $\sigma_2^2 = (0.013)^2$. As σ_1^2 is is equal to $(0.018)^2$ the variance σ_u^2 becomes:

$\sigma_u^2 = (0.018)^2 + (0.013)^2 = (0.022)^2$.

An overall uncertainty of the plutonium content is obtained by multiplying the corresponding standard deviation, 0.022, by a factor two. This corresponds to forming a confidence interval for a confidence level of about 95 percent.

Thus the plutonium content is (88.026 ± 0.044) weight percent.

5. Certified value

On the basis of the results given previously the plutonium mass fraction (880.26 ± 0.44) $\mathrm{g \cdot kg^{-1}}$ is certified. An overall uncertainty of the plutonium mass fraction of an individual one gramme sample was estimated by combining the standard deviation of the plutonium measurements, the standard deviation of the calibration measurements, the standard deviation of the plutonium content of the reference material and the standard deviation characterizing the random variation of the plutonium mass fraction between samples (inhomogeneity) according to the law of error propagation.

6. List of the members of the Working Group

The EC Working Group was composed of representatives from:

- British Nuclear Fuels plc,
  Sellafied, United Kingdom
- Bundesanstalt für Materialprüfung,
  Berlin, Federal Republic of Germany
- Commissariat à l'Energie Atomique, CEA
  Centre d'Etudes Nucléaires
  Fontenay-aux-Roses, France
- Centre d'Etude de l'Energie Nucléaire/
  Studiecentrum voor Kernenergie,
  Mol, Belgium
- Centro di Studi Nucleari, CNEN,
  Casaccia, Italy
- Joint Research Centre,
  Central Bureau for Nuclear Measurements,
  Geel, Belgium
- Netherlands Energy Research Foundation,
  ECN, Petten, The Netherlands
- Risø National Laboratory,
  Roskilde, Denmark
- United Kingdom Atomic Energy Authority,
  Harwell, United Kingdom
- Université de Liège,
  Chimie Analytique et Radiochimie,
  Liège, Belgium
- University of Dublin,
  Trinity College,
  Dublin, Ireland

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A ^{244}Pu SPIKE REFERENCE MATERIAL CBNM IRM-042A

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Abstract

A highly enriched ^{244}Pu Isotopic Reference Material (CBNM IRM-042a) has been prepared and certified for ^{244}Pu isotope concentration. The certified value of (2.257 7 ± 0.004 4).10^{18} atoms $^{244}\mathrm{Pu}\cdot\mathrm{kg}^{-1}$ of solution has been established by isotope dilution mass spectrometry.

The plutonium isotopic composition has been determined by thermal ionization mass spectrometry and calibration of these measurements by means of synthetic ^{242}Pu/^{239}Pu mixtures.

The Isotopic Reference Material is supplied in a sealed glass ampoule containing approximately 10 g of a 5 M nitric acid solution at an approximate concentration of 1µg Pu per g solution.

This Isotopic Reference Material is part of a systematic CBNM programme to supply spike isotopic reference materials of various isotopes at different concentrations.

1. Introduction

^{244}Pu is an isotope which is not normally present in current Pu materials. It is, therefore, used sometimes in Isotope Dilution Mass Spectrometry (IDMS) : both the Isotope Dilution measurement and the isotopic composition can be performed in one measurement.

However, it is a very scarce material : the world's stock is probably only a few grammes. Its use in IDMS does NOT increase the accuracy of the measurement over the use of ^{242}Pu. It should, therefore, not be used in routine analysis, but in special cases only.

CBNM has proceeded to make available an independent Spike Reference Material for such uses. Its characterisation was achieved against well known ^{239}Pu solutions.

2. Preparation of the Candidate ^{244}Pu Isotopic Reference Material

A mother solution was prepared starting from 10 mg ^{244}PuO$_2$ (ORNL batch No. Pu-244-315A). By dissolution in 5 M HNO$_3$, a concentration of about 40 µg $^{244}\mathrm{Pu}\cdot\mathrm{g}^{-1}$ solution was obtained. A weighed fraction of this stock solution was diluted with distilled water in a glass flask, leading to a concentration of about 1 µg $^{244}\mathrm{Pu}\cdot\mathrm{g}^{-1}$ solution. Using a dispenser, 10 ml aliquots of this solution were distributed in glass ampoules in a special "^{244}Pu" glove box. Subsequently the ampoules were transferred out of the glove box and sealed in an oxy-acetylene flame in a fume hood, under adequate Health Physics protection and control. About 230 ampoules were prepared.

3.Preparation of the ^{239}Pu solution

NBS Standard Reference Material, SRM 944 with plutonium content certified as (47.50 ± 0.05) weight percent of plutonium, was used to prepare a solution with known ^{239}Pu concentration. The solution, coded as SRM-944/25, was achieved by dissolving the plutonium sulphate tetrahydrate in 5 M nitric acid to obtain a concentration of 23.605 ± 0.026 mg $^{239}\mathrm{Pu}\cdot\mathrm{kg}^{-1}$ solution.

4. Preparation of the blends for certification measurements

A series of 6 ampoules were randomly taken from the batch of candidate reference material CBNM IRM-042a. A weighed fraction from each ampoule was transferred to a recipient and spiked with a weighed fraction of SRM-944/25. Weighings were carried out using a syringe which was weighed before and after transfer of aliquots into the recipient. After mechanical mixing the samples were stored to be chemically treated before mass spectrometric control measurements.

All uncertainties given are 2s where s = standard deviation of the single determination. They are indicated under the digits of the values to which they apply. A determination does not only include the measurement but also a full independent chemical preparation of the sample and - in the case of isotope dilution - an independent spiking.

Blend	CBNM IRM-042a ampoule number	mass CBNM IRM-042a solution (in g)	mass SRM 944/25 solution (in g)
1	12	5.249 02	2.040 42
2	54	5.596 72	2.110 42
3	100	5.316 32	2.124 42
4	129	5.508 42	2.049 72
5	195	5.327 42	2.102 62
6	244	5.435 92	2.112 82

Table 1 - Metrological data of weighings

5. Chemical Preparation Prior to Mass Spectrometric Measurements

Before mass spectrometric analysis an appropriate chemical preparation has to be applied in order to condition the samples to be analysed and to give them all a similar chemical pre-history. Separation from impurities is also carried out at this occasion.

The solutions were evaporated to dryness and dissolved in 200 µl 2M HNO_3. To achieve valency homogeneization in the Pu^{4+} state, 50 µl 1.25M $FeCl_2$ was added to convert all Pu to the Pu^{3+} valency state. Subsequently, 100 µl 1M $NH_2OH \cdot HCl$ and 100 µl 1M $NaNO_2$ were added. Finally 430 µl concentrated nitric acid was added to obtain a Pu^{4+} 8M HNO_3 solution.

This solution was brought on an anion exchange column AG1-X4. After adding of 8M HNO_3, plutonium was eluted with 0.35M HNO_3. This eluted fraction was evaporated, dissolved in 1M HNO_3 to obtain a 1 mg $Pu \cdot ml^{-1}$ solution and transferred to the mass spectrometer for measurement.

6. The mass spectrometer used in the measurements

Isotope abundance ratio measurements were performed by thermal ionization mass spectrometry using a Finnigan MAT 261 mass spectrometer equipped with a 13 locations sample turret and a fixed double collector. The measurements for the determination of the isotopic composition and of the $^{239}Pu/^{244}Pu$ ratios of the blends were done on single collector by mass jumping and using an electron multiplier when necessary (minor isotopes). The measurements of the $^{239}Pu/^{242}Pu$ ratios of the synthetic mixtures for the purpose of calibrating the mass spectrometer, were done in static mode using the double collector.

7. Sample loading

Loading of the filaments was performed in a glove box. One microlitre of the plutonium solution was put on the filament using a commercial transfer pipette. The droplet was dried by passing an electrical current through the filament. This current was generated by a programmable device allowing to do the filament heating in reproducible steps, standardizing temperatures and heating times. After final adjustment of the filaments, the sample turret was transported to the mass spectrometer according to the existing prescriptions of the local health physics services.

8. Data Acquisition

Generation of the ion currents through highly reproducible heating procedure, was controlled by a HP 9000-216 and performed fully automatically. This was always done in the presence of the analyst in order to notice possible erratic occurrences, in which case the measurement was immediately terminated and discarded.

For each filament the isotope abundance ratios were measured over a time period of about 100 min. During this period 4 groups of ratios were measured. Each group represented 7 scans during which the ion currents of all isotopes were measured alternately. One scan took 4 to 15 s depending on the number of different isotopes to be measured (for 5 isotopes: 15 s per scan, 15 s waiting time, 7 scans totalling to about 17.5 min per group).

A computer program calculated directly the isotope abundance ratios at time t as the mean value of these 7 scans. The results of 4 groups per filament were then fitted versus time using a linear least square fit computer program (FITPY). An interpolation at t=70 min provided the isotope abundance ratio used in all calculations.

9. System calibration

The determination of the correction factor for systematic errors, mainly mass fractionation, was done by means of synthetic $^{239}Pu/^{242}Pu$ isotope mixtures, accurate to 0.01 % /1/.

10. The isotopic composition of the plutonium in the spike solution

The isotopic composition of SRM-944/25 was established by measuring isotope abundance ratios on 12-16 filament loadings, according to the method previously described.The ratios $^{238}Pu/^{241}Pu$ and $^{242}Pu/^{241}Pu$ were measured using an electron multiplier, the abundance ratios $^{239}Pu/^{240}Pu$ and $^{241}Pu/^{240}Pu$ were measured on a Faraday collector

abundance ratio	interpolated and K-corrected value (at t = 70 min)
^{238}Pu/^{241}Pu	0.100 10
^{242}Pu/^{241}Pu	0.054 66 32
^{239}Pu/^{240}Pu	44.073 28
^{241}Pu/^{240}Pu	0.012 25 22
^{244}Pu/^{240}Pu	$\leq$0.000 001 1

The observed values, were corrected for mass fractionation and "EM factor" (correction for electron multiplier error). The isotopic composition and its uncertainty was derived using a fully orthodox data reduction and uncertainty propagation. The correction factor for mass fractionation was determined to be $K(^{239}/^{242}Pu)$ = 0.998 82 with an uncertainty of 0.000 96 (2s) on the measurement of K and of 0.000 10 per mass unit, resulting from the uncertainty of the gravimetric preparation of the synthetic mixtures.

The isotopic composition is valid for 10 December 1986 :

	mol %	mass %
^{238}Pu	0.002 72 28	0.002 70 28
^{239}Pu	97.750 70 1 51	97.741 40 1 51
^{240}Pu	2.217 91 1 40	2.226 99 1 40
^{241}Pu	0.027 18 49	0.027 41 49
^{242}Pu	0.001 49 3	0.001 50 3
^{244}Pu	< 0.000 001 1	< 0.000 001 1
Ar(Pu)	239.074 936 17	

11. The isotopic composition of the candidate spike reference material

The isotopic composition of the candidate spike reference material CBNM IRM-042a has been established in a similar way, measuring 12 to 16 filaments. Following values were calculated by fitting of the observed values and interpolating at 70 min :

abundance ratio	interpolated and K-corrected value (at t = 70 min)
^{238}Pu/^{240}Pu	0.008 0 1 0
^{239}Pu/^{244}Pu	0.000 341 9 4 0
^{240}Pu/^{244}Pu	0.006 918 14
^{241}Pu/^{244}Pu	0.000 684 0 4 4
^{242}Pu/^{244}Pu	0.013 611 12

and

	mol %	mass %
^{238}Pu	0.005 41 68	0.005 28 68
^{239}Pu	0.033 47 40	0.032 79 40
^{240}Pu	0.677 16 30	0.666 20 30
^{241}Pu	0.066 95 44	0.066 14 44
^{242}Pu	1.332 33 10	1.321 69 10
^{244}Pu	97.884 68 40	97.907 90 40
Ar(Pu)	244.006 306 85	

12. Certification of CBNM IRM-042a

The concentration expressed in number of atoms ^{244}Pu per kg of solution can be derived from the general equation:

$$\frac{N_X}{N_Y} = \frac{R_Y - R_B}{R_B - R_X} \cdot \frac{\sum Ri_X}{\sum Ri_Y} \quad (1)$$

When N = c · m with

c = concentration of element (Pu) in numbers of atoms per kg of solution

m = mass of solution in kg

then

$$C(Pu,X) = C(Pu,Y) \cdot \frac{m_Y}{m_X} \cdot \frac{R_Y - R_B}{R_B - R_X} \cdot \frac{\sum Ri_X}{\sum Ri_y}$$

Dividing nominator and denominator by R_Y and rearranging, yields

$$C(Pu,X)\cdot\frac{1}{\sum Ri_X}=C(Pu,Y)\cdot\frac{m_Y}{m_X}\cdot\frac{1-R_B/R_Y}{R_B-R_X}$$

We choose R = $^{239}Pu/^{244}Pu$
then $1/\Sigma Ri_X$ = ^{244}Pu abundance in the unknown material X which must be certified (CBNM IRM-042a)
and $R_Y/\Sigma Ri_Y$ = ^{239}Pu abundance in the spike material Y (SRM-944/25)
Considering that $R_Y \rightarrow \infty$, we obtain

$$C(^{244}Pu,X)=C(^{239}Pu,Y)\cdot\frac{m_Y}{m_X}\cdot\frac{1}{R_B-R_X}\qquad(2)$$

Table 2 gives the values of the number of ^{244}Pu atoms per kg of CBNM IRM-042a solution, obtained for each individual determination.

The uncertainty was determined using the recommended procedure of the Bureau International des Poids et Mésures (BIPM) :

$$C(^{244}Pu)\ \pm\ 2s\ =\ C(^{244}Pu)\ \pm\ 2\cdot\ \sqrt{(\delta x/2)^2+s\bar{x}^2}\qquad(3)$$

where

$C(^{244}Pu)$: mean ^{244}Pu concentration of 12 measurements

δx : uncertainty of systematic nature on the ^{244}Pu concentration due to :
- the concentration of SRM-944/25,
- the spiking,
- the measurements of the correction factor K for isotope fractionation,
- the synthetic Pu mixtures used in the calibration,
- the isotope ratio measurements of SRM-944/25, CBNM IRM-042a, blends, and calculated by error propagation of the individual uncertainty contributions of equation (3) (uncertainties of systematic nature)

$s\bar{x}$: standard deviation on the concentration calculated from the 12 measurements (reproducibility).

The ^{244}Pu concentration of CBNM IRM-042a is certified as:

(2.257 7 ± 0.044)·10^{18} atoms $^{244}Pu\cdot kg^{-1}$ of solution

blend	ampoule	^{244}Pu atoms ($x10^{18}$ kg^{-1} of solution)
1	12	2.256 94 2.256 61
2	54	2.256 60 2.257 75
3	100	2.256 99
4	120	2.256 99 2.257 45
5	195	2.256 33 2.256 77
6	244	2.259 67 2.260 73 2.259 21
mean value		2.257 7 2 8

Table 2 - Determination of the 244Pu concentration of CBNM IRM-042a

Quantity	Relative Uncertainties in 10^{-3}	
	of value concerned	of certified end-value
Weighing of base material	0.095	
Certified purity of NBS SRM-944 47.50 %	1.1	
Atomic weight ^{239}Pu 239.052 157 6	0.000 022	
Avogadro Constant 6.022 134 10^{23}	0.002 0	
Abundance ^{239}Pu in NBS SRM-944 97.741 4 mole %	0.015	1.1
Measurement of R_Y ($^{239}Pu/^{244}Pu$)	0.0	
Calculated correction factor	1.0	
m_Y : mass of SRM 944/25 in blend (approx. 2 g)	0.098	1.1
Measurement of R_X ($^{239}Pu/^{244}Pu$)	12	
Calculated correction factor	1.6	
m_X : mass of IRM 042a in blend (approx. 5 g)	0.038	1.1
Measurement of R_B	0.60	
Calculated correction factor	1.6	2.0
Standard deviation on the mean (n = 12)	0.36	
Total relative uncertainty		2.0

Table 3 - Uncertainty Components Pertaining to Equation (3)

13. References

/1/ J. Broothaerts, M. Gallet, P. De Bièvre, "Synthetic 239Pu/242Pu isotope mixtures as basis for absolute Pu isotopic measurements", this Symposium

DEVELOPMENT OF AUTOMATED ANALYTICAL SYSTEM FOR MIXED OXIDE SAMPLE BY X-RAY AND GAMMA-RAY SPECTROMETRIC METHOD

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Abstract

An automated analytical system for mixed oxide sample has been developed to increase analytical capability as well as to improve timeliness of measurement at the conversion and the mixed oxide fuel fabrication plants.

The analytical system can be divided into a sample preparation system, an x-ray fluorescence and a gamma-ray spectrometer, and a computer system for operation control and data processing. The sample preparation system consists of two revolute robots and various kinds of component devices such as a sample and a crucible storage, a sampling device with a balance, an electric furnace and so on. In this system 0.3 g of sample and 7 g of anhydrous sodium tetraborate are taken and fused in a platinum crucible at 1100°C for ten minutes to prepare a glass bead disk. Concentrations of uranium and plutonium are determined by measuring intensity of L series x-ray line of each element. Isotope composition of plutonium in the sample is also determined by passive gamma-ray spectrometry without sample pretreatment.

The results obtained with this system on 0.2 g of uranium gave a precision of 0.2 - 0.5 % (RSD, n=5) and an accuracy of 0.3 - 0.7 % (relative deviation). The automated sample preparation and measurements for one sample with two sub-sampling are accomplished in 1.5 hours.

1. Introduction

Timely evaluation of measurement results as well as accuracy and precision is very important when destructive analysis techniques are used as one of the safeguards measures. In safeguards implementation in Japan , no delays in the evaluation have been observed for uranium bearing samples at present. The criteria of timely verification are, however, still no met for plutonium bearing samples because of the delays of sample pretreatment and shipment. When a large reprocessing and conversion plant comes into operation, it is quite certain that the number of samples to be verified will increase significantly and delays in evaluation would become more serious unless some new proposal is investigated.

In order to overcome these problems, on-site verification measurement possibilities, e.g., construction of an inspectorate laboratory at the site, use of operator's procedure and/or apparatus with appropriate authentication techniques, are now under consideration. The present paper describes the status of the development of an automated analytical system for mixed oxide powder samples, which will be installed at the on-site inspectorate laboratory or the operator's space of the future large reprocessing and conversion plant in Japan.

2. System Description

2.1 Specification

Specifications of the system design are described below.

(Analytical method)
x-ray fluorescence and gamma-ray spectrometry

(MOX sample)

chemical composition	$(Pu,U)O_2$
Pu/U ratio	0.01 - 2
bulk density	~2.2 g/cm^3
sample size :for XRF	~0.3 g
for HRGS	~10 g

(Analytical capability)
600 samples per year (200 working days) except calibration and quality control samples. Number of samples per analytical batch is up to five.

(Operator)
one including system operation and maintenance

(enclosure)
glove box (4.4m x 1.2m x 1.2m)

2.2 Analytical procedure

Figure 1 shows the standard routine operation procedure of this system for analyzing uranium and plutonium concentrations and isotope composition of plutonium in the mixed oxide powder sample.

The system starts its operation from the positioning test of robots and the moving test of component devices for checking whether their movements are normal or not. After that, the robot A picks a sample vessel and transfers it onto the turn table of a bar code reader for identification of the sample. Then the cap of the vessel is removed and two 0.3 g of mixed oxide powder samples are taken and about 7 g of anhydrous sodium tetraborate is added as a flux into a platinum crucible, respectively, by the sampling device including three vibrating spatula on moving tables and a balance.

After sampling process is completed, the robot B picks the crucible and transfers it into an air cooled electric furnace. The crucible containing the sample and the flux is heated at 1100°C for ten minutes to prepare a glass bead sample disk. By slow movement of the crucible table during the heating process, the crucible is agitated continuously to ensure homogeneity and to aid the expulsion of gas bubbles. The crucible and the charge are allowed to cool to room temperature, and are then inverted, then the bead disk drops out. If no crack is found on the bead disk, the robot B holding a disk carrier transfers the bead disk onto the sample turn table of an x-ray fluorescence spectrometer. Concentrations of uranium and plutonium in the sample are determined by standard x-ray fluorescence spectrometric procedure. In parallel with bead disk sample preparation, gamma-ray spectrum of the sample in the vessel is measured for determination of isotope composition of plutonium. After the measurement process is completded, the bead disk carrier discards the bead disk used into a waste vessel, and the cleaning device removes residual mixed oxide powder on spatula and feeder automatically to avoid cross contamination of samples.

2.3 Details of analytical system

(sample preparation system)

The system consists of various component devices shown in the illustration, Fig. 2: two commercially available robots (MITSUBISHI, MOVEMASTER EX model RV-M1), a sample and a crucible storage, a bar code reader, a cap remover, a sampling and flux feeding sub-system, an air cooled electric furnace, a crack detection device and so on. Maintenance is a prime consideration in designing analytical systems used in the enclosure, Consequently, standard commercially available parts are used whenever possible. Some component devices in this system are actuated pneumatically in place of electrically because pneumatic control has a lot of advantages, e.g.,simpler mechanism, easy control and maintenance, less trouble, low cost and so on. The sample storage in which twelve sample vessels can be stored is surrounded by a lead shielding wall of 5 mm in thickness for personnel radiation protection, and the crucible storage can also store ten platinum crucibles. Prior to transfer of the bead disk onto the sample turn table, any crack on the bead disk can be detected by measuring of pressure change in evacuating line connected to the disk carrier. The standard bead disk storage can store four standard bead disks under dry atmosphere for calibration of the x-ray fluorescence spectrometer.

(gamma-ray spectrometric system)

The system includes a planar type high purity germanium detector(ORTEC, model GLP-25300/13-S), a multichannel analyzer (ORTEC, model 7800-8A2), a high voltage power supply(ORTEC, model 659), an amplifier (ORTEC, model 672) and a computer system (HP, model 360C+ with peripherals). The detector with 1 mm thick cadmium and copper filters is inserted into a lead shield mounted under the glove box. The software for automatic data acquisition and analysis, which has been originally developed for field measurement, is written in HP BASIC.

(x-ray spectrometric system)

A multichannel x-ray spectrometer (SHIMADZU, model VXQ-PF2) having three fixed and one scanning channels is used to measure intensity of L series x-ray spectral line of U, Np, Pu and Am after minor modification. That is to say, LiF(220) crystals are equipped in place of LiF(200) because L series spectral lines of actinides are resolved much better by the former than by the latter. And only a sample turn table is installed in the glove box so that maintenance of other parts of the x-ray spectrometer can be done as usual way. To excite L series spectral lines of actinides preferentially, a rhodium target x-ray tube (Varian Eimac, type OEG-76H) is equipped in this spectrometer. During the system operation, the inside of specimen compartment having beryllium window is passed with nitrogen at flow rate of 10 ml/min to protect the window from oxidation.

(control system)

As shown in the block diagram of the control system, Fig. 3, the system includes three desk-top computers (NEC 98RL, 9801UX and HP 360C+) with peripherals, a control module consisting of a programmable sequencer (MITSUBISHI, MELSEC-A series) which controls not only movement of robots and components devices through drive units but also sequence of the operation, a temperature controller for the furnace and so on. Various digital signals from the system components are incorporated into this module, which links to the computer through a PIO interface. Inside and on the module panel, many switches — for selecting manual, semi-automatic or automatic operation, setting sample and flux size, setting the heating period, etc. — are provided. Each function that the system performs (bar code reading, cap removing, sampling and sub-sampling, fusion, etc.) can be controlled independently by means of switches for each operation. In addition, a pendant digital switch box also allows an operator to manipulate the movement of each component device.

3. Preliminary Tests and Results

Results of preliminary tests of the system developed are briefly described bellow.

Because positioning of the robot having five joints and a pair of finger is the most important function for reliability of the system operation, the repeatability of positioning involving simultaneous movement in many degrees of freedom has been tested. All results obtained are within ± 0.3 mm as described in the technical specifications.

For the safety system operation, temperature distribution on the surface of the air cooled electric furnace during the normal operation at 1100°C has been measured, and it is found that temperature on the surface of the furnace is lower than 33°C.

It is well known that the most common fluxes for preparation of specimen in the form of glass bead disk are sodium tetraborate and lithium tetraborate. In addition to these fluxes, sodium polyphosphate, a mixture of sodium carbonate and silicon dioxide have been tested qualitatively for the resistance of disks to cracking, degree of bubble formation, hygroscopic nature of disks during storage, stickiness of the melts to the crucible, melting point and so on. As a result of these test, it is found that anhydrous sodium tetraborate is preferable to other fluxes for preparing bead disks containing uranium.

X-ray fluorescence spectrum of a mixture of strontium, yttrium and zirconium oxides is measured to confirm the resolution of the spectrometer having LiF(220) crystal. As shown in Fig. 4, K series spectral lines of these three elements are well resolved except overlapping of strontium K beta and zirconium K alpha spectral lines, and wavelength difference of each spectral line is approximate 1.8 degree (2θ). By the fact described above, it can be expected that spectral lines of U, Np, Pu and Am could be still resolved each other in spite of smaller wavelength difference,that is, approximate 0.9 degree (2θ) each.

Figure 5 shows two kinds of calibration curves for uranium in the bead disk. Standard bead disks were prepared as follows; after mixing a known amount of uranium oxide with 7 g of anhydrous sodium tetraborate in the platinum crucible manually, the mixture was fused at 1100 °C for 10 min. Then, the intensity of uranium L series line excited with Rh x-ray tube operating at 40 kV and 50 mA was measured five times for 50 seconds. The upper calibration curve in Fig. 5 was prepared by polynomial fitting of uranium concentraion and line intensity. The curve showed the typical one of higher atomic number analyte in low atomic number matrix. The caribration curve need not be linear to be of analytical value, although certainly a linear curve is preferable. As the lower curve shown in Fig. 5, the linear calibration curve can be obtained by adding high-absorption diluent (e.g. lanthanum oxide) to the flux if necessary.

Analytical results of uranium in synthetic sample bead disks prepared by the above procedure gave a precision of 0.2 - 0.5 % (RSD, n=5) and an accuracy of 0.3 - 0.7 % (relative deviation). In addition, there is no significant difference between analytical results obtained by two calibration curves in Fig. 5. During the tests it was also found that correction of analytical results for weight loss in the fusion process could improve the accuracy of analytical results.

After the measurements, the disks were tested by placing them in an x-ray diffractometer and observing their Debye-Scherrer diffraction patterns. The appearance of no diffraction peaks indicates the absence of residual crystalline material in the bead disk.

4. Discussion

Dissolution of refractory plutonium dioxide is a major problem in the analytical chemistry of plutonium bearing samples. In this case, the most common method used in the past is the use of nitric acid containing a small concentration of fluoride. However, this technique is not adequate to automation because it is almost impossible to detect automatically the time of completion of dissolution process by some sensor. So, our laboratory decided to develop the automated sample preparation system based on the flux fusion technique which provide a homogeneous glass bead disk suitable for direct use as a specimen for x-ray spectromery. The fabrication of the whole system has been completed on the end of last year. As the second step, performance tests of the whole system including the sample preparation and x-ray spectrometry using iron oxide powder sample are now under way to confirm the reliability of system operation. In order to improve the accuracy of concentration determination, a minor modification of the sample prepration process is in progress as follows ; addition of weight loss measurement operation prior to the x-ray spectrometry, and correction of analytical results by using obtained data.

5. Acknowledgments

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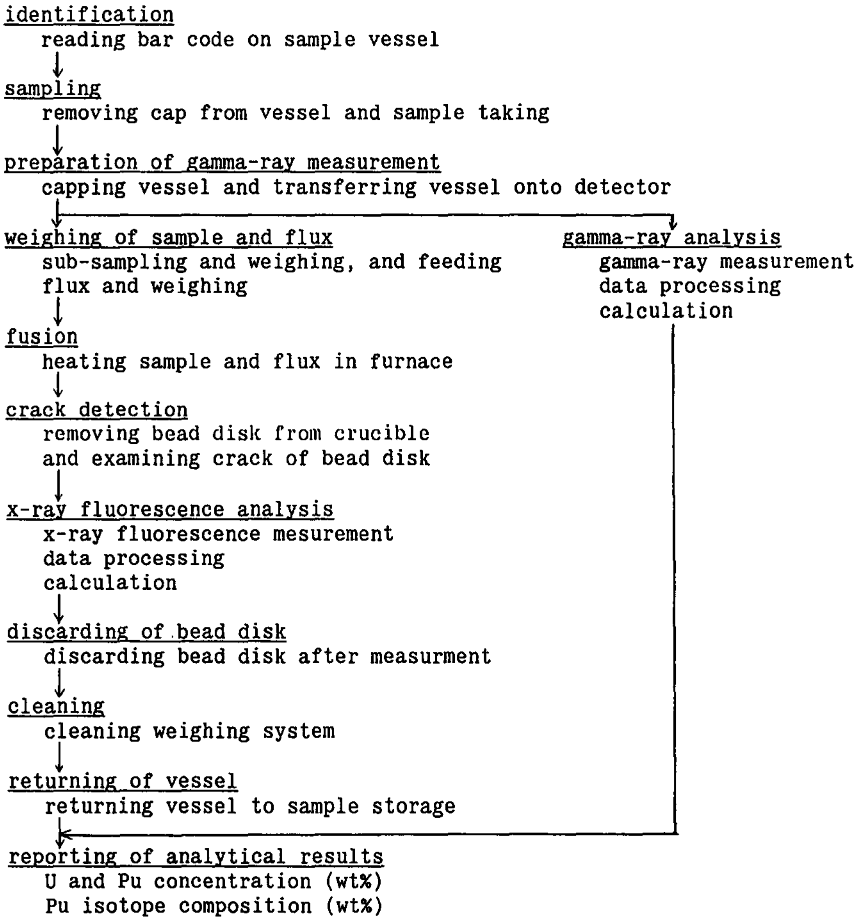

Fig. 1 Analytical Procedure

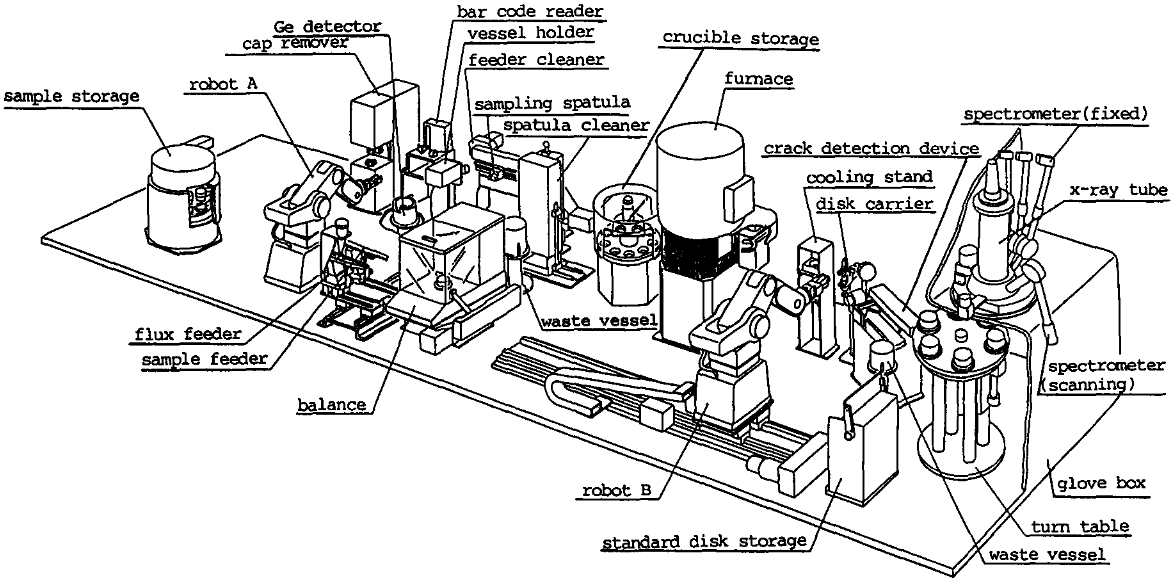

Fig. 2 Automated Analytical System for Mixed Oxide Sample

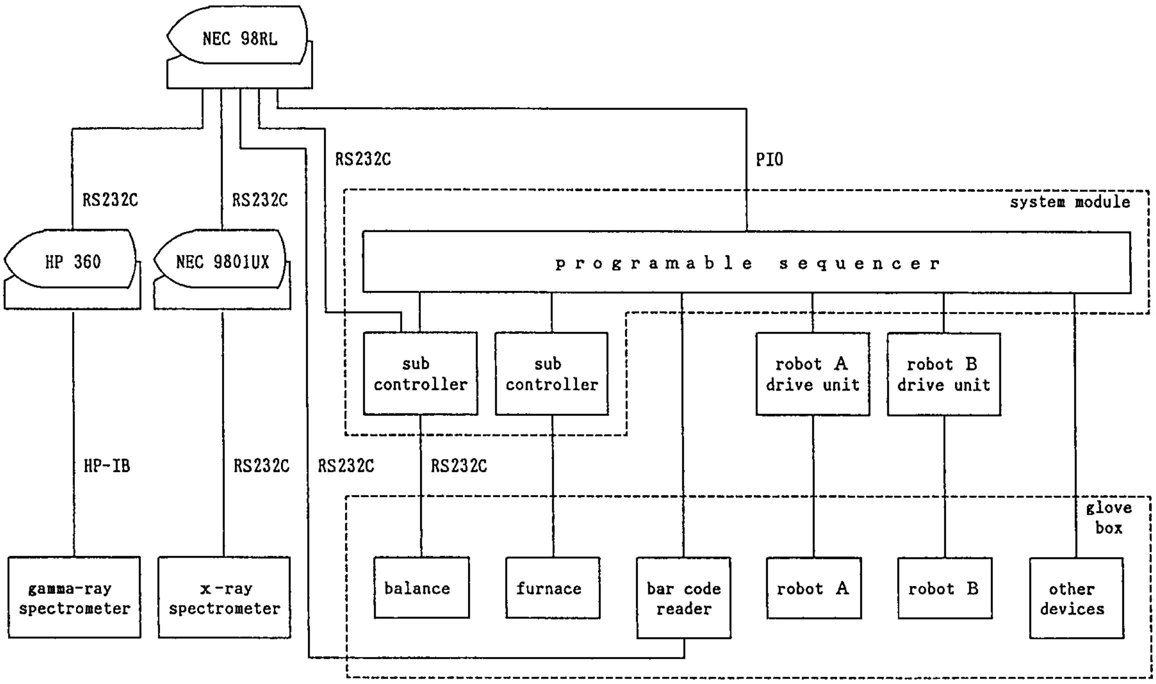

Fig. 3 Diagram of Control System

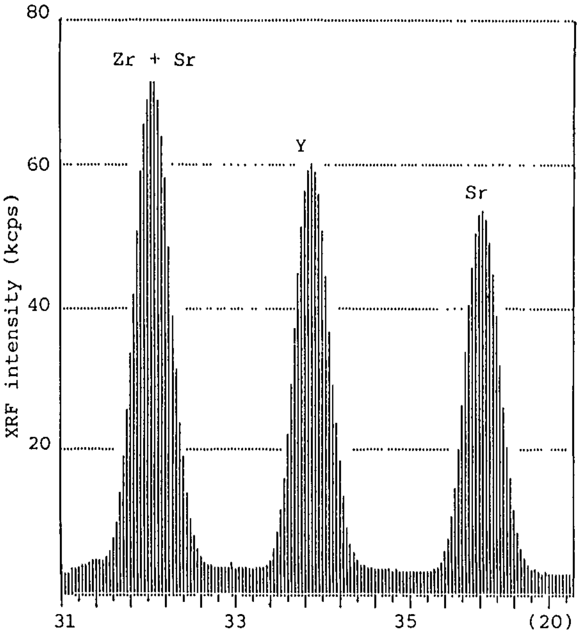

Fig. 4 XRF Spectrum of Sr, Y and Zr

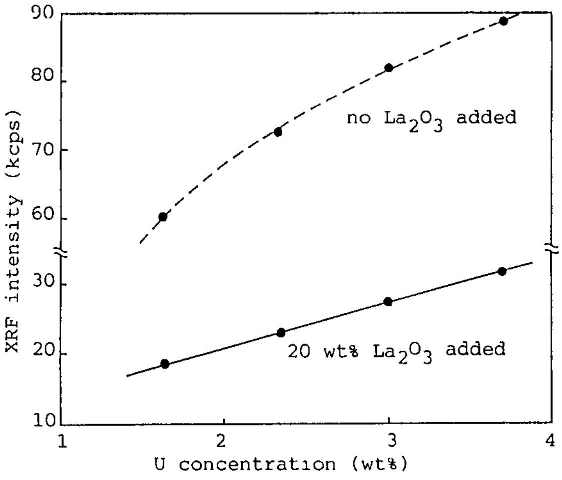

Fig. 5 Calibration Curve of U in Bead Disk

FINAL RESULTS OF THE PIDIE INTERCOMPARISON EXERCISE FOR THE PLUTONIUM ISOTOPIC DETERMINATION BY GAMMA-RAY SPECTROMETRY

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ABSTRACT

Final results from the PIDIE intercomparison exercise organised by the ESARDA Working Group on Techniques and Standards for Non-Destructive Analysis are presented.

The aim of this exercise carried out in 1988 was to test the gamma-ray spectrometry methods used to determine the plutonium isotopic ratios in a large range of isotopic composition, in order to analyse the parameters and the error sources influencing the results.

Sets of seven sealed samples of different plutonium isotopic composition were sent to nine participating laboratories. The final results with uncertainty indicators are reported; they are compared with complementary mass-spectrometry determinations.

No important bias has been observed from this exercise. Significant improvements in plutonium isotopic determination by gamma-ray spectrometry come from both more elaborate spectrum analysis methods and better equipment.

1. INTRODUCTION

In several meetings, the ESARDA Working Group on Techniques and Standards for Non-Destructive Analysis has expressed the need to evaluate the different methods used for the determination of the isotopic composition of plutonium. After an earlier interlaboratory exercise based on measurements of plutonium Certified Reference Materials (CRM's) and completed in 1981 [1], it was decided to organize the international comparison called PIDIE, i.e., Plutonium Isotopic Determination Intercomparison Exercise.

The aim of the PIDIE exercise was to test the X and gamma-ray methods used for measuring plutonium over a wide range of isotopic composition, to give an opportunity to improve them, to investigate sources of error and, if possible, to improve the knowledge of emission probabilities. As a general rule, these methods are based on the X- and/or gamma-ray radiation emitted by the isotopes ^{238}Pu, ^{239}Pu, ^{240}Pu, ^{241}Pu and ^{241}Am and they offer the advantage that the use of external standards is not essential. However, an additional aim was to examine the possible improvement of such measurements by using CBNM pilot reference samples.

Because the PIDIE exercise started about 10 years ago, the aims described above have somewhat changed since then and the questionnaire reflects the problems faced at that time. For example, no part of the exercise was devoted to measurements at high counting rates, low-quantity measurements or other experiments testing screen effects. Nevertheless, due to the strict anonymity and secrecy maintained, the PIDIE exercise can be considered as a real blind test to confirm the present possibilities of X/gamma-ray spectrometry for isotope abundance measurements.

The participants received the sets of plutonium samples and carried out their measurements in 1988. After a synthesis of the first results provided by eight participants, a preliminary report was written in 1989 [2]. Shortly after, a ninth participant sent his results before the deadline. Mass-spectrometry measurements of small samples of each plutonium batch were carried out in 1989 and 1990 by six other laboratories to complete the final analysis of the X/gamma-ray spectrometry results.

The present paper summarizes the final X and gamma-ray spectrometry results [3]. As some of the participants gave several sets of results using different methods for the determination of the plutonium isotopic composition, a total of sixteen sets are available for this final evaluation.

2. GENERAL CONSIDERATIONS

2.1 PIDIE plutonium samples

UKAEA-AERE Harwell was charged with realizing identical sets of seven plutonium samples. The containers were designed about ten years ago and the preparation of the samples began not long after, but the final dispatch was delayed due to obstacles encountered in the export of nuclear materials. Each sample was in the form of a sealed capsule containing 0.45 gram of plutonium oxide in pellet form with the following isotopic abundances:

Batch number	Isotope ratios (%)			
	^{238}Pu/P	^{239}Pu/P	^{240}Pu/P	^{241}Pu/P
1	0.01	94	6	0.2
2	0.02	89	10	0.4
3	0.05	85	14	1.0
4	0.1	78	20	1.8
5	0.1	76	21	2.0
6	1.0	69	25	5.4
7	1.3	65	27	6.8

2.2 Participation

Nine laboratories participated in the PIDIE exercise. An arbitrary code number was allocated to each participant: a subscript - 1, 2 or 3 was added to the code number of the laboratories which sent in several questionnaires. The list of these laboratories is as follows:

ANMCO HARWELL (United Kingdom)
CEC/CBNM GEEL and SCK/CEN MOL (Belgium)
CEC/JRC ISPRA (Italy)
ECN/PETTEN (The Netherlands)
ENEA CASACCIA (Italy)
IAEA (Vienna)
LANL (U.S.A.)
LLNL (U.S.A.)
DAMRI/LMRI (France)

3. ISOTOPIC COMPOSITION DETERMINATION

3.1 General principles

Except for ^{242}Pu, all the isotopes of plutonium emit gamma rays; for this reason, the general procedure used for determining the plutonium isotopic composition is based on the measurement of the X and gamma radiations emitted by these isotopes and, also, by the two other decay products, ^{237}U and ^{241}Am. Combining the information from different peaks in the gamma spectrum with their specific X- or gamma-ray emission probabilities and the half-lives, the atom ratios can be determined after a relative efficiency calibration without the use of external standards.

Two types of methods were used in the PIDIE exercise. The first and most recent type [4] exploits the X and gamma-rays in the 94 to 104 keV region, with some gamma-rays from other regions; the efficiency calibration has to take account of the discontinuity at the K-absorption edge at 121 keV. The second type is related to other methods [5 to 10], where the data processing mainly uses several peaks in the 120 to 500 keV region. One of the main sources of uncertainty is the determination of the ^{240}Pu abundance by its sole gamma-ray at 160 keV, which is not very intense and is perturbed by the interferences.

A letter was assigned to each method to characterize each set of final results. The letter A is for the methods of the first type, the letters B, C and D for the methods of the second type, B when UCRL-52139 [11] or UCRL-53506 [5] photon emission probabilities were used, C when CBNM pilot reference samples [8] were used to get corrected parameters and D when other nuclear data were used [12,13]. Several participants used the same analysis code with similar nuclear data and some participants used the same analysis code several times, but with different nuclear data.

The half-lives used by participants and taken from literature [11 to 13] present some slight differences but the consequences on measurements have been considered as insignificant.

3.2 Instrumentation and measurement conditions

A precise determination of the isotopic composition of plutonium samples is strongly dependent on good energy resolution and, at high counting rates, on good charge collection. The error made in plutonium parameters can be doubled when the resolution at 208 keV goes from 0.7 to 1.0 keV. It is generally recommended to have the experimental ratio FWTM (Full Width at Tenth Maximum)/FWHM (Full Width at Half Maximum) lower than 1.9, as close as possible to the theoretical value of 1.82 obtained for a Gaussian peak shape. Now, the FWFM data (Full Width at Fiftieth Maximum) seems more appropriate to estimate the detector tailing.

One participant used a coaxial n-type germanium detector, the others used small-sized planar intrinsic germanium detectors, 1.4 to 10 cm^3 in volume, which had an energy resolution better than 600 eV at 125 keV and 700 eV at 208 keV at low counting rates. Because of the small quantity of plutonium in the samples, it was impossible to test the instrumentation at high counting rates, the highest value was about 9000 counts per second. It would be interesting to test this factor in a future exercise.

Most participants used a cadmium absorber, 1 to 2 millimeters thick, and sometimes, a cooper absorber in addition. Generally, the samples were measured close to the detector window, with a source-to-detector distance below 4 centimeters. Under these conditions, the counting rates were low, a few kilo-counts per second.

4. EVALUATION RULES

4.1 Compilation of individual results

For participant k giving an i^{th} partial result designated $x_{i,j,k}$ for the j^{th} component, and a corresponding uncertainty designated $\Delta x_{i,j,k}$, the final result adopted is given by the weighted mean $X_{j,k}$ estimated as follows:

$$X_{j,k} = \frac{\sum_{i=1}^{m} \frac{x_{i,j,k}}{(\Delta x_{i,j,k})^2}}{\sum_{i=1}^{m} \frac{1}{(\Delta x_{i,j,k})^2}}$$

[m = number of experiments]

Because the number of experiments was limited (3) and the participants evaluated their uncertainties following different rules, the attribution of comparable final uncertainties was not obvious. Provided that systematic components were considered predominant in most cases, it was considered that repeated measurements under identical conditions could not improve the uncertainties significantly. Then, the uncertainty $\Delta x_{j,k}$ in the final result was taken as the maximum value of the mean of the uncertainties in the partial results quoted by the laboratories, and the standard deviation of the weighted mean.

4.2 Final results

Taking account of the results of each participant, it was possible to calculate the mean values and various uncertainties for the j^{th} component defined as follows:

- unweighted mean:

$$\bar{X}_j = \sum_{k=1}^{n} \frac{X_{j,k}}{n} \quad \text{(n = number of results)}$$

- relative standard deviation of a result (from the unweighted mean)

$$S_{X_j} = \frac{1}{\bar{X}_j}\left[\sum_{k=1}^{n}\frac{(X_{j,k} - \bar{X}_j)^2}{(n-1)}\right]^{\frac{1}{2}}$$

- relative standard deviation of the mean (from the unweighted mean)

$$S_{\bar{X}_j} = \frac{S_{X_j}}{\sqrt{n}}$$

- average of relative uncertainties

$$\Delta X_j = \sum_{k=1}^{n}\frac{\Delta X_{j,k}}{n}$$

where $\Delta X_{j,k}$ is the uncertainty in the result $X_{j,k}$

- weighted mean:

$$\bar{X}_{\pi j} = \frac{\sum_{k=1}^{n}\frac{X_{j,k}}{(\Delta X_{j,k})^2}}{\sum_{k=1}^{n}\frac{1}{(\Delta X_{j,k})^2}}$$

- relative standard deviation of the weighted mean (also called the external error)

$$S_{\bar{X}\pi j} = \frac{1}{(n-1)} \cdot \left[\frac{\sum_{k=1}^{n}\frac{(\bar{X}_{\pi j} - X_{j,k})^2}{(\Delta X_{j,k})^2}}{\sum_{k=1}^{n}\frac{1}{(\Delta X_{j,k})^2}}\right]^{\frac{1}{2}}$$

5. RESULTS FROM THE PIDIE EXERCISE

5.1 ^{238}Pu to ^{241}Pu and ^{241}Am results by X and gamma-ray spectrometry

a) Comparison of final results

The following parameters are given in Table 1 and Figure 1:

- unweighted means with the corresponding relative standard deviations of a single result and of the mean for all D.A. and N.D.A. results, and results from the X/gamma-ray spectrometry methods A,B,C and D. Because the number of results available for methods C and D is limited, i.e., 2, the relative standard deviation is not significant and is not, therefore, given.
- weighted means for all N.D.A. results with, in the Table, the corresponding relative standard deviations of the mean (external errors).
- average of the relative uncertainties for all N.D.A. results and for methods A,B,C and separately.

The dispersion obtained in all the X/gamma-ray spectrometry results versus atom ratios were about: 1 to 4% for ^{238}Pu/P, 0.2 to 1.4% for ^{239}Pu/P, 2 to 3.5% for ^{240}Pu/P, 1 to 2% for ^{241}Pu/P, 3 to 5% for ^{241}Am/P.

b) Comments

Several observations can be made with respect to the above results:

- The PIDIE exercise is based on aims which were defined about ten years ago, therefore it is not designed to take account of recent technologies at best. New analysis techniques which are tested are utilized by a number of participants which is too limited for studying a possible specific bias.
- Many parameters, such as measuring conditions, equipment, nuclear data, range of energies, softwares, differ from one participant to another, so that conclusions relate to general points only. Nowadays, it could be possible to obtain more specific conclusions, by limiting the number of influencing parameters and taking account of recent developments.
- In general, the relative standard deviation for a single result is of the same order of magnitude as the average of the relative uncertainties, which means, on average, that the latter have been correctly estimated.
- Except for ^{241}Am where the bias is analyzed below (Section 5.3), the N.D.A. results are, overall, in agreement with the D.A. results. The latter are, nevertheless, characterized by a very small scatter of the ^{239}Pu/Pu, ^{240}Pu/Pu and ^{241}Pu/Pu ratios.
- With respect to the determination of ^{240}Pu, which turns out to be rather difficult by gamma-ray spectrometry, the results are in agreement with the D.A. values. Nevertheless, it can be shown after a careful analysis that the results obtained with the type B methods, are, on average, lower than the D.A. values, while the results obtained with type A methods are slightly higher. In contrast the results of the determination of ^{239}Pu from the type B methods are somewhat higher than the D.A. values.
- With respect to the other methods based on gamma-ray spectrometry, significant progress can be seen with the type A methods. The results obtained, in agreement with the D.A. values are characterized by uncertainty levels, relative standard deviation of a single result and average of the relative uncertainties, definitely lower than those obtained with the other methods. In addition, for the determination of ^{238}Pu, ^{241}Am and occasionally, for the determination of ^{241}Pu these type A methods appear to be more precise than the destructive methods.

c) Results from X/gamma-ray spectrometry using an additional absorber

The participants were asked to measure samples N°2 and N°6 with an additional absorber, thus, reducing the efficiency for low-energy peaks. This part of the exercise shows that the analysis of Pu spectra in the KX-ray energy region, which provides significant improvements, appears to be limited in the presence of screens. Then, usual methods turn out to be an indispensable complement in some applications, such as the measurement of nuclear wastes or hold-up. Other experiments relating to screen thicknesses and low quantities measurements, seem worthwhile.

d) Corrections based on the use of the CBNM pilot references samples

TABLE 1 - FINAL RESULTS FOR PIDIE EXERCISE GIVEN AS ATOMIC RATIOS ^{238}Pu/P, ^{239}Pu/P, ^{240}Pu/P, ^{241}Pu/P, ^{241}Am/P, WHERE P IS THE SUM OF ATOMS OF ^{238}Pu, ^{239}Pu, ^{240}Pu, ^{241}Pu.

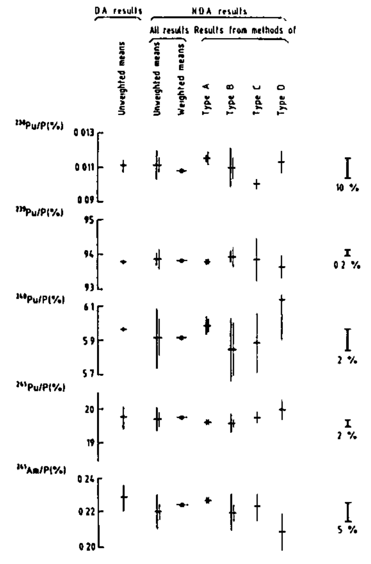
n = number of results

$\bar{X}$ = mean value of results, S_X = relative standard deviation on a single result, $S_{\bar{X}}$ = relative standard deviation on mean

Δ = average of the relative uncertainties on results

The letter "A" is attributed to the methods using mainly the X-ray region; for the methods using the 120 to 500 keV region, the letter "B" is given when the UCRL-52135 or 53506 nuclear data were used, "C" when a correction derived from the CBNM Pilot samples was applied, "D" when other photon emission probabilities were used.

Sample ratio Pu/P	Mass-spectrometry results (n = 7)			Final results from X/γ-ray spectrometry																		
				For all results, unweighted means (n = 16)				For all results, weighted means (n = 16)			For type A methods (n = 3, unweighted means)				For type B methods (n = 9, unweighted means)				For type C methods (n = 2, unweighted means)		For type D methods (n = 2, unweighted means)	
	$\bar{X}$	S_X(%)	$S_{\bar{X}}$(%)	$\bar{X}$	S_X(%)	$S_{\bar{X}}$(%)	Δ_X(%)	$\bar{X}_\pi$	$S_{\bar{X}_\pi}$(%)	Δ_X(%)	$\bar{X}$	S_X(%)	$S_{\bar{X}}$(%)	Δ_X(%)	$\bar{X}$	S_X(%)	$S_{\bar{X}}$(%)	Δ_X(%)	$\bar{X}$	Δ_X(%)	$\bar{X}$	Δ_X(%)
Sample 1																						
^{238}Pu/P	0.0111	3.8	1.5	0.0111	8.5	2.1	5.5	0.0108	1.4	5.5	0.0116	0.99	0.57	3.6	0.0110	10.3	3.4	6.2	0.0101	3.5	0.0113	7.1
^{239}Pu/P	93.822	0.0089	0.0034	93.873	0.21	0.050	0.34	93.836	0.022	0.34	93.801	0.070	0.040	0.070	93.941	0.19	0.063	0.35	93.900	0.65	93.645	0.40
^{240}Pu/P	5.969	0.11	0.043	5.919	3.4	0.84	2.4	5.917	0.025	2.4	5.993	1.0	0.60	0.75	5.850	3.1	1.0	2.8	5.895	2.0	6.141	3.1
^{241}Pu/P	0.1977	1.7	0.64	0.1971	1.7	0.43	0.8	0.1975	0.22	0.81	0.1966	0.13	0.075	0.52	0.1963	1.9	0.62	0.80	0.1976	0.80	0.2006	1.3
^{241}Am/P	0.2287	3.2	1.3	0.2204	4.4	1.1	2.2	0.2237	0.24	2.2	0.2269	1.2	0.69	0.52	0.2202	4.8	1.6	2.1	0.2231	3.4	0.2086	4.0
Sample 2																						
^{238}Pu/P	0.0227	3.1	1.2	0.0232	3.8	0.96	4.4	0.0225	0.68	4.4	0.0229	3.4	2.0	2.1	0.0230	4.1	1.4	4.9	0.02325	3.3	0.0242	6.8
^{239}Pu/P	89.463	0.010	0.0039	89.552	0.26	0.067	0.40	89.431	0.017	0.40	89.456	0.070	0.040	0.26	89.577	0.31	0.10	0.37	89.625	0.59	89.510	0.54
^{240}Pu/P	10.071	0.088	0.033	9.980	2.4	0.59	1.7	10.051	0.34	1.7	10.075	0.64	0.37	0.53	9.958	2.7	0.92	1.7	9.910	1.9	10.010	3.2
^{241}Pu/P	0.4428	0.52	0.20	0.4431	1.5	0.38	0.87	0.4413	0.22	0.87	0.4437	0.65	0.38	0.40	0.4416	1.2	0.40	0.92	0.4410	0.75	0.4514	1.4
^{241}Am/P	0.2633	1.2	0.49	0.2522	5.3	1.3	2.3	0.2567	0.25	2.3	0.2615	2.2	1.3	0.44	0.2519	6.0	2.0	2.2	0.2514	3.4	0.2403	4.2
Sample 3																						
^{238}Pu/P	0.0475	1.0	0.40	0.0477	2.4	0.60	3.4	0.0473	0.55	3.4	0.0481	1.0	0.59	1.8	0.0472	2.6	0.85	3.5	0.0480	3.2	0.0489	5.5
^{239}Pu/P	84.835	0.011	0.0041	84.814	0.42	0.10	0.40	84.784	0.031	0.40	84.786	0.040	0.023	0.14	84.863	0.54	0.18	0.42	84.705	0.50	84.745	0.59
^{240}Pu/P	14.128	0.049	0.018	14.148	2.5	0.63	2.0	14.153	0.23	2.0	14.170	0.26	0.15	0.52	14.103	3.3	1.1	2.3	14.265	1.9	14.200	2.7
^{241}Pu/P	0.9900	0.48	0.13	0.9883	0.86	0.22	1.0	0.9835	0.27	1.0	0.9902	0.55	0.32	0.43	0.9855	0.98	0.33	1.2	0.9900	0.70	0.9965	1.4
^{241}Am/P	0.6249	2.9	1.2	0.6062	3.2	0.79	2.2	0.6130	0.21	2.2	0.6170	0.71	0.41	0.48	0.6052	3.7	1.2	2.0	0.6015	4.0	0.5990	3.8
Sample 4																						
^{238}Pu/P	0.1096	1.2	0.45	0.1091	1.4	0.34	2.5	0.1092	0.41	2.5	0.1108	1.0	0.59	1.2	0.1083	1.2	0.39	2.3	0.1091	3.1	0.1102	4.5
^{239}Pu/P	78.237	0.017	0.0065	78.289	0.51	0.13	0.56	78.256	0.089	0.56	78.217	0.20	0.12	0.34	78.372	0.63	0.21	0.48	78.015	0.62	78.295	0.65
^{240}Pu/P	19.816	0.048	0.018	19.771	2.0	0.50	1.7	19.889	0.29	1.7	19.840	0.71	0.41	0.52	19.694	2.5	0.84	1.8	20.050	2.1	19.735	2.2
^{241}Pu/P	1.837	0.29	0.11	1.831	1.2	0.31	0.90	1.831	0.32	0.90	1.835	0.68	0.39	0.42	1.826	1.3	0.42	0.93	1.828	0.80	1.851	1.5
^{241}Am/P	1.573	2.0	0.83	1.560	3.4	0.85	1.8	1.575	0.25	1.8	1.567	0.91	0.52	0.41	1.561	3.6	1.2	1.5	1.559	3.5	1.548	3.7
Sample 5																						
^{238}Pu/P	0.1330	1.1	0.42	0.1321	1.6	0.40	2.3	0.1319	0.43	2.3	0.1338	0.97	0.56	1.2	0.1314	1.7	0.57	2.1	0.1310	3.1	0.1336	4.3
^{239}Pu/P	76.504	0.025	0.0096	76.457	0.67	0.17	0.50	76.397	0.084	0.50	76.429	0.14	0.081	0.16	76.578	0.79	0.26	0.56	76.045	0.55	76.370	0.70
^{240}Pu/P	21.300	0.070	0.027	21.349	2.4	0.60	1.7	21.353	0.33	1.7	21.366	0.45	0.26	0.53	21.235	2.8	0.94	1.9	21.775	2.0	21.410	2.2
^{241}Pu/P	2.063	0.27	0.10	2.061	1.3	0.32	0.91	2.064	0.27	0.91	2.069	0.24	0.14	0.39	2.057	1.5	0.49	0.96	2.048	0.82	2.082	1.5
^{241}Am/P	1.748	2.7	1.1	1.735	3.3	0.84	1.9	1.751	0.22	1.9	1.751	0.49	0.28	0.43	1.732	3.5	1.2	1.6	1.725	3.4	1.732	3.7
Sample 6																						
^{238}Pu/P	0.9695	0.65	0.25	0.9661	1.7	0.42	2.5	0.9718	0.21	2.5	0.9747	0.21	0.12	0.64	0.9598	2.0	0.67	2.4	0.9690	3.3	0.9790	5.1
^{239}Pu/P	68.890	0.016	0.0062	69.070	1.0	0.26	1.1	68.813	0.13	1.1	68.684	0.10	0.058	0.32	69.249	1.2	0.40	1.1	69.210	1.3	68.705	1.6
^{240}Pu/P	24.703	0.048	0.018	24.561	2.9	0.73	2.8	24.816	0.34	2.8	24.906	0.23	0.13	0.69	24.420	3.6	1.2	3.1	24.420	2.8	24.815	4.5
^{241}Pu/P	5.438	0.29	0.11	5.404	2.1	0.53	1.5	5.415	0.23	1.5	5.439	0.33	0.19	0.53	5.372	2.5	0.82	1.7	5.400	1.2	5.498	2.6
^{241}Am/P	3.775	4.1	1.7	3.860	3.3	0.82	2.5	3.872	0.14	2.5	3.877	0.19	0.11	0.59	3.862	3.2	1.1	2.4	3.846	3.8	3.833	4.4
Sample 7																						
^{238}Pu/P	1.322	1.6	0.62	1.335	2.0	0.50	2.6	1.344	0.33	2.6	1.342	0.82	0.47	0.70	1.328	2.5	0.82	2.5	1.338	3.4	1.352	5.3
^{239}Pu/P	65.132	0.086	0.032	65.428	1.4	0.34	1.3	64.956	0.32	1.3	64.960	0.60	0.35	0.62	65.545	1.6	0.53	1.3	65.720	1.5	65.310	1.8
^{240}Pu/P	26.781	0.094	0.036	26.496	3.5	0.87	3.2	26.889	0.64	3.2	26.915	1.3	0.75	0.74	26.424	4.1	1.4	3.5	26.195	3.9	26.490	4.5
^{241}Pu/P	6.765	0.25	0.097	6.739	2.0	0.50	1.6	6.735	0.47	1.6	6.786	0.50	0.28	0.63	6.699	2.4	0.79	1.7	6.745	1.3	6.847	2.8
^{241}Am/P	3.677	3.6	1.5	3.709	3.1	0.78	2.7	3.746	0.24	2.7	3.742	0.73	0.42	0.67	3.717	3.1	1.0	2.6	3.666	4.1	3.666	4.6



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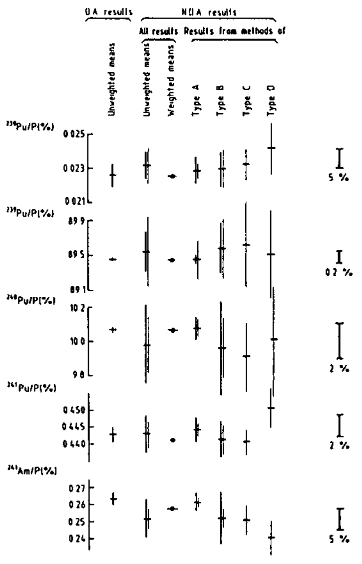

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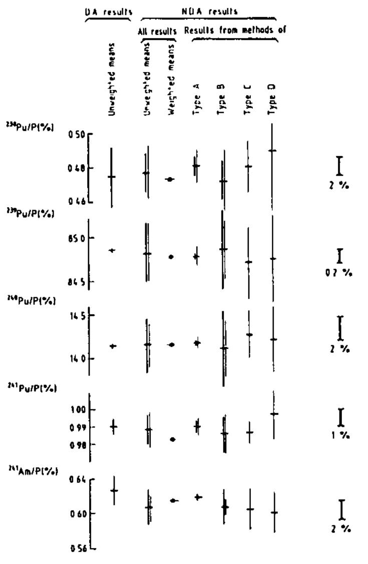

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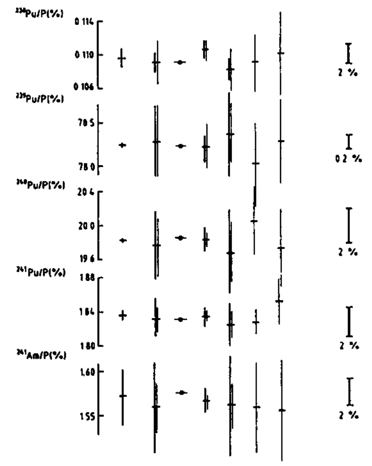

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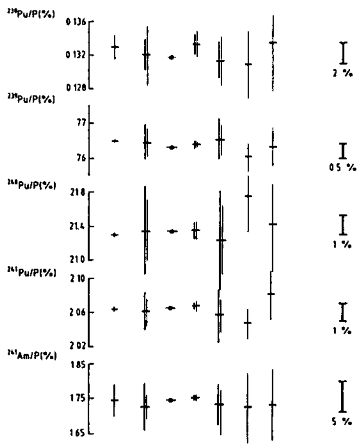

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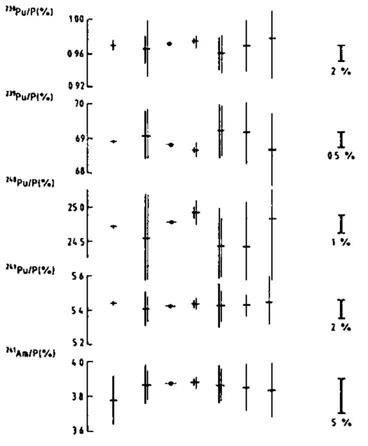

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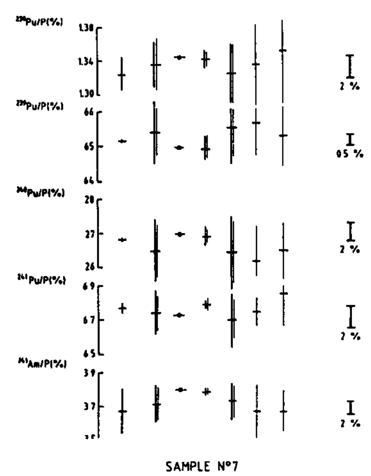


SAMPLE N°7

LEGEND :
P : sum of abundances of ^{238}Pu to ^{241}Pu
——: relative standard deviation for a single result
——: average of the relative uncertainties on result
D.A results : results from destructive analysis techniques
N.D.A. results : results from non-destructive analysis techniques (X/γ-ray spectrometry)
Type A : methods using mainly the X-ray region
Type B : 120 - 500 keV region analysis procedures with UCRL data
Type C : 120 - 500 keV region analysis procedures after correction with CBNM reference samples
Type D : 120 - 500 keV region analysis procedures using other data

FIGURE 1 : FINAL RESULTS FOR PIDIE EXERCISE

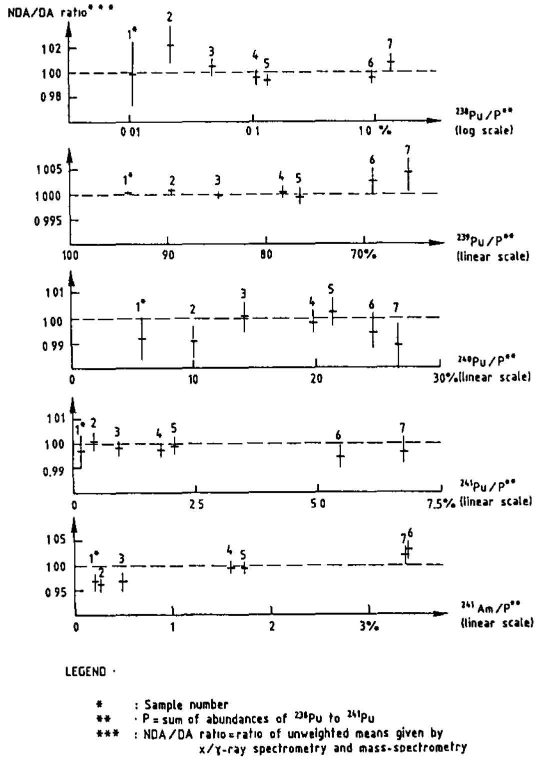
As too few participants used the CBNM pilot reference samples, it is not possible to draw general conclusions. However, it can be seen that the differences between the N.D.A. results and the D.A. results were reduced by a factor of 3 for ^{239}Pu, ^{240}Pu and ^{241}Pu.

5.2 Results from destructive analyses (D.A. results)

In order to compare the X/gamma-ray spectrometry results with other values obtained differently, several PIDIE samples were distributed to a number of laboratories for destructive analysis - D.A.-Mass spectrometry was used to determine the abundances of all the plutonium isotopes except ^{238}Pu, and alpha counting or another technique was used for the measurement of ^{239}Pu and ^{241}Am abundances. The results of seven sets of PIDIE destructive analyses collected recently [15] are also reported in Table 1 and compared with the N.D.A. results in Figure 2. The ratios of the average N.D.A. values to average D.A. values are thus represented with standard deviations resulting from the quadratic summing of the primary standard deviations of the average values.

5.3 Remark for the determination of ^{241}Am abundance

A significant bias can be seen from Figure 2 for the determination of ^{241}Am; at low concentrations, the values determined by X/gamma-ray spectrometry are lower than those obtained from destructive analyses, on the other hand, the converse holds at high concentrations. The Figure 3 shows that this bias is also observed with the type A methods; in addition, it is similar to the bias observed between the results obtained by mass spectrometry using the double spike technique and those obtained by alpha-counting. This observation implies that there is probably a systematic error in the alpha-counting.



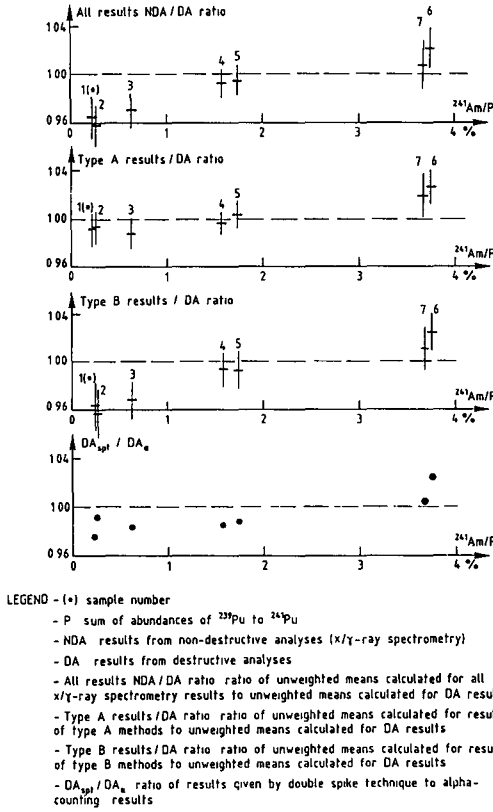
LEGEND -

* : Sample number

** · P = sum of abundances of ^{238}Pu to ^{241}Pu

*** : NDA/DA ratio=ratio of unweighted means given by x/γ-ray spectrometry and mass-spectrometry

FIGURE 2 COMPARISON OF N D A and D A RESULTS



LEGEND - (•) sample number

- P sum of abundances of ^{239}Pu to ^{241}Pu
- NDA results from non-destructive analyses (x/γ-ray spectrometry)
- DA results from destructive analyses
- All results NDA/DA ratio ratio of unweighted means calculated for all x/γ-ray spectrometry results to unweighted means calculated for DA results
- Type A results/DA ratio ratio of unweighted means calculated for results of type A methods to unweighted means calculated for DA results
- Type B results/DA ratio ratio of unweighted means calculated for results of type B methods to unweighted means calculated for DA results
- DA_{spl} / DA_{α} ratio of results given by double spike technique to alpha-counting results

FIGURE 3 BIAS OF ^{241}Am RESULTS . COMPARISON OF X/γ-RAY SPECTROMETRY RESULTS WITH D A RESULTS

5.4 Measurements of ^{242}Pu by isotopic correlations

^{242}Pu cannot be measured by X/gamma-ray spectrometry. The questionnaire included an optional question for participants willing to give a result for ^{242}Pu using isotopic correlations. Eight results from five laboratories were received. Rather large dispersion can be observed (around 20% relative standard deviation) for the results. This may be explained by the different ways of dealing with the ^{241}Am/^{241}Pu ratio correction. A significant bias relative to the destructive analysis results may be noted for low abundances (around 100% bias below 0.2% ^{242}Pu abundance) that could be partially explained by the unknown history and mixed origins of the samples. But this could representative of future actual materials, such as re-cycled fuels. A possible conclusion of this exercise (which was not the main purpose of PIDIE) is the need to study new isotopic correlation formulas.

5.5 Remark

Due to large number of factors influencing the measurement, X/gamma-ray spectrometry equipment, different experimental conditions, methods, and nuclear data, PIDIE cannot be expected to yield general conclusions. By fixing some factors: same equipment, same measurement conditions, same samples, the PERLA exercise, which was initiated more recently [16], is turning out to be a valuable follow-up since it makes it possible to validate, in principle, the methods and to analyze their specific biases.

6. CONCLUSION

From this comparison, it is evident that there is no significant bias between the values given by X/gamma-ray spectrometry and the values provided by destructive analyses with the exceptions of ^{242}Pu and ^{241}Am.

Appreciable progress in both precision and accuracy have been recorded, due to a combination of better equipment and improved procedures, as for example, the method based on the analysis of the plutonium X/gamma-ray spectrum in the low-energy region.

Characterized by different and independent analytical methods, the PIDIE samples can turn out to be a very useful means for verifying the isotopic composition of plutonium.

Thus, PIDIE has made it possible to understand better the possibilities offered by X and gamma-ray spectrometry for the non-destructive monitoring of nuclear materials.

ACKNOWLEDGMENTS

The authors wish to thank all the participants who found ways and means to make their contributions to this exercise. PIDIE is the result of the work of many people, in particular, the original initiative from Mr. A.S. Adamson and his contribution together with that of Mr. H. Ottmar in formulating PIDIE and obtaining the nuclear material for the samples. We wish to pay special tribute to the continued efforts of the late Mr. F.J.G. Rogers during the long period before the samples were dispatched to the participants.

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FISSILE MATERIAL MEASUREMENTS AND DETECTION LIMITS BY GAMMA-RAY SPECTROMETRY IN 220 l WASTE DRUMS

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Abstract

The estimation of the content of fissile materials in routinely produced waste drums necessitates appropriate measurements to meet material balance requirements as well as sorting criteria for disposal.

About 500 typical 220 l waste drums containing incinerator ashes and compactable materials have been assayed by passive gamma-ray spectrometry. The detection system has been calibrated by ^{152}Eu sources. Selfattenuation corrections and varying source inhomogeneities have been investigated. From the measurement results the fissile material content in the wastes has been determined and detection limits are predicted.

The performance of a relatively simple and fast measurement technique for fissile material content in typical waste drums provides valuable field data in support of material balance system studies and waste segregation policies.

1. Introduction

Part of the characterization of nuclear waste barrels is the determination of the inventory and the quantity of the radionuclides present in the barrel.

Generally, the methods for the determination of the nuclide inventory of a container are of a non-destructive nature, because in many cases it is not feasible to take a representative sample for destructive analysis. These NDA methods generally rely on gamma-ray spectrometry or neutron counting of the radiation emitted by the drum. Both passive and active techniques have been investigated and applied in the past.

Of those methods high-resolution gamma-ray spectroscopy (HRGS) has proven its utility over the years. It has however inherent limitations because of gamma-ray attenuation which is dependent on gamma-ray energy as well as on matrix material density /1/.

Using techniques which correct for this phenomenon, it has been possible to assay some 500 drums that differed in matrix density, composition, origin or drum size.

The experience gained by the routine use of HRGS on this batch of waste drums is reported here.

2. Experimental set-up and equipment

2.1. Drum scan mechanism : Segmented gamma-scanner

The assay mechanism for the drums consists of a turntable, a vertical motion unit and a detector with collimator.

The rotation of the drum during the measurement averages out the radial inhomogeneity of the assayed material. The vertical motion (with 5 predetermined, computer controlled measurement steps) in combination with the use of a detector collimator enables the measurement of the drum in 5 segments.

By performing a separate assay on each segment, the effect of vertical inhomogeneity is reduced. A transmission source on the opposite side of the drum with respect to the detector provides data for the matrix attenuation correction in each segment.

2.2. Detector

The detector in this set-up is a Philips Ge(Li) detector coupled to a preamplifier and mounted on a 35 l cryostat. The efficiency of the detector is 10.9 %.

2.3. Electronics

The electronic units consist of a spectroscopy amplifier (Canberra Model 2020) and a fast ADC (Canberra Model 8075) linked to the Canberra System 100. This is an advanced PC based Multichannel Analysis Workstation that features high performance data acquisition, analysis and report generation. Further it corrects for the dead time and pulse pile-up effects in the shaping amplifier and ADC.

It runs under Microsoft Windows which expands the DOS operating system allowing multiple application programs to be run simultaneously on the host computer (Compaq 386/25). The spectra are stored on the hard disk.

Spectra analysis was hand made through the indication of the regions of interest. Peak centroid determination, background subtraction and peak integration is made by software. Energy calibration was carried out with the ^{152}Eu source. The efficiency curve was registered in a separate measurement with an ^{152}Eu source. These elements allowed the identification of the nuclides and the determination of their quantities.

A more sophisticated peak analysis program (SPECTRAN-AT) is available. It fits a semi-gauss curve to the peak and decomposes multiple peaks. With the use of an efficiency curve, the nuclide vector can be calculated.

3. Sample description

The 500 waste drums that have been assayed to the present time with the segmented gamma scanner, can be classified in a few main groups : they may contain ashes, compressible wastes or concrete conditioned waste.

Both 180 l and 220 l size drums had to be measured. In some cases 180 l drums were overpacked in 220 l drums.

For the ashes from the incineration of combustible waste a density of about 450 kg/m³ was reported. The concrete filled barrels (always 220 l size) had net weights between 500 and 550 kg. On the other hand the density of the compressible waste varied from very low (100 kg/m³ like thermal insulation materials) to high (1000 kg/m³ for metal or glass parts).

4. Measurement procedures

All barrels have been scanned in 5 segments and each segment was measured for 300 seconds. The spectra accumulated for each individual segment were added so that finally one spectrum per barrel was registered.

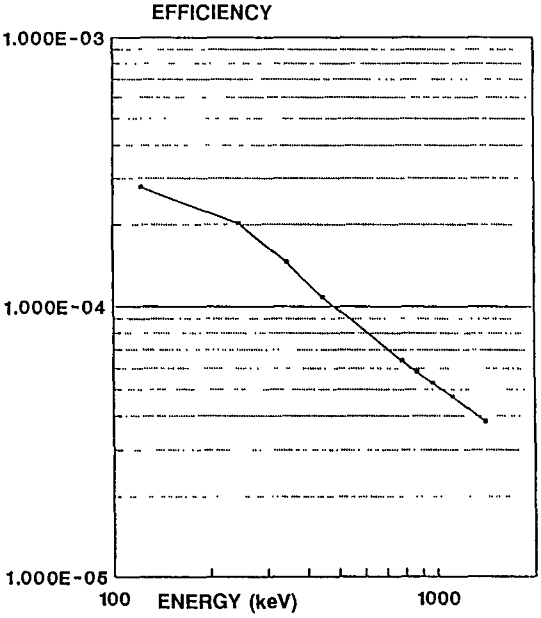
The interpretation of the experimental results was different depending on the waste category.

4.1. Ash barrels

The ashes of the Evence Coppée furnace had a mean density of 400 to 450 kg/m³ and were supposed to be homogeneous over the entire barrel volume. Calcined clay balls have been shown to provide a satisfactory non-active simulator for these ashes.

The determination of the efficiency curve was carried out with this clay matrix using an ^{152}Eu source located in different radial positions. The efficiency showed little radial dependence, disregarding very excentric positions, and thus represented a fair calibration for the actual measurements. All attenuation effects of the emitted gamma's were intrinsically included in the efficiency curve. An example of efficiency curve is given in figure 1.

FIGURE 1 : Efficiency curve by ^{152}Eu source empty barrel



The nuclide source strength was then calculated with the well known formula :

$$I = \frac{A}{\Delta T \,.\, B \,.\, \varepsilon}$$

where
I = the nuclide intensity
A = the area under the peak
ΔT = the measurement time
B = the branching ratio
ε = the efficiency

4.2. Non-combustible wastes

For the case of non-combustible, compressible wastes, the preceding procedure is not applicable because of the unknown matrix density.

In order to get an adequate attenuation correction factor over the full energy range, an ^{152}Eu transmission source was measured together with the waste barrel.

The comparison with an empty barrel measurement provided the energy dependent attenuation curve and thus allowed the calculation of a correction factor, which was related to half a barrel thickness, as a function of gamma energy.

The obtained spectrum was analyzed, using the efficiency curve for the case without matrix present and the nuclide content was augmented by using its corresponding correction factors from the attenuation measurement. So the formula mentioned in 4.1 is corrected by $e^{\mu x}$ wherein μ is the energy dependent attenuation and x is the waste barrel radius.

4.3. Concrete barrels

Because of the high attenuation of gamma rays due to the concrete matrix, the methods previously discussed cannot be applied and gamma measurements on concrete barrels seem meaningless in general.

However for the case of a homogeneous distribution of the radioactive material in the concrete matrix, a method /2/ has been described to establish a correlation between the specific activity of voluminous radioactive waste and the count rate in a collimated external gamma detector.

5. Experimental reuslts

5.1. Detection limits

Detection limits have been determined for ^{235}U and ^{239}Pu as they could be measured by their gamma emissions. They are based on the analysis of a typical spectrum, and although this calculation is spectrum dependent and affected by the presence of other radionuclides, the order of magnitude is representative. The procedure that was followed has been described by Currie /3/.

The lower limit of detection L_D is given by the following formula :

$$L_D = 2.71 + 4.65\ \sigma_B$$

wherein σ_B means the standard deviation on the observed background value.

The coefficients used in the expression correspond to a 95 percent confidence level.

The activity detection limit a_D can be obtained from the following formula :

$$a_D = \frac{L_D \, . \, C}{\varepsilon \, . \, B \, . \, \Delta T}$$

wherein
C = $\Delta T . (1-e^{-\lambda . \Delta T})$
ε = counting efficiency for the gamma-ray considered
B = the photon emission probability.

Applied to our example we found the results as reported in table I.

TABLE I : Detection limits for fissile isotopes isotopes for ash barrels

	Background uncertainty σ_B	Detection limit counts	Efficiency	Detection limit mg
^{235}U				
185 keV	727	3382	24.10^{-5}	204
^{239}Pu				
129 keV	485	2258	28.10^{-5}	37
375 keV	283	1319	13.10^{-5}	188
413 keV	390	1816	12.10^{-5}	294

5.2. Mean composition of different waste barrels.

All waste barrels have been scanned and the gamma spectra were processed as described in paragraph 4. After analysis, the nuclid inventory could be determined : the distribution over the barrel population is given in table II.

Considerable differences can occur between the different waste barrels and values vary between zero and the maximum. The distribution is given as the number of barrels containing a given percentage of that maximum quantity. The numbers are reproduced for three classes of wastes : pool ashes, pool compressible wastes and miscellaneous wastes.

It becomes clear from the distributions that the maximum quantities are only obtained in a few cases while the highest frequency lies in the category containing up to one quarter of that maximum amount.

There are also serious differences between different groups of ashes and compressible wastes, depending in some cases on the origin and the reactor type.

Less isotopes could be identified in the compressible waste category because of the measurement method applied : a transmission source was used, thus increasing artificially the background and masking the presence of weak isotopes.

5.3. Mean attenuation in the matrix

Matrix attenuation has been taken into consideration in a particular way as described in paragraph 4. In case of the ash barrels, the matrix could be simulated by non active material and attenuation effects were accounted for in the efficiency curve.

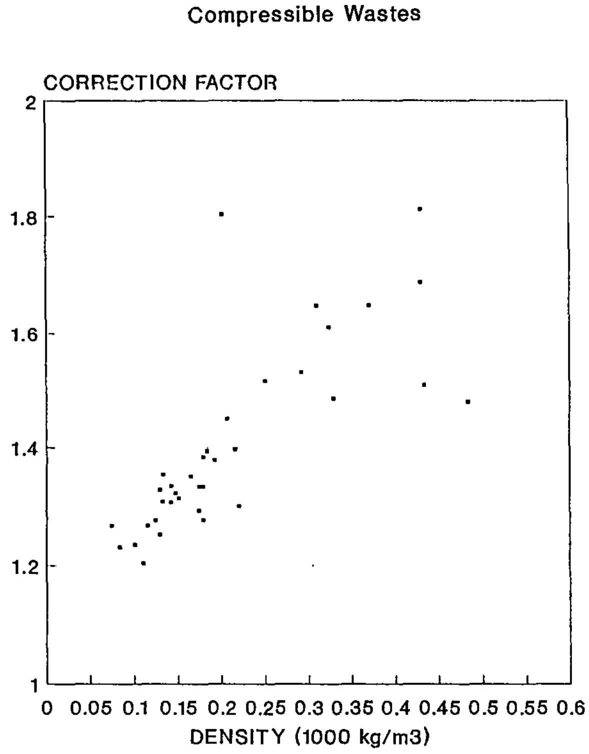
TABLE II : Distribution of Radionuclides in the waste barrels

	Maximum content	Number of barrels with inventory relative to maximum value					
	MBq	0	0-10 %	10-25 %	25-50 %	50-75 %	> 75 %
Pool Ashes - Total barrels 189							
^{57}Co	84.65	179	188	-	-	-	1
^{60}Co	777	0	32	124	21	3	1
^{54}Mn	6.7	5	34	76	69	7	3
^{134}Cs	9.3	115	156	26	4	0	3
^{137}Cs	176.2	7	174	12	1	1	1
^{235}U	4.01	155	164	14	5	4	2
^{125}Sb	31.3	11	77	52	44	13	3
Pool Press - Total barrels 84							
^{60}Co	104	26	26	53	0	4	1
^{54}Mn	9	74	74	8	0	0	2
^{137}Cs	31.4	57	57	26	0	0	1
Miscellaneous Waste - Total barrels 70							
^{60}Co	56.7	9	66	2	1	0	1

In case of non combustible but compressible waste, a transmission measurement was made with a ^{152}Eu source. Comparison with an empty barrel, provided a series of correction values over the energy region of 120 keV to 1.4 MeV, to which a function of the form $y = a_1 \exp(a_2x)$ was fitted, with y the correction factor and x the gamma-ray energy.

Because this procedure was rather complex for routine measurements, a simplified method was investigated, based on the weight determination and calculation of the mean density of the matrix.

FIGURE 2 : Correction factor distribution for ^{60}Co



A correction factor for the ^{60}Co line is given in figure 2 as a function of matrix density. It turns out that all corrections can be made within an error of 10% over the entire energy region.

6. Conclusion

Different kinds of waste categories have been investigated routinely for the determination of the nuclide inventory. For this purpose HRGS has been used succesfully. Gamma-ray attenuation correction over the entire energy region could be simplified by the determination of the barrel density.

Although not particularly suited for the determination of the alpha content in voluminous waste barrels the detection limits for U and Pu have been investigated and amount to approximately 200 mg per barrel for the lower density wastes.

7. References

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UNATTENDED MODE MONITORING OF HIGH RESOLUTION GAMMA-RAY SPECTRA

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Abstract

An Isotope Monitoring System (IMS) for unattended spectrum acquisition is described. This consists of a simple low cost flexible software package running on a Compaq 80386 and controlling up to 4 Canberra Packard System 100 multi-channel analyzer (MCA) cards. The IMS permits the independent configuration of each of the 4 MCA cards for different monitoring cycles and for different spectrum acquisition cycles each being based upon different trigger criteria. In this way IMS is able to automatically identify events, time tag them, and acquire and store valid spectra corresponding to those event.

An additional feature of IMS permits to run a Multi-Group Analysis (MGA) software package for the determination of plutonium isotopic compositions in batch mode.

One particular application is discussed which consists of 4 high resolution gamma-ray detector systems connected together to the IMS for unattended spectrum acquisition. The off-line batch mode analysis of the spectra using MGA is also discussed.

1. Introduction

The Euratom Safeguards Directorate is being increasingly called upon to develop and install non-destructive assay (NDA) systems for unattended operation in new large scale nuclear facilities in the European Communities. Some of these NDA systems consist not only of neutron coincidence counters but also of high resolution gamma-ray detectors. The introduction of NDA measurement systems for unattended operation becomes obligatory:

- when nuclear material is handled automatically in remote controlled nuclear facilities
- where nuclear material is inaccessible for measurement or sampling by inspectors in the normal way
- to reduce the radiation dose to operator staff and inspectors caused by the increased number of measurement needed on large quantities of plutonium, i.e. higher throughputs and larger quantities in a single item
- when the investment in new measurement systems for unattended operation becomes cost effective due to the considerable increase in manpower overheads associated with the use of conventional attended measurement systems in these new large scale nuclear facilities.

The first substantial effort to design NDA safeguards measurement systems for unattended operation was initiated in the Los Alamos National Laboratory (LANL) for neutron coincidence counters in the PFPF project/1/. Recently the Euratom Safeguards Directorate has also developed an acquisition system having similar features/2/. LANL thus laid down the fundamental guide-lines for future development of unattended safeguards measurement systems, and in particular neutron coincidence counter systems. These principles can be summarised as follows:

- redundancy in neutron coincidence electronics and computers
- continuous collection with short measurement times, e.g. 1 minute
- infrequent data review, e.g. 1 per month
- sealed cabinets
- continuous cable runs
- software self-diagnostics
- data collection in electronics cabinets near to the detectors.

The Euratom Safeguards Directorate took these guide-lines and investigated the possibility of adapting them to the case of unattended gamma-ray spectra acquisition. The following principles were thus established:

- separate spectrum acquisition from spectrum analysis
- no monitoring mode operation, i.e. the monitoring mode of the neutron coincidence counters can be considered as being part of the C&S measures
- no connection between gamma-ray spectra and external hardware, e.g. neutron coincidence acquisition electronics, etc.
- use of commercial electronics and spectrum presentation software
- a hardware platform based upon the IBM compatible personal computer
- use of closed cycle helium refrigerators for the gamma-ray detectors
- reduction of the data to be stored through "front-end data processing" involving:
    - triggering only on the item once positioned in front of the detector
    - only storage of spectra of interest
    - user definable thresholds to exclude background spectra from spectra of interest
    - user definable frequency for logging the correct functioning of the hardware

- status log indicating the type of acquisitions performed.

There are many different existing high resolution gamma-ray systems having sophisticated automated acquisition cycles for unattended operation but all of them appear to be linked to external hardware. As an example, one recent application of the TRIFID/EPICS system/3/ used a hardware trigger from an external controller to start the spectrum acquisition. Another important different between the approach described here and that often implemented elsewhere is that in the other systems the spectrum analysis is performed immediately after acquisition. In the approach described here the immediate analysis is not possible since the system is connected to 4 different spectrum acquisition chains and the execution of any complex analysis routines would adversely affect the performance of the internal triggering software and would cause conflicts should events occur simultaneously at two, or more, of the acquisition chains.

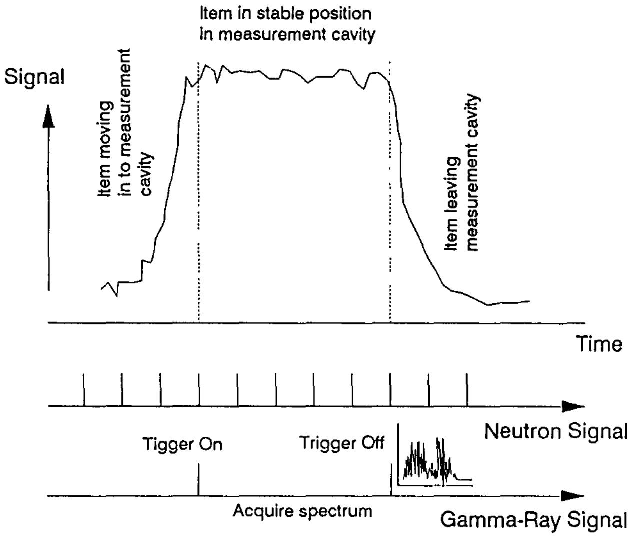


Fig. 1 The Measurement Signal as a Function of Time

The difference between the approach for neutron coincidence raw data acquisition and spectrum acquisition is best shown by reference to Fig. 1. The neutron coincidence counter raw data is collected in a way which is independent of the passage of the items through the measurement cavity, i.e. there is no triggering of acquisition as a function of the items passage through the detector cavity. It is true that the raw neutron coincidence data are analysed during collection to determine if they belong to a background event, a source, or the passage of a plutonium item, however it is only during the review phase that data is identified for subsequent analysis. This approach can not be efficiently used for the unattended acquisition of gamma-ray spectra since spectra should only be acquired when meaningful, i.e. when the item has stopped in the correct measurement position. The archival overhead for the collection of uninteresting spectra, e.g. from background or during the arrival or departure of the item, would be substantial if the system were to have, say, 30 days autonomy. Another important boundary condition was the optimisation of the acquisition time since for some applications the residency time of the item in front of the detector is as little as 4 minutes. It was thus not possible to adopt a triggering approach which would reduce the time available for spectrum acquisition, i.e. the spectrum acquisition must be started as soon as the item is positioned correctly and stopped just before the item moves on.

Once the principles mentioned above were established the Euratom Safeguards Directorate decided to adopt a commercial multi-channel analyser (MCA) card, the Canberra S100 MCA card, and an IBM compatible personal computer, and to develop application software using the software supplied with the MCA card selected.

The Euratom Safeguards Directorate considers that the selection of the S100 MCA card and personal computer have the following advantages:

- installation of up to 4 MCA cards in an IBM compatible personal computer
- software provides a common user interface, Microsoft Windows 3, with other applications under development
- flexible commercial spectrum presentation package
- professional user descriptions/manuals
- limited cost for purchase and maintenance
- flexible, reasonably priced, development engine for safeguards applications.

As mentioned above a custom applications software was developed, the Isotope Monitoring System (IMS). The role of IMS is to acquire spectra only when a plutonium item is positioned in front of the gamma-ray detector and to do this automatically for 4 different measurement systems in an unattended way. Once started IMS controls the 4 different gamma-ray acquisition systems in a monitoring, or cycle, mode. This involves collecting continuously, with each of the S100 cards, spectra for short periods of time. Each spectra is examined for the total counts in a user defined peak region of interest (ROI) and for the ratio of that peak ROI with a user defined background ROI. A flag is set if the total counts in the ROI is in excess of some user defined threshold. This, in principle, indicates that an item is arriving. However, in order to distinguish this from a simple increase in background, a second flag is set if a user defined ROI ratio threshold is also exceeded. If neither of the thresholds are exceeded, for a given spectrum, it is erased and a new monitoring cycle started.

When the user defined thresholds are exceeded IMS changes from the monitoring, or cycle, mode to acquisition, or sampling, mode. This change is made only for the measurement system where an item has been detected, the other measurement systems remain in monitoring mode. For the acquisition, or sampling, mode one or more spectra can be acquired and stored for future analysis. Each spectrum acquired corresponds to a user defined time slot, or period, corresponding to a period when the items is known to be correctly placed in front of the gamma-ray detector. When the acquisition, or sampling, mode is completed the monitoring, or cycle, mode is reintroduced until the next item is detected.

2. Automatic Plutonium Gamma-Ray Spectra Acquisition

System Hardware

The system hardware can be separated in to two parts, that linked with the gamma-ray detector system, and that linked with the MCA card and computer. The first type of hardware will be listed but not described in detail. The following components were used:

- HPGe LEGe detector 200mm^2 x 10mm
- Canberra CRYOLECTRIC electrically refrigerated Cryostat
- Canberra Model 3105 High Voltage Power Supply
- Canberra Model 2025 Fast Research Amplifier
- Canberra Model 8077 Fast ADC
- Canberra Remote ADC Transmitter/Receiver Units.

The gamma-ray detector hardware layout has three novel features:-

(i) The use of closed cycle helium refrigerators running continuously unattended.

(ii) The use of custom ADC transmitter/receiver units in order to connect the ADC's to the S100 MCA cards. The ADC's were placed about 7m from the gamma-ray detectors and the S100 MCA cards and computer were connected via the ADC transmitter/receiver units. The connection between the ADC transmitters and receivers was over twisted pairs which can be up to 500m long and provide full ADC data transfer.

(iii) The fact that the gamma-ray detectors are mounted on to neutron coincidence counters so that simultaneous measurements are performed.

The Canberra S100 MCA card was selected because of its known compatibility with the other Canberra equipment used in the measurement chain and because of the flexible software that is supplied with the card. One S100 master card and 3 S100 slave cards were used.

The four S100 MCA cards were installed in a Compaq 386-20e. The hardware layout is shown in Fig. 2.

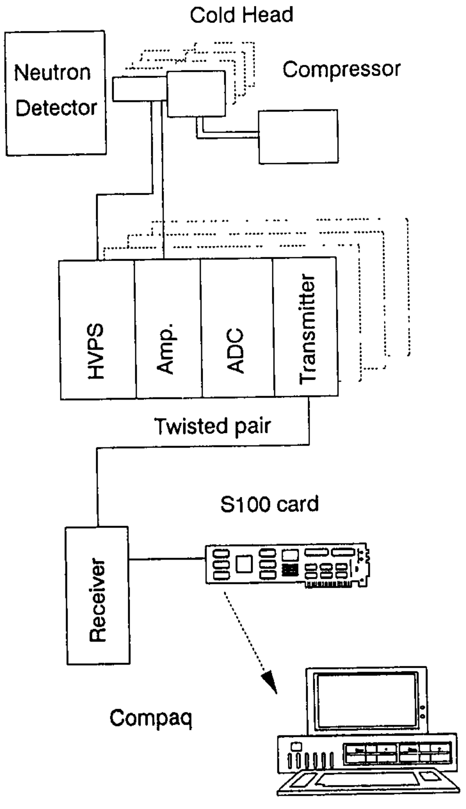


Fig. 2 Hardware Layout

System Software

The system software relies upon two different sets of software, the first the commercial software for the Canberra S100 MCA card, the second the specific application routines programmed for the Isotope Monitoring System (IMS).

Application Software

The S100 MCA card was packaged with Microsoft Windows 3 and the Canberra Application Manager. The Application Manager came with three components, the first the application manager itself which provides for the control of the S100 cards, second the Microsoft spreadsheet EXCEL providing the communication link in to the S100 cards, and the MCA command summary which provides EXCEL with a library of commands.

The custom software, IMS, was thus written using EXCEL macros and the library of S100 commands. The use of EXCEL has many advantages since custom menus, dialogue boxes, and user messages can be employed to enhance the user friendliness of the system.

Isotope Monitoring System

As has been stated above the user, in starting the Isotope Monitoring System (IMS), initiates the program through a macro in an EXCEL spreadsheet. The presence of up to 4 S100 MCA cards are detected automatically, the communication DDE link established, and the cards configured according to a set of configuration files.

Once the S100 cards have been configured the main menu of IMS is presented, see Fig. 3. The main user menu provides options for:

- starting monitoring
- configuring each of the 4 MCA cards
- accessing the status file
- accessing the spectrum files
- leaving system.

The first three options are linked directly to the IMS, whilst the last two are service options.

Start Monitoring

The start monitoring option accesses an additional menu screen, see Fig. 4, where the user can select:

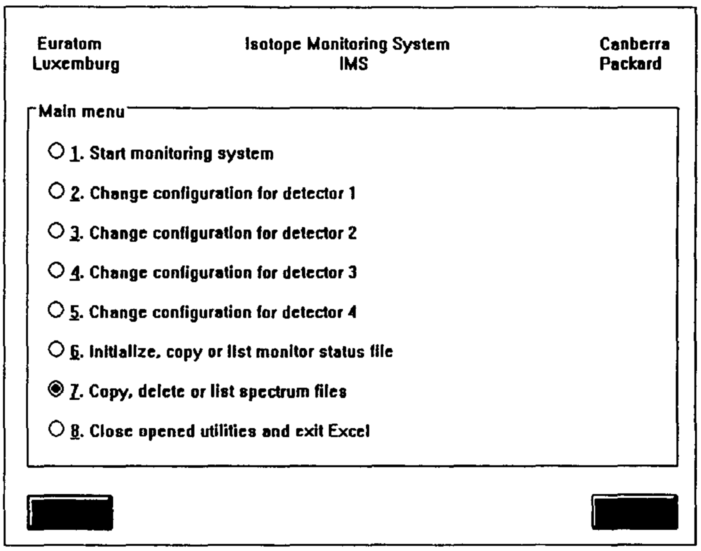
- which of the 4 S100 MCA cards are to be used
- the frequency with which status information on the correct functioning of the system is to be written to the status file
- the first spectrum index file number to be used.

The status information is a state of health signal written to the status file so that the user, upon review, can confirm that the system was functioning correctly. This status information is only written if nothing happened during the time period specified. The status file is updated if nothing occurs, say, during 1 hour, and continues to be updated every hour.

MCA Card Configuration

The option to configure one, or more, of the S100 MCA cards leads the user to another menu screen, see Fig. 5, the gamma-ray detector configuration menu. This menu is where the different monitoring thresholds and sequences are programmed.

A schematic of the monitoring and acquisition modes is shown in Fig. 6, and is described in detail below.



Euratom
Luxemburg

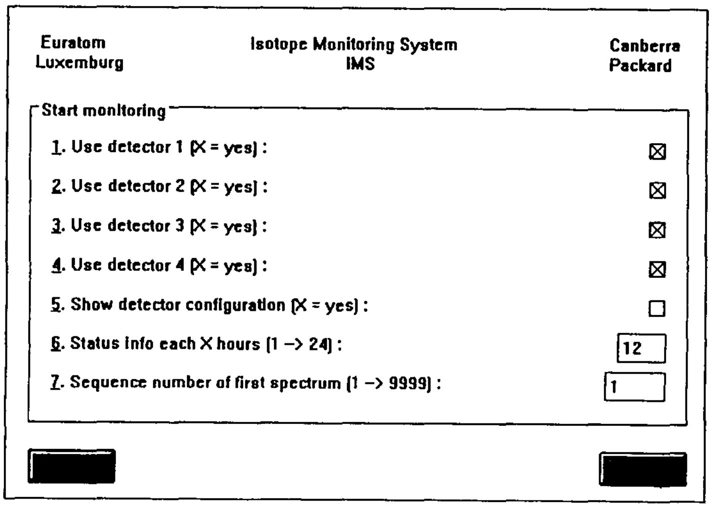
Isotope Monitoring System
IMS

Canberra
Packard

Main menu

○ 1. Start monitoring system
○ 2. Change configuration for detector 1
○ 3. Change configuration for detector 2
○ 4. Change configuration for detector 3
○ 5. Change configuration for detector 4
○ 6. Initialize, copy or list monitor status file
● 7. Copy, delete or list spectrum files
○ 8. Close opened utilities and exit Excel

Fig. 3 The IMS Main Menu



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IMS

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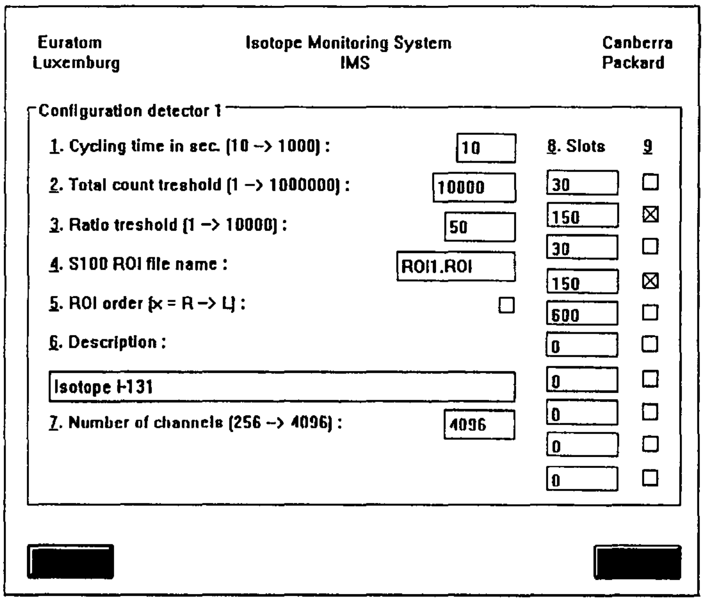
Packard

Start monitoring

1. Use detector 1 (X = yes) : ☒
2. Use detector 2 (X = yes) : ☒
3. Use detector 3 (X = yes) : ☒
4. Use detector 4 (X = yes) : ☒
5. Show detector configuration (X = yes) : ☐
6. Status info each X hours (1 -> 24) : 12
7. Sequence number of first spectrum (1 -> 9999) : 1

Fig. 4 The IMS Start Monitoring Menu

Fig. 5 The IMS Gamma-Ray Detector Configuration Menu



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IMS

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Packard

Configuration detector 1

1. Cycling time in sec. (10 -> 1000) : 10
2. Total count treshold (1 -> 1000000) : 10000
3. Ratio treshold (1 -> 10000) : 50
4. S100 ROI file name : ROI1.ROI
5. ROI order (x = R -> L) : ☐
6. Description :
Isotope I-131
7. Number of channels (256 -> 4096) : 4096

8. Slots	9
30	☐
150	☒
30	☐
150	☒
600	☐
0	☐
0	☐
0	☐
0	☐
0	☐

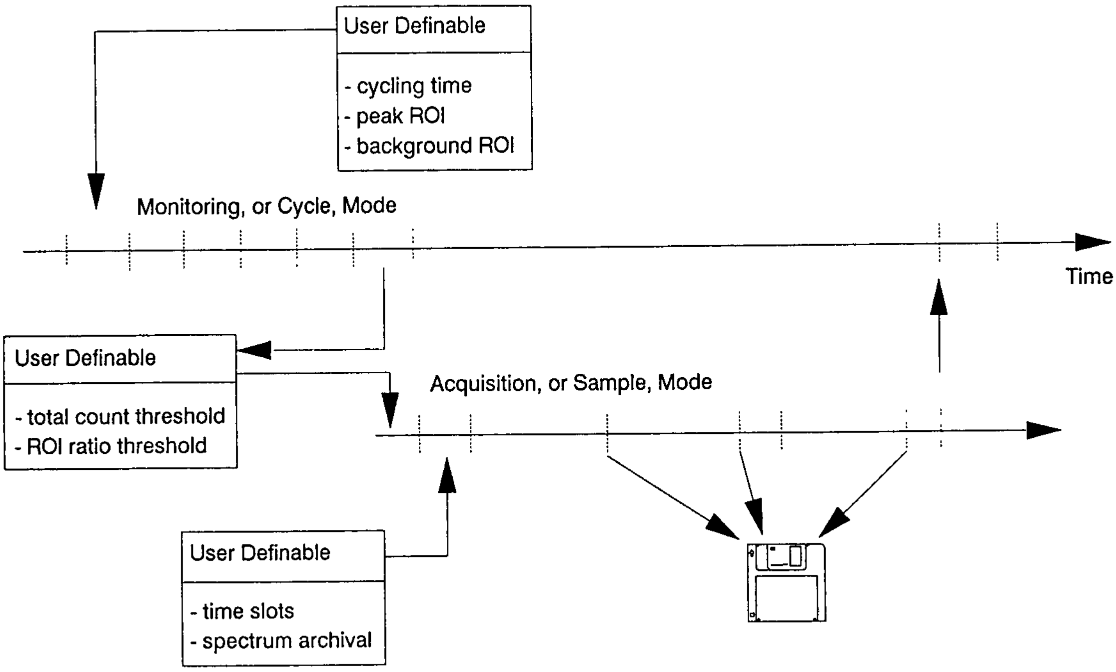


Fig. 6 Schematic of Monitoring and Acquisition Modes

The user is presented with two distinct sets of configuration parameters, the first related to the ability of the system to detect an item arriving, the second related to the acquisition of one, or more, spectra.

The first set of fields, numbers 1 to 5, relate to the detection of an item, i.e. the monitoring, or cycle, mode. The definition of the fields are as follows:-

1. Cycling time in sec. (10 -> 1000)

The acquisition time for which a "background" spectrum will be acquired and evaluated to determine if an item is arriving. This time must be set as a function of the speed of movement of the items as they are handled and presented before the gamma-ray detector. The user can define a period between 10 and 1000 seconds.

2. Total count threshold (1 -> 1000000)

A integral count is calculated for a predefined cycle time, see field 1, and a predefined ROI, see field 4, and compared to the user defined total count threshold. If the threshold is exceeded then IMS considers that an item is arriving and the spectrum acquisition sequence is triggered. This threshold can be set so as to ensure that an acquisition is not triggered by a background signal or by event defined not to be of interest. The user can define a threshold of between 1 and 1000000 total counts in the ROI.

3. Ratio threshold (1 -> 10000)

A ratio value is also calculated for the same cycle period as for the threshold calculation, field 2. The ratio is defined by the ROI field 4 which provides a file name where the two ROI's are defined and by the location of the background ROI to the peak ROI, field 5. The ratio threshold provides for a isotope specific trigger independent of the total count trigger. It is thus possible to use the total count and ratio thresholds in order to finely select the type of event for which spectra are required, and to ensure that the triggering of spectrum acquisition is resistant to background changes or the passage of items having other isotope signatures. The user can define a threshold of between 1 and 10000 for the ratio of the ROI's.

4. S100 ROI file name

This file contains the two user defined ROI's which are used for the determination of the total count threshold and the ratio threshold. The user can identify, and define, the exact ROI's to be used with the normal S100 application manager software.

5. ROI order (x=R -> L)

This indicates which of the two ROI's is the peak ROI and which the background ROI.

Field 6 provides for an identifier tag which would be useful if the different S100 MCA cards were set to trigger on different isotope specific ROI's. The field 7 defines the number of channels to be acquired in the S100 spectra.

Once a item has triggered either the total count threshold or the ratio threshold the S100 must be configured for the acquisition of spectra for archival and subsequent off-line analysis, i.e. the acquisition, or sampling, mode. Fields 8 and 9 provide for the user to configure the acquisition time and number of spectra to be collected. The slots are user definable sequential time periods, or slots, during which the S100 MCA card will be instructed to acquire spectra. The field 9, corresponding to each time slot in field 8, permits the user to indicate if that particular spectrum, once acquired, should be stored on the hard disk of the computer for future off-line analysis. This arrangement of user definable time windows and spectrum archival options provides the user with the following options:-

(i) Not to store a spectrum acquired during an initial period when the item is known still to be moving. If the trigger thresholds are set to be sensitive to small items then larger items will trigger the acquisition prematurely. It is for this reason that the user can decide not to store the first spectrum acquired.

(ii) The user can ask to store one, or more, spectra during the period when the item is known to be in position in front of the gamma-ray detector.

(iii) If it is known that the item will move at predetermined times and that different portions of the item will be automatically presented to the gamma-ray detector for measurement then the user can insert short periods where no spectra will be stored.

(iv) At some specific time the item will be automatically removed from in front of the gamma-ray detector and the acquisition of the last spectrum will have been stopped and the spectrum stored. If the control fo the S100 MCA card were to be handed back directly from the acquisition mode to the monitoring mode then it is probable that the acquisition mode would be retriggered by one, or other, of the thresholds. In order to avoid this an additional time period can be inserted at the end of the acquisition cycle although no spectrum would be stored for that period.

Each of the 4 S100 MCA cards can be configured differently in the gamma-ray detector configuration menu.

Status File

The status of an active S100 MCA card is regularly dumped to a status file and can be read and listed by the user though the main menu. Each status line in the status file, or status log, is date and time tagged and has a syntax ff ff ff ff with each ff representing one of the S100 MCA cards. The ff can have the following different states:

- 00 means that the S100 MCA card did not receive a trigger
- 10 means that the total count threshold was exceeded
- 01 means that the ratio threshold was exceeded
- 11 means that both the trigger thresholds were exceeded.

From this status log the user can clearly see the source of all triggering operations and when they occurred. In addition the status log provides a rapid overview of the number of spectra that have been acquired and stored.

There is a second type of status log with a syntax f f f f, with each f representing one of the S100 MCA cards. The f can have the following states:

- 0 means that particular cards was not selected by the user
- 1 means that monitoring was initiated with that card
- 2 means that the card was not available at the end of a monitoring cycle
- 3 means that the card became again available at the end of a monitoring cycle
- 8 means that a spectrum storage problem occurred
- 9 means that a communication problem occurred with the card.

The 0 and 1 are written when a monitoring cycle is initiated and 2 and 3 written when the monitoring cycle is completed.

Other Features

Access to the spectrum files is through an additional user menu which provides the user with options to copy and delete spectra, to display or print spectrum file lists by month, day, S100 MCA card, etc.

3. Gamma-Ray Spectra Analysis Using MGA

In section 2 the gamma-ray spectrum acquisition routines were described. The resultant gamma-ray spectra once acquired and stored are available for further off-line analysis. The gamma-ray spectrum acquisition routines are configurable for the acquisition of any type of gamma-ray spectrum, i.e. they are not specific to the acquisition of plutonium gamma-ray spectra. In the particular application described above the objective was to acquire plutonium gamma-ray spectra in an automated and unattended manner, and to provide for the off-line analyse of those spectra using the Multi-Group Analysis (MGA) program developed at (LLNL)/4/.

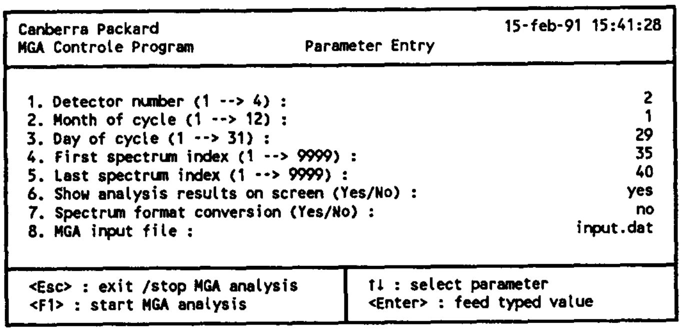
Once the different plutonium gamma-ray spectra have been acquired in unattended operation the user can intervene manually to perform the analysis using MGA in a "batch mode", see Fig. 7.

The menu screen is divided in to three parts:

- the upper header contains title information and the date and time
- the central part shows the program parameters
- the lower part shows the menu options and information about the command keys.

The program parameters are as follows:

- gamma-ray detector number x: only spectra mmddiiii.dex where x is the number (1-4) of the gamma-ray detector will be analysed
- month mm of cycle: only spectra mmddiiii.dex where mm is the month during which acquisition took place will be analysed
- day dd of cycle: only spectra mmddiiii.dex where dd is the day during which acquisition took place will be analysed
- first spectrum index $iiii_1$: index of the first S100 spectrum mmddiiii.dex of the series of spectra to be analysed
- last spectrum index $iiii_2$: index of the last S100 spectrum mmddiiii.dex of the series of spectra to be analysed
- show MGAPC analysis flag: the user can select either to see the MGA analysis on screen and initiate the next analysis, or to have the series of analyses performed automatically
- MGAPC spectrum storage flag: the user can select to have the S100 spectrum file format converted to an ASCII format according to the a proposed "ASCII Transfer Spectral Data File Format"/5/
- MGAPC input file name fffffffff: during the execution of



```
Canberra Packard                                    15-feb-91 15:41:28
MGA Controle Program            Parameter Entry

 1. Detector number (1 --> 4) :                                      2
 2. Month of cycle (1 --> 12) :                                      1
 3. Day of cycle (1 --> 31) :                                       29
 4. First spectrum index (1 --> 9999) :                             35
 5. Last spectrum index (1 --> 9999) :                              40
 6. Show analysis results on screen (Yes/No) :                     yes
 7. Spectrum format conversion (Yes/No) :                           no
 8. MGA input file :                                         input.dat

 <Esc> : exit /stop MGA analysis    ↑↓ : select parameter
 <F1> : start MGA analysis          <Enter> : feed typed value
```

Fig. 7 The Front End Menu to MGA

MGA the program requests for user input are directed to an input file generated by the user to permit automatic analysis.

Upon execution of the program the first spectrum file to be analysed is that defined by the gamma-ray detector number, day and month of acquisition, and the spectrum index number. The program ensures that the MGA analysis is performed for each spectrum having an index number between iiii$_1$ and iiii$_2$. The spectra are analysed according to the input data in the MGA input file fffffffff generated by the user. A completed analysis is then printed if the correct instruction is embedded in the input file fffffffff. Should MGA produce an error message, via its error file, the error is classified in to fatal or non-fatal. Fatal errors stop the control program, non-fatal errors require user input to continue. The S100 spectra can also be converted to the ASCII format if requested.

4. Discussions

The use of a Canberra S100 MCA card when coupled with the special software IMS provides a reasonably versatile system for both unattended spectrum acquisition. The position of the Euratom Safeguards Directorate is that this solution provides one approach to the problem of unattended spectra acquisition and subsequent off-line data analysis. The Euratom Safeguards Directorate views this solution as being particularly effective when:

- unattended gamma-ray spectra acquisition is required without connection to external hardware to obtain triggers
- only assay functions are needed, i.e. no C&S role
- when immediate data analysis is not required, i.e. when the inspector only expects to service the data acquisition system, say, every week or every month
- when subsequent off-line data analysis is acceptable, i.e. when there are only a limited number of items passing through the detector cavity.

The implementation as described here is particularly addressed to the case where the item handling routines are automated, very well defined, and reproduced identically for each item presented before the gamma-ray detector. In addition it is assumed that the time available for acquiring the spectrum is normally very short, i.e. 4-5 minutes, so that the trigger technique must maximise the time available for spectrum acquisition. A future area of development for the IMS application software could be the addition of other trigger techniques for determining when to start and stop spectrum acquisition, i.e. monitoring of the stability of throughput or dead time, etc..

The IMS application software itself is not linked to the acquisition of plutonium spectra for analysis with MGA. Other applications could involve the unattended performance of enrichment measurements or, through an improved status log, the simple counting and identification of radioactive items passing before the gamma-ray detector. The S100 card and the IMS could equally be adapted for NaI type detectors.

Areas where the Euratom Safeguards Directorate is concentrating its future developments for unattended measurement systems are:

- unattended operation of combined neutron and gamma-ray measurement systems
- networking data acquisition systems to provide rapid data analysis and failure detection
- automatic recognition and analysis of events of safeguards importance.

5. Conclusions

This paper describes a simple, low cost and flexible solution to the unattended acquisition of gamma-ray spectra.

The solution consists of a dedicated software package running on a Compaq 80386 and controlling up to 4 Canberra Packard System 100 multi-channel analyzer (MCA) cards. This system is called the Isotope Monitoring System (ISM) and permits the independent configuration of each of the 4 MCA cards for different monitoring cycles and for different spectrum acquisition cycles each being based upon different trigger criteria. The IMS is able to automatically identify events, time tag them, and acquire and store valid spectra corresponding to those event.

The IMS permits the independent customization of the monitoring mode for each of the 4 MCA cards. Each of the MCA cards has its own monitoring cycle and each provides for time tagged spectrum acquisition when both a gross count threshold and a isotope specific ratio threshold are triggered. Up to 10 separate spectra, each for a different measurement time, can be acquired per acquisition cycle.

In addition a time tagged status file is updated when ever either of the thresholds are triggered. This status file also contains data on the active status of each MCA card and various diagnostic indicators.

An additional feature of the present implementation of IMS permits to run a Multi-Group Analysis (MGA) software package for the determination of plutonium isotopic compositions in batch mode. The MGA package is treated as an independent engine and multiple cycles of MGA can be initiated in batch mode from a menu employing a separate user generated MGA input file.

One particular application is discussed which consisted of 4 high resolution gamma-ray detector systems connected together to the IMS for unattended spectrum acquisition.

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GAMMA RADIATION DETECTOR UNITS ON THE BASIS OF P-I-N CADMIUM TELLURIDE DETECTORS FOR NONDESTRUCTIVE CONTROL OF NUCLEAR FUEL

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Abstract

The results of creation of spectrometry ionizing radiation units on the basis of p-i-n structures from cadmium telluride are presented in the report. The detector and the head-section of the preamplifier were cooled to -35°C by a small-sized thermoelectric cooler. P-i-n structure of the detector allowed to achieve energy resolution on line 122 keV of ^{57}Co isotope - 1,78 keV and 662 keV of ^{137}Cs isotope - 6,41 keV. Peak/valley ratio of the spectral line 122 keV makes the value more than 80.

The possibility of application of the detector for nondestructive control of nuclear fuel arises from research results.

1. Introduction

The technique of nondestructive control of nuclear fuel with gamma spectrometry is widely used in the IAEA for the organization of production and processing of nuclear fuel.

Germanium and scintillation detectors are used mainly for detection of gamma radiation of nuclear fuel.

But lately due to successful creation of detectors with acceptable energy resolution on the basis of cadmium telluride and mercuric iodide there is an opportunity to have devices on the basis of these detectors and methods of control /1,2,3/. Limitations in application of these detectors occur because of the fact that their values of energy resolution and efficiency of registration are not high enough. Values of the parameters depend on the characteristics of initial materials and technological and constructive peculiarities of their manufacture. For precise spectrometry measurements the most important are: energy resolution, range of detected energies, peak/valley ratios, efficiency and sensitivity of registration, stability in time. These parameters are interdependent functions, very often discrepant, it means that of one of the parameters impores the others deteriorate.

Works on creation of detectors on the basis of cadmium telluride were help mainly in manufacturing of detecting units of the M-S-M (metal-semiconductor-metal) type. The best results in this field have been achieved by the scientists of the firm EURORAD (France) /4/: energy resolution of the detectors 2,5 keV on line 59,6 keV and 3,2 keV on line 122 keV. The specifications of such detectors are defined mainly by the parameters of the initial cadmium telluride.

At the present moment it is practically impossible to manufacture detectors on the basis of M-S-M structures with better characteristics. The value of the product of lifetime to the mobility of charge carriers in cadmium telluride is not high. But just this value defines the spectrometry characteristics of the detector ($\mu_e\tau_e \le 3\cdot10^{-3}\,cm^2V^{-1}$, $\mu_p\tau_p \le 3\cdot10^{-4}\,cm^2V^{-1}$).

But at the present time M-S-M CdTe detectors and detecting units on their basis are successfully used for practical purposes in nondestructive analysis of nuclear fuel materials. The authors /1/ put into practice the commercial detectors of EURORAD in the complex with the system of pulse selection and achieved energy resolution < 5 keV at the semiheight for gamma radiation with energy 662 keV. But it is worth mentioning that in this case high energy resolution was achieved by reducing of efficiency count to 20÷50%. With the help of this system the quantitative parameters of the burnup fuel were measured. The results of these measurements agree with the values defined with a high resolution Ge(Li) detector.

The Riga Research Institute's detecting unit with the CdTe detector of semispherical geometry was successfully used for the control of fresh and burnup fuel of NPP /2/. The detecting unit had the energy resolution better then 15 keV for gamma radiation with the energy 662 keV and was able to distinguish in the spectrum gamma-lines of the isotopes ^{134}Cs(605 keV, 796 keV) and ^{137}Cs(662 keV).

The methods of measurements used by the authors is based on measuring gamma-spectra in the region of high energies from 500 to 1000 keV. The range of energies 100÷500 keV is of great interest in solving the problems of nondestructive control of nuclear fuel. And in particular for the analysis of content of Uranium and Plutonium isotopes in mixed fuel. This demands high value of the energy resolution of the detectors just in this region.

The results of the creation of CdTe detectors with p-i-n structure and detecting units on their basis having a wide range of registered energies from 6 to 1000 keV with high energy resolution and contrasting spectral lines allowing to solve various tasks of nondestructive control of nuclear fuel are presented in the report.

2. Results

For manufacturing detectors with p-i-n structure chlorine doped high resistivity cadmium telluride ($10^7 \div 10^9$ oΩ cm) of p-type conductivity was used. It was grown by the method of horizontally oriented crystallization from the melting of stoichiometric composition. Monocrystals of cadmium telluride had the value of the product of mobility to lifetime $(0,5 \div 1) \cdot 10^{-3} cm^2V^{-1}$ for electrons and $(0,1 \div 0,5) \cdot 10^{-4} cm^2V^{-1}$ for holes.

The technology of the creation of p-i-n structures consists of the creation of electron-hole and heterojunctions on the substrates of monocrystals of the initial cadmium telluride. After thermical treatment the plates with p-i-n junction were treated mechanically and chemically for the leakage current reduction. Experimental dependencies of dark current and voltage for the detector with p-i-n structure and M-S-M structure are given in Fig.1.

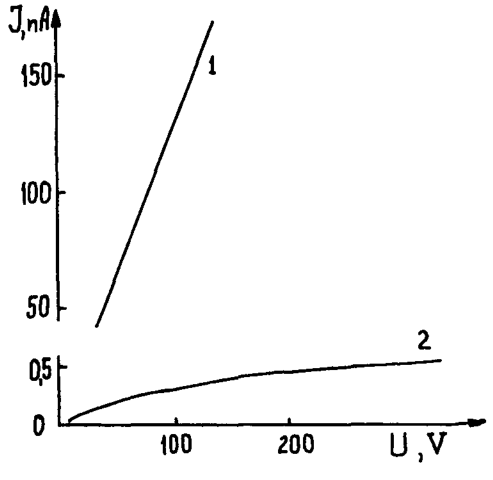

Fig.1. Reverse current as a function of bias voltage for M-S-M structure (1) and p-i-n structure (2).

The advantage of p-i-n structure to M-S-M structure when comparing the value of dark current at the equal voltage is evident. So the usage of p-i-n structure allows to solve the task of realization of high tension of the electric field in the sensitive volume of the detector at considerably less current noises.

The dark current in p-i-n structure has the value $(1 \div 5) \cdot 10^{-9}$A cm^{-2} at 300K and is defined by the processes of generation in the space charge region:

$$I = \frac{e\,n\,W}{2\tau}\text{, where} \qquad [1]$$

e - charge of the electron,
n - concentration of charge carriers in cadmium telluride,
τ - lifetime of charge carriers, c,
W - width of space charge region.

Temperature dependence of the dark current is:

$$I(T) \simeq \exp(-E_g/2KT), \qquad [2]$$

where $E_g(CdTe)$ = 1,48 eV at room temperature.

At temperature -30 ÷ -40°C the value of the dark current is $10^{-12} \div 10^{-13}$A cm^{-2}, giving energy resolution 30 eV which is comparable with the current of germanium and silicon detectors at the temperature of liquid nitrogen. It is possible to achieve such temperatures for p-i-n structure with a help of small-sized thermoelectric coolers based on Peltier effect.

P-i-n structures were influenced by high and low temperatures, infrared radiation (the results will be published separately). The tests show that the width of the space charge region in p-i-n structure which is the sensitive region of the detector limits itself by the concentration of deep impurities with energy levels near the middle of the forbidden zone. In our crystals the concentration of deep impurities has the value $N_T \le 10^{12} cm^{-3}$ allowing to have the width of the space charge region up to 500 mkm. The measurements of the distribution of the electric field in p-i-n structures showed correlation between the thickness of the sensitive region and the lifetime of holes. So in our opinion the main obstacle for having p-i-n detectors with high values of operation volume for cadmium telluride crystals grown according to our technology is absence of perfection in part of concentration of acceptor centres whose energy levels are situated near the middle of the forbidden zone.

When obtaining cadmium telluride crystals with the lifetime of holes $3 \div 5 \cdot 10^{-6}$s it is possible to manufacture p-i-n detectors with sensitive (depleted) thickness area $1 \div 1,5$ mm and high energy resolution. French colleagues /4/ have crystals with such characteristics.

On the basis of the detector with p-i-n structure a detecting unit consisting of a head section and a service unit has been manufactured. The head section is a thermostat on the basis of small-sized thermocooler situated inside the shell-radiator transmitting heat from the hot joint of the thermocooler. The detector and the head section of the chargesensitive preamplifier are on the cold joint and are cooled to the optimal temperature $-30 \div 40$°C. The head detecting unit is connected with the service unit by a flexible cable. The service unit contains a source of a high voltage for the detector , a regulated amplifier for agreeing signals from the output of the detecting unit with the input of the standard pulse analyser (ORTEC 7500, NOKIA or others of the same type).

The energy spectra of ^{241}Am, ^{57}Co and ^{137}Cs received with the help of the detecting unit on the basis of the p-i-n detector are given in Fig.2,3,4.

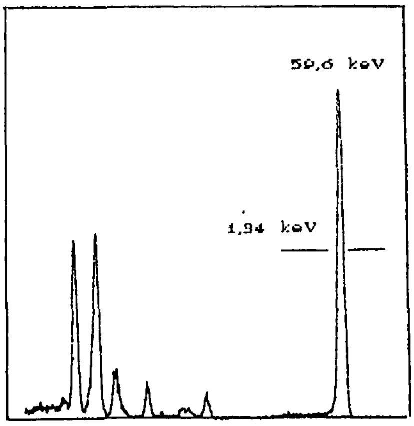

Fig.2. ^{241}Am spectrum detected with p-i-n cadmium telluride detector

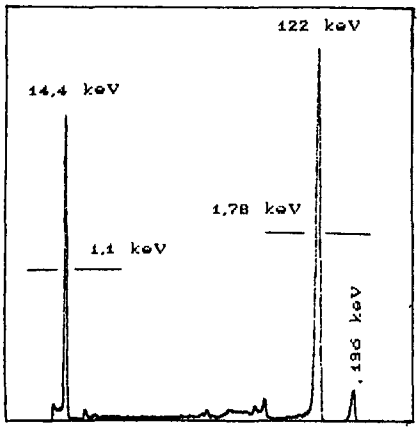

Fig.3. ^{57}Co spectrum detected with p-i-n cadmium telluride detector

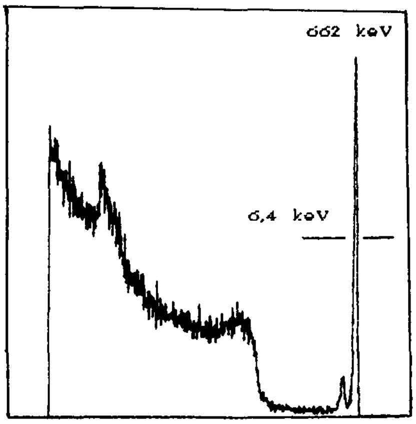

Fig.4. ^{137}Cs spectrum detected with p-i-n cadmium telluride detector

The area of the sensitive service of the detector is 30 mm^2, thickness of the sensitive region - 300 mkm. Detector voltage - 700 V, temperature - 35°C. Energy resolution on a half of amplitude distribution height for energy 144 keV is 1,1 keV at the noise of the generator 1 keV is due to the capacity of the detector 10 pF. The increase of the thickness of the sensitive region to 1 mm allows to decrease the capacity impact of the detector to the noise of electronics and using special lownoise field transistors the noise of preamplifier is achieved with a detector' value of 500 eV. The width of spectral lines 59,6 keV and 122 keV give values 1,34 keV and 1,78 keV respectively. One of the peculiarities of energy spectra of ^{241}Am and ^{57}Co isotopes is contrast and symmetry of spectral lines. Rate peak/valley for 59,6 keV and 122 keV lines is more than 80 and a very typical for CdTe detectors on the basis of M-S-M structure pulling of the left part of spectral lines is absent.

At the detection of gamma radiation of ^{137}Cs isotope the energy resolution on line 662 keV is 6,4 keV at the rate peak/compton more than 3.

3. Conclusion

The given results prove that the detecting unit on the basis of p-i-n cadmium telluride detector can be used for nondestructive analysis of nuclear fuel according to methods used in the works /1,2,3,5/. But detecting units with p-i-n detector will have advantages in comparison with cadmium telluride detectors for better energy resolution as compared with hemispherical detectors and do not demand usage of additional units for pulse selection which are used with the detectors of the firm EURORAD.

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IMPROVED END-POINT PREDICTION IN ISOTHERMAL CALORIMETRY

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Abstract

An "Expert System" software procedure has been developed for use with isothermal calorimeters in order to improve the sample end-point power prediction process. The procedure examines the measurement data and determines the onset of the exponential end-point prediction procedure. It also determines when sample thermal equilibrium conditions have been reached. The procedure has been tested using archived data from two different calorimeters and sample power predictions are obtained in a total measurement time of about 2 hours with sample power errors of less than 0.5%.

1. Introduction

Recent measurements with isothermal calorimeters/1,2,3/have demonstrated both the accuracy in the technique and reductions in measurement times compared to conventional heat-flow calorimeters. The present work has been directed towards further reductions in measurement times. This is motivated, in part, by the needs of safeguards inspectors for short measurement times.

2. Measurements

Table 1 contains results from isothermal calorimeter measurements performed using PERLA plutonium oxide standards. Although the results are generally satisfactory, typically a period of about 4 hours was required for each measurement to achieve a satisfactory predicted sample end power. The reason for the long time period to reach an adequate prediction value can be seen in Fig. 1. The figure is a plot of the measured sample power P (Po-IC) and the predicted sample power P (Pred) as a function of time. From the figure it is clear that the predicted sample power reaches an asymptotic value after a period of about 3 hours and only after a large number of power measurement data points are included in the exponential fitting algorithm. It would appear that the prediction process is being biased by early data points which do not fit the assumed single exponential form of the prediction equation.

3. Analysis

On the basis of measurement data from two calorimeters an analysis of the behaviour of the prediction procedure was undertaken. In particular the behaviour of the coefficient of the exponential was examined. It was found that for most measurements this exponential coefficient reached an asymptotic value in a range of time from about 20 minutes to 60 minutes from the start of the measurement. It was further observed that if the onset of prediction was delayed until the exponential coefficient achieved an asymptotic value a better prediction was obtained at an earlier time. This is in contrast to the previous prediction procedures where the prediction was started after a fixed delay period (usually 15 minutes) following the start of the measurement (sample insertion into the calorimeter measurement chamber).

A software procedure has now been devised to control the prediction process. A rolling value of the exponential coefficient of the fitting function is calculated using 18 averaged sample power data points and the achievement of an asymptotic value is tested using a rolling linear least squares fit to the last six values of the calculated exponential coefficient. The test criteria involve a number of parameters including the slope and standard deviation in slope of the coefficient itself. Once the criteria are satisfied the single exponential fitting/prediction procedure is initiated and similar tests and criteria are used to determine the earliest acceptable value for the predicted sample power.

4. Results and Conclusions

The new procedure has been tested on data from two calorimeters including measurements of both plutonium metal and oxide samples. The results are displayed in Table 2. Sample numbers with a suffix of P1 or P2 have been preheated for one or two hours respectively.

The results are very satisfactory and show that the new procedure works well. At the time of the "first guess", which corresponds to the first predicted value, the error (when compared to equilibrium or certified values) is less than 5% and this is achieved in times ranging from about one hour to about 90 minutes for samples with no preheating. The prediction achieves an acceptable value (error less than 0.5%) in about two hours. These results represent a significant improvement in measurement times when compared to the results from the conventional prediction procedures. It is anticipated that additional refinements to the prediction procedure will further improve the results.

This work was partially supported by the UKAEA Nuclear Materials Control Office.

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JRC Ispra Plutonium Assay Calorimeter - Measurements with PERLA PuO_2 Standards

Sample No.	Data File	Calculated Power (Watts)	Measured Power (after 3 Hrs) (Watts)	Predicted Power (after 3 Hrs) (Watts)	Error ΔP (%)	Final Predicted Power (after 5 or 6 Hrs) (Watts)	Error ΔP (%)	Sample Treatment
Pu113		15.44	14.866	15.4971	+ 0.32	15.4425	+ 0.02	no preheating (V1.5)
Pu113	30019001	15.511	14.06	15.574	+ 0.38	15.60	+ 0.58	1 hr preheat (V1.7)
Pu113	30019003	15.512	14.93	15.585	+ 0.44	15.565	+ 0.34	1 hr preheat, Al, (V1.7)
Pu113	01029001	15.513	14.79	15.473	- 0.25	15.512	0.00	no preheating, Al (V1.7)
Pu113	02029001	15.514	14.73	15.559	+ 0.29	15.523	+ 0.06	0.5 hr preheat
Pu113	05029001	15.516	14.77	15.582	+ 0.42	15.548	+ 0.20	1 hr preheat
Pu113	06029001	15.517	14.85	15.478	- 0.26	15.587	+ 0.45	2 hr preheat
Pu113	07029001	15.518	15.10	15.460	- 0.37	15.528	+ 0.06	3 hr preheat
Pu113	08029001	15.519	14.80	15.506	- 0.08	15.528	+ 0.06	0.25 hr preheat
Pu106	I23039001	1.566	1.512	1.578	+ 0.78	1.5615	- 0.28	1 hr preheat, canister top
Pu106	I22039001	1.566	1.458	1.571	+ 0.32	1.5706	+ 0.29	1 hr preheat, canister centre
Pu106	I21039001	1.566	1.496	1.554	- 0.77	1.5555	- 0.70	1 hr preheat, canister bottom
Pu111	31019002	5.872	4.556	5.753	- 2.0	5.859	- 0.22	1 hr preheat, brass packing
Pu111	I301101	5.845	5.20	5.863	+ 0.3	5.8398	- 0.08	
Pu115	I20039001	22.440	21.245	22.5307	+ 0.43	22.466	+ 0.11	Po without canister
Pu115	I19039001	22.438	21.07	22.466	+ 0.10	22.436	- 0.00	Po with canister
Pu120	ISP281102	37.96	36.48	38.04	+ 0.21	37.918	- 0.12	no preheating

$$\text{Error } \Delta P\ (\%) = \frac{(\text{Predicted Power}) - (\text{Calculated Power})}{(\text{Calculated Power})} \times 100$$

Table 1

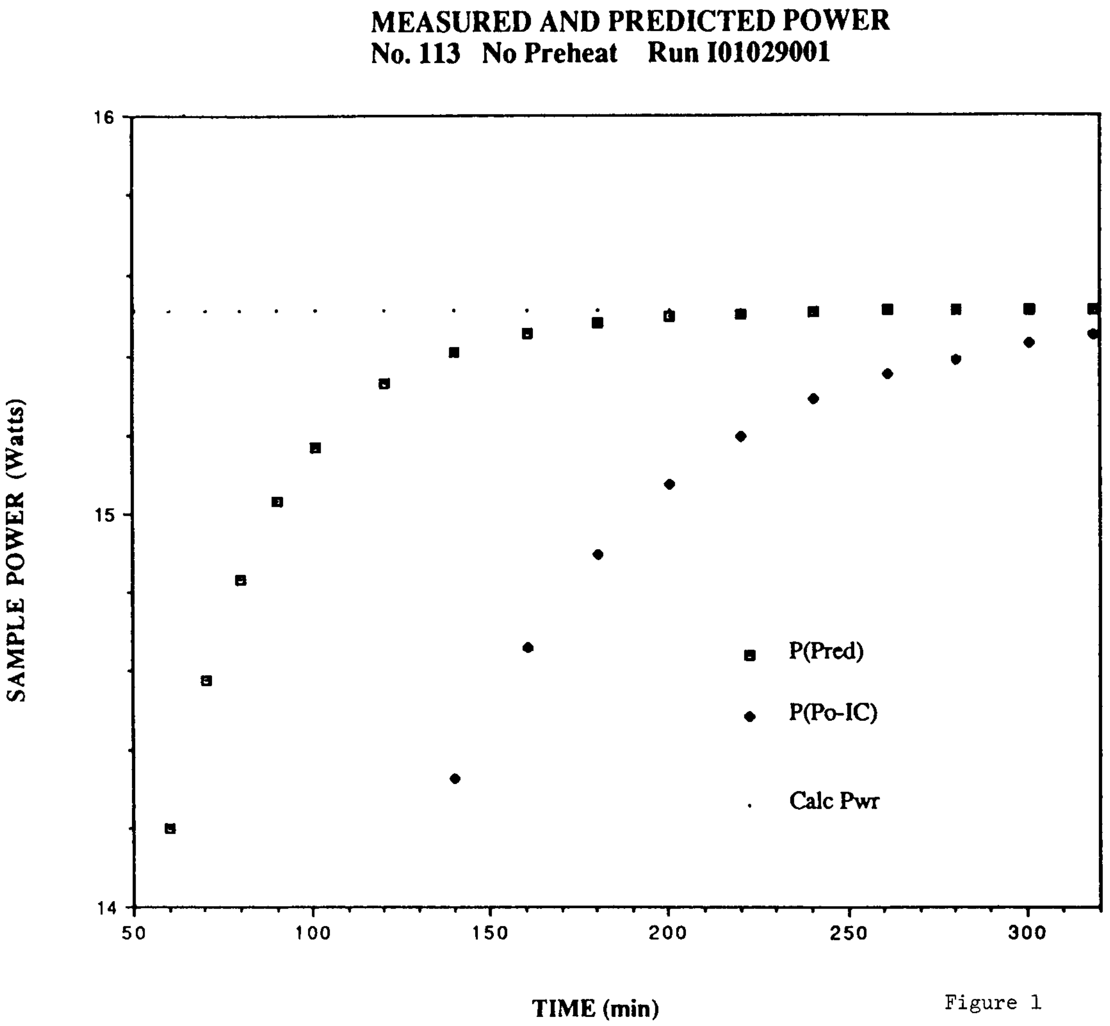

Figure 1

MEASUREMENT RESULTS USING EXPERT SYSTEM SOFTWARE

SAMPLE N°	END POWER (W)	FIRST GUESS		PRED. RESULT	
		TIME (min)	% ERR.	TIME (min)	% ERR.
A	15.731	69	2.1	138	0.2
B	15.723	71	2.9	108	0.3
C	15.724	81	1.1	112	0.2
D	15.723	56	4.9	109	0.2
E	15.723	80	1.5	124	0.5
F	15.715	69	2.0	121	0.2
G	15.726	71	3.6	96	0.9
HP2	53.653	46	0.6	82	0.1
IP1	57.542	44	0.8	83	0.2
JP1	59.880	36	3.4	81	0.3
KP2	58.481	40	3.7	73	0.4
L	39.010	46	0.3	94	0.3

TABLE 2

A REVIEW OF PLUTONIUM MEASUREMENT BY PASSIVE NEUTRON COINCIDENCE COUNTING

B W HOOTON
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Abstract

The paper presents a summary of a comprehensive review of the theory and practice of passive neutron coincidence counting for the determination of plutonium. It describes the methods used to extract the spontaneous fission component in a train of pulses from neutron detectors and it examines the latest methods used to correct for neutron multiplication effects. Examples of measurement stations and electronics are presented with procedures for their calibration and quality assurance. An assessment of errors under field conditions is also presented.

1. Introduction

For many years the determination of plutonium by passive neutron coincidence counting has been the basis for plutonium verification in international safeguards. It has been the cornerstone of the strategy for independent verification of the plutonium account. The early instruments were rather crude by today's standards, having a detection efficiency of perhaps 7% compared to 18% for instruments in routine use today and up to 50% for special systems. The coincidence method is well established but significant development work is still in progress and safeguards can probably look forward to an even better reliability of results as methods for a more detailed analysis of the data, using improved counting systems, come into routine use. The Review attempts to cover all aspects of the technique but since some readers will not wish to dwell on the mathematical derivations most of the underlying theory is given in Annexes.The Review covers the areas given below but since I will not have time to cover every aspect of the technique this presentation will concentrate on describing the state of the practice today and what further development of the technique can be expected in the near future. The Review does not cover active neutron methods.

The Contents of the Review are:

1. Introduction

2. Principles of the Coincidence Method

- **2.1 Spontaneous Fission**
- **2.2 A General Model**
- **2.3 The Variable Dead Time Counter**
- **2.4 The Shift Register**
- **2.5 Alpha/n Effects**
- **2.6 Moisture and Matrix Effects**

3. Neutron Multiplication Effects

- **3.1 Induced Fission**
- **3.2 Self-Multiplication Correction**
- **3.3 Monte Carlo Calculations**

4. Equipment

- **4.1 HLNCC-Type Counters**
- **4.2 Neutron Collar**
- **4.3 Other Systems**

5. Measurement of Plutonium

- **5.1 Quality Control and Calibration**
- **5.2 Measurement Strategy**
- **5.3 Measurement Uncertainties**
- **5.4 Waste Measurement**

6. Time Correlation Analysis

- **6.1 The Interpretational Model**
- **6.2 Electronic Pulse Train Analysis**
- **6.3 Time Correlation Examples**

Annex A Nuclear Data

Annex B Neutron Counters and Electronics

Annex C Shift Register Theory

Annex D Neutron Production by Alpha/n Reactions

Annex E Time Correlation Theory

References

The Review is intended to be a source document for anyone with a general interest in coincidence counting. It will include a wealth of basic nuclear data on the neutron emmission characteristics of nucleides, both by spontaneous and induced fission. It will also include data to facilitate the calculation of (α,n) yields in mixtures. There are so many results on measurements in the literature that it has not been possible to cover many, and the Review was not intended as a complete survey of the implementation. Nevertheless I hope it will cover sufficient to illustrate the chief principles behind measurements and a measurement strategy. In summarising the Review I have selected areas of interest to this audience. I will use a general model to lead into the shift register as the current state of the practice and then I will look at time correlation analysis since it may well come into more routine use as improved equipment, electronics and methods of analysis are developed.

2. General Model

The measurement techniques used to determine plutonium by coincidence counting uses equipment designed to analyse the pulse train produced by a set of neutron counters. The system components are illustrated in Fig 1.

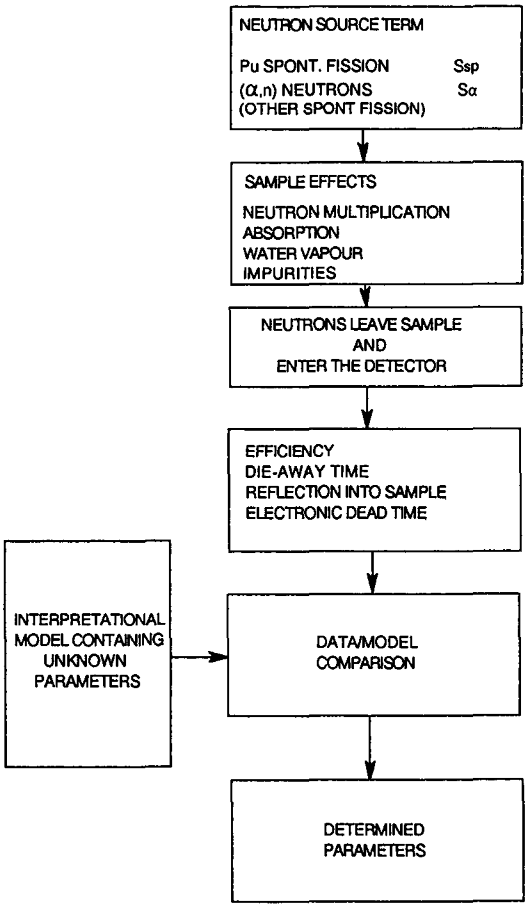
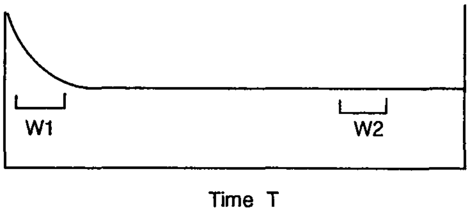


Fig 1
The General Model

The experimental data are collected and then compared with a theory based on an interpretational model. The model may be simple, as in the case of the shift register or it may involve a complex analysis of the pulse train using a more sophisticated interpretational model. In the case of the shift register the model is simple in its assumptions and easy to interpret. It assumes that the source is a mixture of random neutrons from a/n reactions and bursts of neutrons from spontaneous fission. The equipment is assumed to have a constant detection efficiency and a fixed neutron decay time. Under these circumstances the "coincidence" signal is proportional to the spontaneous fission strength of the sample and a simple calibration will yield the constant of proportionality.

Instruments in routine use today apply the shift register algorithm, which can be explained in terms of the time distribution of pulses shown in Fig 2.



A distribution function for the number of neutrons at a time T before the arrival time of each neutron

Random neutrons plus fission bursts

Fig 2.

A random pulse train has a constant distribution for the intensity of pulses in a period at any time T after a pulse has occurred; i.e. there are no correlations, but if fission occurs a small number of neutrons will be close together in time. Under these circumstances the distribution is greater for small time intervals. It has an exponential decay from time zero with a decay constant equal to the moderation and detection time for neutrons in the equipment. This distribution function is well known as the Rossi - Alpha distribution from the theory of reactor noise analysis.

The number of neutrons in a time interval dt at a time T after (or before) any neutron in a random pulse train is constant and given by the mean number of neutrons per second (R) multiplied by the time interval dt. The number measured in the time window w2 is:

$$N2 = R \, . \, W2$$

The number measured in the time window W1 is equal to the random rate plus the extra contribution from fission. The difference between the two areas is proportional to the fission rate even for large samples and, in the absence of neutron multiplication effects, it is a measure of Pu240(eff).

The shift register measures the extra contribution from fission by sampling the distribution in the window W1 and subtracting the background signal in W2. This is achieved using the electronics illustrated in fig 3.

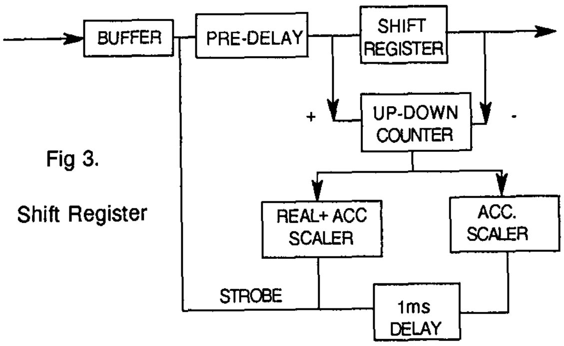


Fig 3.

Shift Register

Consider the shift register response to a burst of n neutrons, all within the gate width of the register W1. The first neutron to arrive will see zero in the gate, the second neutron will see the first in the gate, the third will see the first two and so on. The recorded signal is therefore:

$0 + 1 + 2 + 3 + \ldots (n-1)$; which equals $n(n-1)/2$.

This response is characteristic of the shift register. If two neutrons arrive the response equals 1 and the shift register is recording the number of pairs. If more than two neutrons arrive the shift register records, 3 for a burst of 3 and 6 for a burst of 4. The overall response is proportional to $\overline{n(n-1)}$, which is itself proportional to $\overline{\nu(\nu - 1)}$.

The quantity $\overline{\nu(\nu - 1)}$, is defined as:

$$\overline{\nu(\nu - 1)} = \sum_{\nu} \nu(\nu - 1)\, P(\nu)$$

where $P(\nu)$ is the probability for the emission of ν neutrons from fission. It is also, by definition, the second moment of the neutron multiplicity distribution

The quantity measured in the shift register $\overline{n(n-1)}$ is directly proportional to Pu240(eff) but this statement needs confirmation since the multiplicity of neutrons emitted by plutonium is not the same as the multiplicity of those detected. Consider the probability for the detection of i neutrons in a detection system of efficiency ε when ν neutrons are emitted by a source

$$\mathrm{Prob}(i,\nu,\varepsilon) = \varepsilon^{i} \binom{\nu}{i} (1 - \varepsilon)^{\nu - i}$$

Where $\binom{\nu}{i} = \frac{\nu!}{(\nu - i)!\, i!}$ (the binomial coefficient)

Since the number entering the detector is not constant and has a distribution with a mean value of $\bar{\nu}$ the probability of detecting i neutrons after fission is given by:

$$\mathrm{Prob}(i,\varepsilon) = \varepsilon^{i} \sum_{\nu = i}^{N} \binom{\nu}{i} (1 - \varepsilon)^{\nu - i} P(\nu)$$

The summation is up to N, the maximum number of neutrons emitted in a fission event.

When i neutrons are detected the shift register response is $i(i - 1)/2$ and the overall response is given by

$$R = \sum_{i=2}^{N} \frac{i(i-1)}{2} \,.\, \mathrm{Prob}(i,\varepsilon)$$

substituting for Prob (i,ε) then R can be shown to be equal to

$$R = \varepsilon^{2} \sum_{2}^{N} \frac{\nu(\nu - 1)}{2} P(\nu)$$

It is interesting to note that this complete response depends only on ε^2 even though it includes the effects of triple and higher order bursts.

This response must now be modified to take into account the electronics, which measures only a fraction of R depending on the time window W and its position. The experimental distribution is not perfect close to the origin since there is a limit on how close together two pulses can be. This is taken into account by having a delay time between the input pulse and the window W. The fraction of the area under the exponential measured by a gate width W and a pre-delay P is given by:

$$e^{-\frac{p}{\tau}} . (1 - e^{-\frac{W}{\tau}})$$

where p is the predelay time, typically 5 μs. If these parameters are constant then this fraction is constant and the shift register response is given by

$$R = \mathrm{Pu240(eff)} \,.\, \varepsilon^{2} \,.\, e^{-\frac{P}{\tau}} (1 - e^{-\frac{W}{\tau}}) \sum_{\nu}^{N} P(\nu) \frac{\nu(\nu - 1)}{2}$$

The quantity R is only proportional to the amount of plutonium in the absence of any perturbation to the stream of neutrons detected by the shift register. Absorption or multiplication effects will perturb the neutron leakage from the sample and any variations in detection efficiency or the die-away time of the detectors will also change the measured response. There are several ways for dealing with multiplication effects and for the special case where the (α,n) rate can be calculated and the efficiency and die-away time are constant the self-multiplication correction method of Ensslin /1/ can be applied.

I think it is fair to say that the current state of the practice in coincidence counting is based on the shift register approach with a calculated (α,n) contribution to deal with multiplication. In a high percentage of measurements this method can be used. It is unfortunate that it does not have a built in check to confirm the validity of the (α,n) calculation since if the calculation is in error it may escape un-noticed. There are some checks that can be used to give added confidence in the result, such as an examination of the multiplication parameter itself to see if it conforms to expectations /2/, but this requires some data on how the multiplication parameter M is expected to vary with plutonium mass.

The effect of moisture on the sample has been carefully examined in recent years /3/ and the most significant effect is on the (α,n) rate because of the added oxygen. The effect of hydrogen on the moderation and neutron multiplication is not large and in any case the multiplication, whatever its cause, is determined by the model. There is a minor effect due to the change in the energy spectrum of the neutrons but this is not the only way that the energy can change and the sample and its container will normally degrade the spectrum to a greater extent than the effect of moisture.

3. Further Development

The shift register produces only two pieces of information on the sample, the total count and the coincidence rate. In reality the system has many unknowns and the models must make assumptions about the system parameters to reduce the unknowns to a manageable level. The chief unknowns are:

Fission Source Intensity (Pu240(eff))	S_{sf} (or F_s)
Ratio of (α,n) to spont. fiss. neutrons	α (or S_α)
Efficiency	ε
Induced Fission	p (or M)
Die-away Time	λ

The efficiency is a variable since it is a function of neutron energy. The energy of the neutrons entering the detection region may vary a little with isotopic composition, the (α,n) component and multiplication but the biggest perturbation comes from the sample size and composition. Most of the secondary interactions in the sample, e.g. induced fission, take place quickly at epithermal energies and there is often ample justification for treating the die-away parameter as a constant. Nevertheless the system would still contain four unknowns.

A method for dealing with more unknowns was suggested by Bondar /4/ at the 1982 IAEA Technical Symposium. It was based on a detailed analysis of the pulse train to obtain more than two pieces of information. The approach is generally refered to as Time Correlation Analysis and the shift register can be viewed as a rather simple example of the time correlation analysis of the pulse train. The approach was pursued by workers at Ispra with parallel development of the electronics and the interpretational models. Hage /5/, /6/ has produced a formal theoretical treatment which enables the method to be applied on a sound foundation. Let me explain the approach and discuss its limitations.

The instrumental basis of the Euratom Time Correlation Analyser is the opening of a gate of length g either at a regular period (Pulse Fluctuation Analysis) or by each pulse in the pulse train (Pulse to Pulse Analysis) and thereafter the counting of the number of pulses, k, which appear within the gate length. This leads to a count rate distribution R_{kg}. A typical distribution is shown in Fig 4. /7/ which also shows how critical the dead time corrections can be to the detailed shape of the distribution This distribution clearly has a lot of information on the pulse train but the usefullness of the approach depends on how sensitive the data are to the independent unknowns.

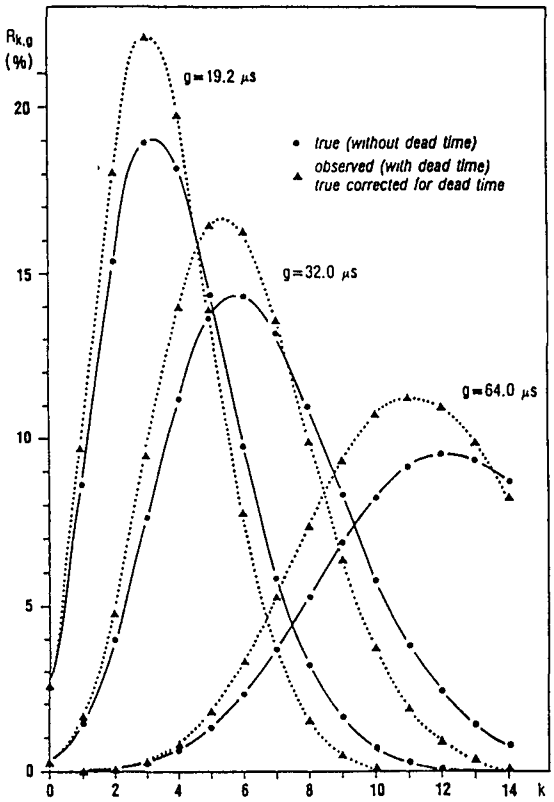


Fig 4.
Distributions from Pulse to Pulse Analysis

An interpretational model which may be used to determine the unknowns from the data has been formulated by Bondar /4/. The treatment is general and can be viewed as in two stages. In the first instance the problem is defined in terms of the intensities of singlets, doublets , triplets etc. originating from a single random event, either spontaneous fission of alpha/n. These are designated C_1, C_2, C_3, etc. and the only other unknown necessary to define the pulse train is the die-away time λ. There is no need to consider the efficiency since the parameters C_i relate to detected groups.

This specification of the pulse train is complete and it leads to a theoretical expression for $R_{k,g}$ in terms of the values of C_i and λ.

$$R_{k,g} = F(C_1, C_2, C_3, \dots, \lambda)$$

The precise form of this function is given in ref /4/.

The next stage is to use a model to derive equations for the C_i components in terms of the amount of Pu240(eff), the alpha/n source strength, the efficiency of the detection system and the neutron multiplication parameter p. The equations can be expressed by adding the individual contributions to each group, as follows:

$$\text{Singles} = C1 = S_{sp}.P(1,\varepsilon,p) + pS_\alpha.\ Q(1,\varepsilon,p) + \varepsilon\ (1-p)S_\alpha$$

$$\text{Doubles} = C2 = S_{sp}\ .\ P(2,\varepsilon,p) + pS_\alpha\ .\ Q(2,\varepsilon,p)$$

$$\text{Triples} = C3 = S_{sp}\ .\ P(3,\varepsilon,p) + pS_\alpha\ .\ Q(3,\varepsilon,p)$$

where S_{sp} and S_α are the spontaneous fission and alpha/n event intensities, ε is the detection efficiency, $P(i,\varepsilon,p)$ is the probability for the detection of i neutrons when induced fission takes place with a probability p, and $Q(i,\varepsilon,p)$ is the probability for the detection of i neutrons from the alpha/n source with the same induced fission probability p. The values of $P(i,\varepsilon,p)$ and $Q(i,\varepsilon,p)$ are calculated using

$$P(i,\varepsilon,p) = \sum_{\nu \geq n} \frac{n!}{\nu!\ (\nu - n)!}\ \varepsilon^n\ (1-\varepsilon)^{\nu - n} P_\nu(p)$$

where $P_\nu(p)$ is the probability that n neutrons are emitted from a fission event with an induced fission probability of p. The procedure used to analyse data is to determine the unknowns S_{sf}, S_α, ε, and p by a least square fit of the $R_{k,g}$ data to the theoretical predictions. It can be seen from fig 4. that there are sufficient degrees of freedom to carry out a minimum χ^2. In practice the sensitive to p may be small and there may be a large error on the source terms. The accuracy of the measurement improves as the efficiency increases to give a greater contribution from the triples and higher order terms.

An alternative treatment by Cifarreli and Hage /5/ is based on the moments of the R_{kg} distribution, defined as:

$$m_{R(\mu)} = \sum_{k=\mu}^{\infty} \binom{k}{\mu} R_{k,g}$$

These moments are calculated from the measured data R_{kg}. They are then used to used to obtain values for the correlated singlets, doublets and triplets in the pulse train which, using the notation of Cifarelli and Hage /5/ are expressable as:

$$R_1 = \varepsilon\ F_s\, M\ \nu_{s(1)}\ (1 + \alpha)$$

$$R_2 = \varepsilon^2\ F_s\ M^2 \left[\ \nu_{s(2)} + (M-1)\ \frac{\nu_{s(1)}\ \nu_{I(2)}}{\nu_{I(1)} - 1}\ (1+\alpha)\right]$$

$$R_3 = \varepsilon^3\ F_s\ M^3$$

$$\left[\ \nu_{s(3)} + 2\ (M-1)\ \frac{\nu_{s(1)}\ \nu_{I(2)}}{\nu_{I(1)} - 1} + (1+\alpha)(M-1) \dots\right.$$

$$\left. \dots \frac{\nu_{s(1)}}{\nu_{I(1)} - 1} \left\{ \nu_{I(3)} + 2\ (M-1)\ \frac{\nu_{s(1)}\ \nu_{I(2)}}{\nu_{I(1)} - 1} \right\}\right]$$

The R_1, R_2 and R_3 values, above, have been obtained from the experimental data R_{kg} and they include the effect of the die-away time, the gate width and the predelay in the electronics. The expressions on the right hand side of each equation are the result of an interpretational model and they have been derived with the following assumptions:

1. Instantaneous neutron multiplication
2. A constant probability for induced fission
3. A pure exponential die-away time $1/\lambda$
4. A single value detection probability

The nuclear data constants in the equations are the moments of order μ for the neutron distributions associated with spontaneous fission, $\nu_{s(\mu)}$, and induced fission $\nu_{I(\mu)}$. Within the constraints of the model there still remains the practical limitations of dead time corrections and the sensitivity of the results to each unknown.

In practice the method is limited to three unknowns, at best, since the accuracy on triples is already poor and there seems to be little point in attempting to measure higher moments. The effects of dead time are critical at high counting rates and although the technique has had significant success with waste, by treating the efficiency as a variable, the application to high mass samples will only be established with the implementation of high quality dead time corrections.

In summary, although the determination of plutonium by passive neutron coincidence counting is subject to some minor problems of moisture and difficulties if the (α,n) contribution is unknown it is in a healthy state for most samples. The shift register method is well developed and on a sound foundation. However, if further development takes place it can be expected to be an implementation of the more sophisticated time correlation analysis models. These will require careful attention to dead time losses but the method has a special merit for waste measurement if the detection efficiency varies betwen samples. It may also prove to be usefull for large samples if the equipment has a high efficiency and the electronics can accurately process a high count rate.

5. References

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NEUTRON MULTIPLICITY COUNTER DEVELOPMENT*

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Abstract

We have designed and built two multiplicity counters to test the usefulness of multiplicity counting for the safeguards assay of plutonium-bearing materials. The first prototype counter has been characterized and a variety of plutonium-bearing materials have been measured with it. Assays accurate to ~0.7% have been obtained for both pure and impure plutonium oxide samples in reasonable measurement times. Assays accurate to ~5% have been obtained for metal samples. A second multiplicity counter has been designed using experience gained from the first as well as Monte Carlo simulations. The second counter was designed to be more suitable for in-plant measurement of pyrochemical process materials. This paper presents the results of characterization studies of the two instruments.

1. Introduction

A conventional passive neutron coincidence assay of plutonium measures the first and second moments of the neutron multiplicity distribution of a sample and deduces from them the sample's fertile mass. The problem with this technique is that there are actually three variables in the measurement problem: the sample mass, the (α,n) neutron rate of the sample, and the multiplication of neutrons by the sample. Thus, conventional coincidence techniques only provide accurate assays for pure materials; for samples whose impurities are known well enough to compute the ratio, α, of their (α,n) neutron rate to their spontaneous-fission rate; or for impure samples whose self-multiplication is known /1/.

The goal of neutron multiplicity counting is to obtain a third quantity from the multiplicity distributions and to deduce all three variables from measured quantities. Our first assays using multiplicity counting and a conventional thermal-neutron detector were obtained from measurements performed in a modified, active-well counter using specialized shift-register electronics designed and built just for the purpose /2/. These assays were accurate to ~2% (1σ) for a variety of pure and impure materials. However, very long measurement times were necessary to obtain this accuracy for large samples with an α greater than about 0.5 because of the large statistical error in the measured triples to doubles ratio.

Assays of this accuracy can be obtained in more reasonable measurement times if the neutron counter is designed to have a very high detection efficiency relative to most thermal-neutron counters used for safeguards measurements and a low die-away time. Short electronic deadtime is also essential if accurate deadtime corrections are to be performed. This paper reports on two thermal-neutron counters that have been designed and built at Los Alamos to achieve the goal of accurate multiplicity assays in reasonable measurement times. The first prototype counter was built to study the effects of counter design on multiplicity assay. It was designed to be used with multiplicity-sorting electronics that could sort multiplicities up to seven and a moment analysis technique that greatly reduces the measurement times needed to obtain good statistical precision for large samples. Using the experience gained from this first counter, we designed and built a second counter to be more applicable to facility measurements.

2. The Dual-Mode Multiplicity Counter

The first multiplicity counter consists of 130, 50.8-cm-active-length ^{3}He tubes. A schematic diagram of this counter is shown in Fig. 1. In the counter's initial configuration, each of the ^{3}He tubes was wrapped in a 0.7-cm-thick sleeve of high-density polyethylene followed by an outer, 0.081-cm-thick sleeve of cadmium. Each wrapped tube was then placed in the aluminum core as shown in the diagram. From the Monte Carlo design calculations we expected this configuration of the counter to have a low detection efficiency but a very low die-away time. The short die-away time was essential so that short coincidence-gate widths could be used without loss of counting precision. Short gate widths were necessary so that the multiplicity circuit would not overflow for large samples. The counter was called the dual-mode counter because it was also designed to operate in a high-efficiency mode with the cadmium sleeves removed.

Early characterization studies of the counter in its low-efficiency mode (cadmium sleeves in place) revealed a detection efficiency of 17% for a ^{252}Cf source placed in the center of its 16.5-cm-wide by 25.4-cm-high sample chamber and a

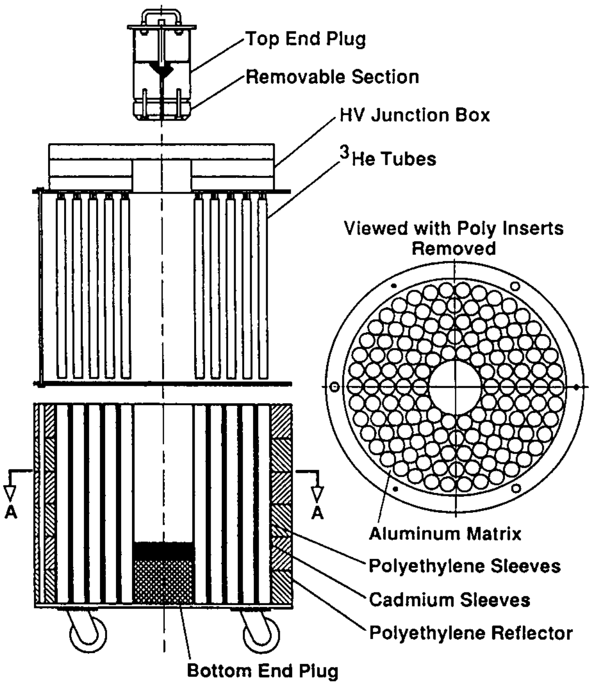

Fig. 1. Schematic of the dual-mode multiplicity counter.

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die-away time of 11.8 μs. In the high-efficiency mode, the measured detection efficiency was 53% for ^{252}Cf and its die-away time was 57 μs.

Using assay variance as a figure-of-merit for multiplicity counters, Ensslin et al. /3/ have written a program that predicts the statistical accuracy of a multiplicity assay as a function of a detector's efficiency and its die-away time; a sample's effective ^{240}Pu content, self-multiplication, and (α,n) neutron rate; and the measurement count time. They have calculated the precision of assays performed by this prototype counter for samples whose α is less than 5 and whose ^{240}Pu-effective content is between 7.6 and 143 g. These calculations predicted that the dual-mode counter will perform more accurate assays in the high-efficiency mode than in the low-efficiency mode for samples of this type. In the high-efficiency mode, however, the shift-register multiplicity circuit had to be upgraded to handle higher multiplicities. In Ref. 3, Ensslin et al. report on measurements made with this counter in the low-efficiency mode using the original multiplicity circuit that could sort multiplicities up to seven. Measurements made in the high-efficiency mode with a new multiplicity circuit capable of sorting multiplicities up to 32 are reported here.

Figures 2 and 3 show the spatial response of the dual-mode counter in the high-efficiency mode measured with a ^{252}Cf source centered in the sample chamber and moved axially and radially. This detector's response is uniform in the central region of the cavity, decreases near the end plugs, and increases near the cavity's outer walls.

Eight pure plutonium oxide samples, seven impure oxide samples, and eight plutonium metal samples were measured using the new multiplicity electronics package and the dual-mode counter in the high-efficiency mode. For the oxide samples, the predelay was set at 3 μs and the coincidence gate width was 32 μs. For the metal samples, the predelay was also 3 μs, but the gate width had to be decreased to 12 μs to keep the multiplicity circuit's sorting register from overflowing. All samples were centered in the sample cavity where the detector's response was most uniform. The oxide samples were measured for 5000 s each; the metal samples were measured for 1000 s each.

Because the multiplicities counted with the new circuit ranged from 1 to 32 for these samples and available deadtime correction schemes could not provide accurate correction for multiplicity distributions this large, a new scheme was derived so that measurements from the new circuit could be properly corrected. This new technique by Dytlewski is described in Ref. 4. All assays were based on measured multiplicity distributions corrected using this approach. The analysis of the corrected multiplicity distributions to yield assay results was based on the emitted moments calculated by Boehnel /5/ and Cifarelli and Hage /6/. The calibration parameters were the detector's efficiency for a nonmultiplying, pure plutonium oxide sample; gate fractions derived from the die-away response to ^{252}Cf; and known values for the first, second, and third induced-fission moments for ^{239}Pu at 2 MeV.

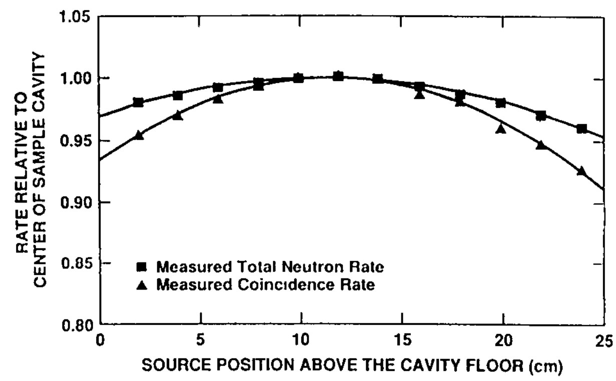

Fig. 2. Axial response to a ^{252}Cf source of the dual-mode multiplicity counter in the high-efficiency mode.

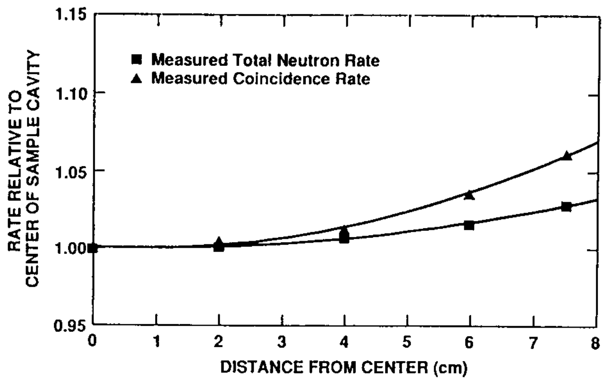

Fig. 3. Radial response to a ^{252}Cf source of the dual-mode multiplicity counter in the high-efficiency mode.

The multiplicity assay results for the 23 samples are given in Table I. Figures 4 and 5 give the assay results derived as above for these samples relative to their known reference values. Included in these plots, for comparison, are the assay results obtained using conventional passive, neutron coincidence assay methods. For the oxide samples, the conventional two-parameter assay analysis included a multiplication correction based on a value for α calculated from the known isotopic ratios for the samples. For the metal samples, no multiplication correction was performed. The calibration parameters for the conventional assays were derived from the characterization with ^{252}Cf and a relationship derived for the pure oxides between the multiplication-corrected coincidence rate and the known ^{240}Pu-effective mass.

Table II gives a comparison of the overall assay results obtained for these samples with the multiplicity approach to the conventional assay results. The improvement attained with the multiplicity analysis is striking. The results of the multiplicity assay compared to reference values for the pure and impure oxide samples display no statistically significant bias, and the scatter in the results is consistent with the expected uncertainty in the reference values and the statistical precision predicted by Ensslin et al. /3/ A more recent effort has been made to predict the statistical precision of multiplicity assays from measured quantities. The results of this effort are reported in Ref. 7.

The metal sample results display a bias and their scatter is slightly larger than expected. Although the reason for these discrepancies is not yet completely understood, several factors have been identified that may be responsible for them. One factor is that the reference values for three of the samples were suspect. These samples are starred in Table I. However, removing the results for these samples from the calculation of the average and standard deviation of the assays only improves the latter. The bias remains about the same.

Several factors may contribute to this bias. One factor is that the die-away response for this counter was not exponential. Figure 6 displays the measured die-away response for this counter for ^{252}Cf with an exponential fit overlayed. At early times the die-away is faster; at later times there is a

TABLE I. Multiplicity Assay Results Obtained with the Dual-Mode Multiplicity Counter in the High-Efficiency Mode

Sample Type	Total Pu (g)	^{240}Pu-effective (g)	M	Alpha	Multiplicity Assay/Reference
Pure oxide	59.97	10.10	1.025	0.436	0.993
Pure oxide	171.9	29.29	1.038	0.426	1.001
Pure oxide	321.9	54.33	1.050	0.418	1.009
Pure oxide	384.8	65.10	1.056	0.416	1.002
Pure oxide	543.8	92.14	1.070	0.416	1.010
Pure oxide	612.7	104.5	1.078	0.418	1.013
Pure oxide	848.6	144.3	1.097	0.420	1.006
Pure oxide	876.6	149.1	1.084	0.413	1.011
Impure oxide	19.97	1.977	1.014	0.807	1.000
Impure oxide	74.89	7.414	1.022	0.766	1.005
Impure oxide	149.8	14.83	1.031	0.766	0.999
Impure oxide	299.6	29.66	1.043	0.747	1.014
Impure oxide	458.4	43.33	1.057	0.705	1.005
Impure oxide	614.0	64.95	1.063	0.710	0.998
Impure oxide	777.7	81.31	1.074	0.649	1.014
Metal	585	34.46	1.208	0.858	0.836
Metal	1000	57.66	1.590	0.024	0.936
Metal	1352[a]	81.66	1.119	0.847	1.021
Metal	1678[a]	98.33	1.175	0.682	0.830
Metal	1729[a]	101.7	1.410	0.198	0.912
Metal	2000	115.3	2.070	0.113	0.845
Metal	2200	130.8	1.611	0.086	0.898
Metal	2962	141.3	2.086	0.112	0.873

[a]Reference values may not be accurate

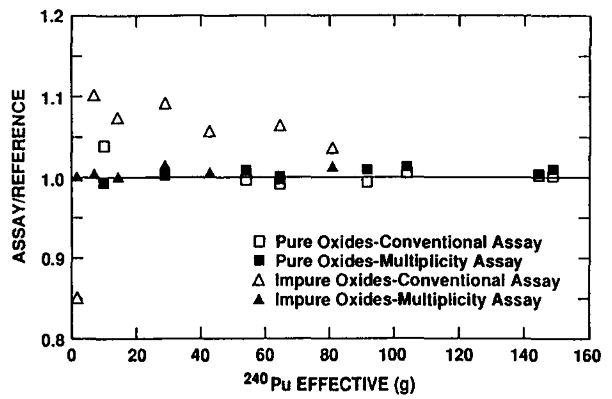

Fig. 4. A comparison of the multiplicity assay results obtained with the dual-mode multiplicity counter in the high-efficiency mode to conventional assay results for plutonium oxide samples.

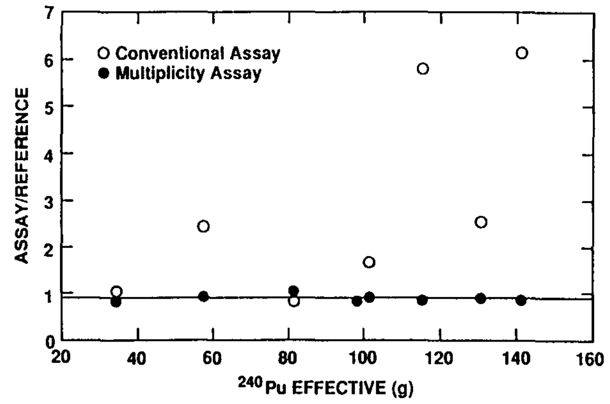

Fig. 5. A comparison of the multiplicity assay results obtained with the dual-mode multiplicity counter in the high-efficiency mode to conventional assay results for plutonium metal samples.

TABLE II. Comparison of the Overall Multiplicity Assay Results Obtained with the Dual-Mode Multiplicity Counter in the High-Efficiency Mode to Conventional Assay Results

Average Assay/Reference with Standard Deviation		
Sample Type	Conventional Assay Results	Multiplicity Assay Results
Pure plutonium oxides	1.004 ± 1.4%	1.006 ± 0.66%
Impure plutonium oxides	1.039 ± 8.2%	1.005 ± 0.68%
Plutonium metal	2.665 ± 81%	0.894 ± 7.1%
Plutonium metal (subset) using relative calibration		0.975 ± 4.9%

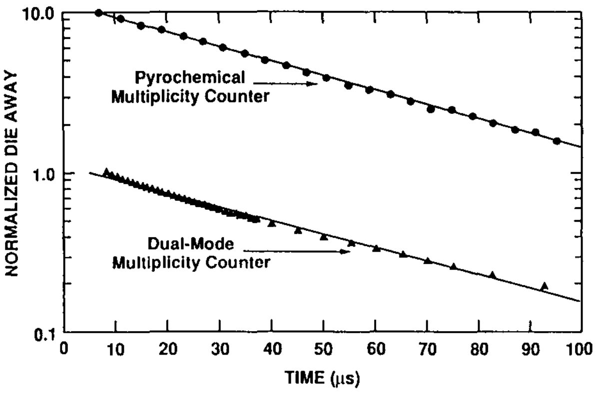

Fig. 6. The die-away response of the dual-mode multiplicity counter in the high-efficiency mode and the pyrochemical multiplicity counter for ^{252}Cf. The former is normalized to 1; the latter is normalized to 10.

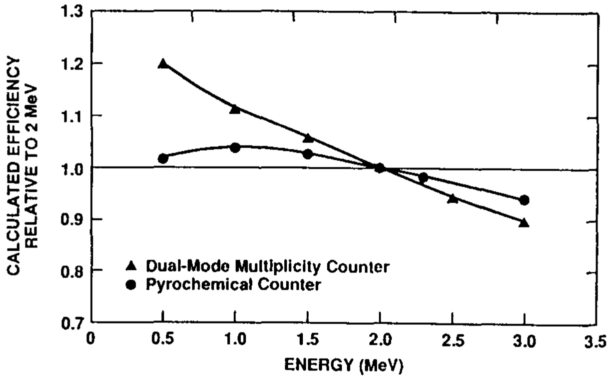

Fig. 7. The calculated efficiency of the dual-mode multiplicity counter in the high-efficiency mode and the pyrochemical multiplicity counter for a monoenergetic point neutron source in the center of the sample chamber.

tail. This behavior complicates the determination of the effective die-away time for the counter and the subsequent determination of the gate fractions needed to analyze the detected multiplicity distributions. The oxide assays are not as sensitive to errors that may result as a consequence of this complex die-away behavior because their third moment is small (that is, their self-multiplication is small). However, the metal assays are much more sensitive to such errors, because their third moments are substantially higher relative to their first and second moments.

Another possible source of the bias is that the average energy of the neutrons emitted by the metal samples is probably slightly different from that for the oxide samples. The multiplicity assay calibration was based on a small plutonium oxide source. Spontaneous-fission neutrons from ^{240}Pu have a mean energy of ~2.0 MeV. Induced-fission neutrons have a mean energy of ~2.2 MeV; (α,n) neutrons from oxygen have a mean energy of ~1.8 MeV. The average energy of the neutrons emitted by these samples will be a complex combination of these neutron energy spectra and spectra resulting from (α,n) reactions with any other impurities. The calculated energy response of the dual-mode counter in the high-efficiency mode relative to 2 MeV is given in Fig. 7. These calculations were made with MCNP version 3B /8/ for a monoenergetic point source placed in the center of the sample chamber. From these calculations, if the mean energy of the neutrons emitted by the metal samples is larger on average than for the oxide samples, the detection efficiency of the dual-mode counter will be lower and the assay will be biased low. A multiplicity analysis will be especially sensitive to such effects because the detected third moment depends on the third power of the detection efficiency.

The metal assays were re-evaluated, for the subset of samples whose reference values were believed to be accurate, using a relative calibration based on a small metal sample whose multiplication was known. Using this relative calibration approach instead of a calibration based on plutonium oxide substantially reduces the bias, which suggests that the bias is indeed caused by an effect related to the general sample composition. The overall results for this relative calibration method are included in Table II. More study is needed to understand the effect of sample composition on multiplicity assays.

3. The Pyrochemical Multiplicity Counter

The assay results discussed above suggest that not only does a multiplicity counter need to have a high efficiency and a low die-away time, it should also have a detection efficiency that does not vary with the emitted neutron energy spectrum and a well-behaved, exponential die-away response. The dual-mode counter was designed as a research tool and was not optimized in this sense. Also, its sample cavity is too small to accommodate the large range of container types that may be encountered in facilities.

A second counter has been designed and built at Los Alamos to be more suited to in-plant use and to optimize, as much as reasonably possible, the parameters that have thus far been identified as important in multiplicity assays by thermal-neutron detectors. We determined the optimum arrangement of tubes in the body of the counter and the best choice for end-plug materials to yield a high and invariant detection efficiency by using Monte Carlo simulations. These calculations are reported in detail in Ref. 9. The body of the counter was constructed only from polyethylene to simplify its die-away response. In Figure 7 we compare the calculated energy response of this new counter to that of the dual-mode counter.

The new counter, named the pyrochemical multiplicity counter, consists of 126 ^{3}He tubes with 71-cm active lengths arranged in four rows. Figure 8 gives a schematic diagram of this new counter as it was conceived after the calculations. The cadmium-lined sample cavity is 24 cm in diameter and 38 cm high, and its efficiency profile was optimized for cans up to 20 cm wide and 36 cm high. The end plugs are made of graphite. To allow the counter to be used for both at-line and in-line applications, it was built in two identical halves. To accommodate the physical separation of the halves, one tube from the third ring was moved to the outermost ring and the tubes in each ring were moved together slightly to allow for the split in the junction box. The calculations were not repeated for this new configuration because we did not expect it to significantly affect the results. Each half in the final counter then contains 63 tubes. The halves can be brought together to form a single free-standing counter or can be split and then placed around a glove-box well for in-line applications.

The calculations predicted that the counter would have an average detection efficiency of 57.0%, an exponential die-away behavior, and a die-away time of 49.4 µs. The measured efficiency of the counter is 57.7% for ^{252}Cf and its measured die-away time is 47.2 µs. The differences between the calculations and the measured quantities are consistent with the above-mentioned design changes. The measured die-away response of the pyrochemical multiplicity counter is given in Fig. 6 along with an exponential fit. These data do not display the nonexponential behavior of the dual-mode counter.

Figures 9 and 10 show the spatial response of the pyrochemical counter measured with a ^{252}Cf source centered in the sample chamber and moved axially and radially. Several of the calculated points are overlayed for comparison. This detector's response is more uniform in the central and lower regions of the cavity than the dual-mode counter and increases less near the cavity's outer walls. The agreement between the calculations and measurements suggests that the Monte Carlo model may also be useful to understand sample-related variations in the detector response that cannot practically be ascertained any other way.

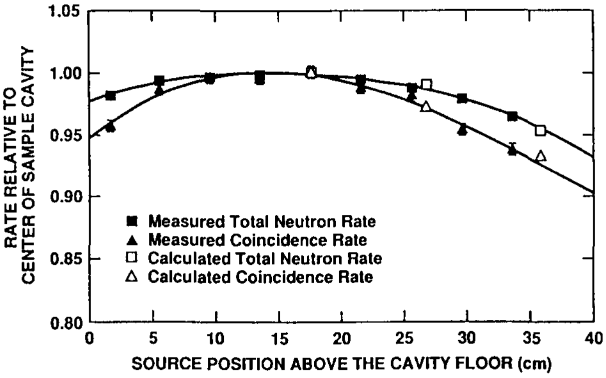

Fig. 9. Axial response of the pyrochemical multiplicity counter to a ^{252}Cf source.

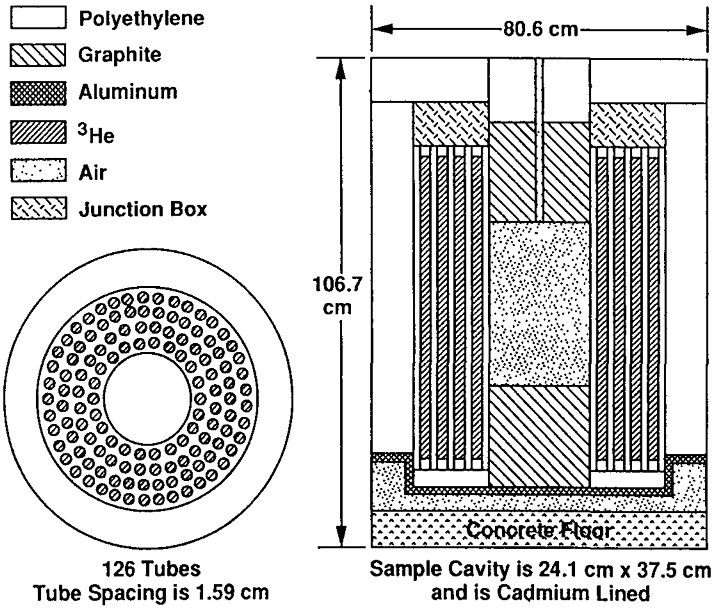

Fig. 8. Schematic of the pyrochemical multiplicity counter.

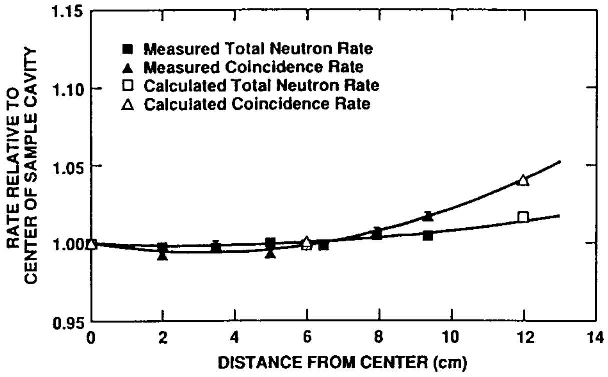

Fig. 10. Radial response of the pyrochemical multiplicity counter to a ^{252}Cf source.

4. Future Development

Although these early characterization studies demonstrate that the goals identified for the new counter from the lessons learned from the dual-mode counter have been achieved, actual sample measurements need to be performed in it. Some preliminary assays of very small samples have been successful, but measurements of more characteristic samples have been postponed until a new multiplicity electronics package is completed. This new package will allow for sorting multiplicities up to 256. Thus, larger coincidence gate widths can be used and the precision of the results for high-rate samples will improve. Also any effects as a result of fluctuations in the die-away response caused by the nature of the sample should be reduced by using longer gate widths.

Because it is probably not possible to build a thermal-neutron detector in which the detection efficiency does not vary, future measurements will include an analysis of correlations between sample size and composition and the relative counting rates of the individual rings of the pyrochemical counter. The effect of reducing the number of rings in a multiplicity counter will also be studied to determine if a multiplicity counter, which is smaller and less costly but perhaps is limited to a smaller range of sample types, is practical.

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SIMPLE AND EFFECTIVE METHOD OF DETERMINING MULTIPLICITY DISTRIBUTION LAW OF NEUTRONS EMITTED BY FISSIONABLE MATERIAL WITH SIGNIFICANT SELF - MULTIPLICATION EFFECT

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1. Introduction

At developing new methods of non-destructive determination of plutonium full mass in nuclear materials and products being involved in uranium - plutonium fuel cycle by its intrinsic neutron radiation, it may be useful to know not only separate moments /1/ but the multiplicity distribution law itself of neutron leaving this material surface using the following as parameters
- firstly, unconditional multiplicity distribution laws of neutrons formed in spontaneous and induced fission acts of the given fissionable material corresponding nuclei and unconditional multiplicity distribution law of neutrons caused by (α,n) reactions at light nuclei of some elements which compose this material chemical structure;
- secondly, probability of induced fission of this material nuclei by an incident neutron of any nature formed during the previous fissions or (α,n) reactions.

An attempt to develop similar theory has been undertaken, for example, in /2/. Here the author proposes his approach to this problem.

The main advantage of this approach, to our mind, consists in its mathematical simplicity and easy realization at the computer. In principle, the given model guarantees any good accuracy at any real value of induced fission probability without limitations dealing with physico-chemical composition of nuclear material.

2. Theory

Preliminarily let us make some comments and give definitions.

Primary neutrons - neutrons born in spontaneous fission acts of fissionable material nuclei or in (α,n) reaction acts at light element nuclei being part of its composition.

Secondary neutrons of the first generation - neutrons born in induced fission acts of this material corresponding nuclei from the part of primary neutrons.

Secondary neutrons of the second generation - neutrons born in result of induced fissions of material nuclei from the part of secondary neutrons of the first generation.

Secondary neutrons of the third generation - neutrons born by the part of neutrons of the second generation, etc.*

Number of primary or secondary neutrons formed in the corresponding single production act is a random value which distribution law is called an unconditional multiplicity distribution law of primary or secondary neutrons.

From the point of view of processes of born primary and secondary neutrons interaction with the fissionable material nuclei, only two of them are significant: processes of fission and scattering. The influence of the rest interaction processes is neglected.

We consider that "prehistory" of any secondary neutron origin has no influence on its ability to cause induced fission of the fissionable material either nucleus. This ability is quantitatively described by so-called induced fission probability p.

The specified unconditional multiplicity distribution laws of neutrons and induced fission probability are determined a priori for examined object with nuclear material and are included into the proposed model as constants.

If fissionable material contains uranium in its composition, from the point of view of its specific activity, it is considered to be stable in comparison with plutonium.

Thus the mathematical idea of the method consists in the following (see Fig.1): if induced fission from primary neutron is absent in principle (K=0 in Fig.1), "random" value which is a number of neutrons leaving this material surface for this case is $J^{(0)}=1$.

In case of strict assumption that this primary neutron gives only one induced fission (K=1), random value $J^{(1)}$ of neutrons number passing through this material surface for this case is (see also Fig.1):

$$J^{(1)} = X + (1-X)\sum_{M} J^{(0)} \qquad (1)$$

where X - random value, taking value "1" if induced fission from primary neutron is absent and "0"- in opposite case (so-called characteristic random value of absence of induced fission at primary neutron);

M - random value equal to number of secondary neutrons formed in this induced fission act.

In other words, random value $J^{(1)}$ with probability (1-p) is equal to 1 and with probability p it equals to sum of random number of M units (independent "random" values $J^{(0)}$).

Now let us assume that this primary neutron produces sctrictly two induced fissions (K=2). Then random value $J^{(2)}$ of neutrons number passing through the fissionable material surface for this case is :

$$J^{(2)} = X + (1-X)\sum_{M} J^{(1)} \qquad (2)$$

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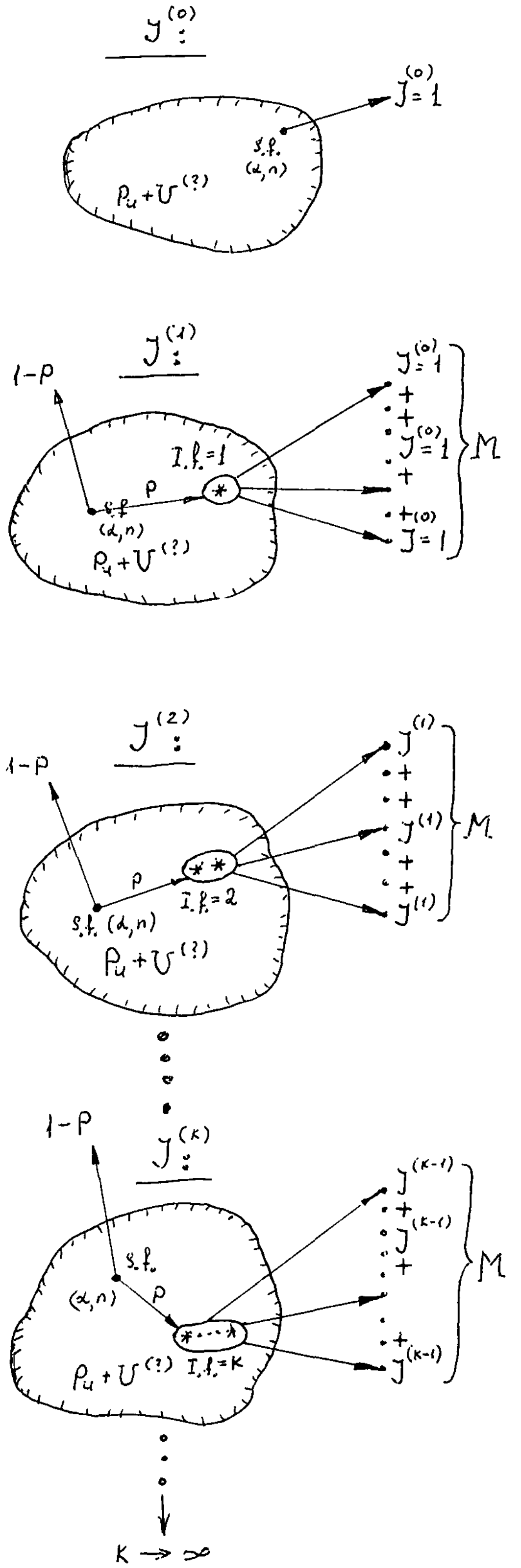
* If we are speaking about secondary neutrons in general, we mean their combination over all the generations.



S.f. (α,n) - primary neutron production act
I.f. - induced fission act
fig. 1

because each of M secondary neutrons of the first generation forms a situation described above for K=1 (see Fig.1).

Extending the similar considerations, it can be stated that for a chain of K induced fissions ("K" generations of secondary neutrons) random value of neutrons number passing through the material surface is :

$$J^{(K)} = X + (1-X)\sum_{M} J^{(K-1)} \qquad (3)$$

$$K = 1, 2, \dots$$

Now let us determine distribution law $W^{(K)}_{(j)}$ for this random value.

First of all, it is obvious that "random" value $J^{(0)}$ has distribution law $W^{(0)}_{(j)} = \delta^{j,1}$, where $\delta^{j,1}$ - Kronecker symbol. Further let us note that distribution law of finite sum of independent random values can be described by means of successive usage of mathematical convolution operations (the distribution law convolution of the random summands first pair is calculated, then the convolution of this result with the third summand distribution law is determined, etc.). From this it follows that conditional distribution law $W^{(1)}_{(j|m)}$ of fixed number m sum of independent "random" values $J^{(0)}$, having, as specified, distribution laws $W^{(0)} = \delta^{j,1}$ is:

$$W^{(1)}_{(j|m)} = \underbrace{\left\{\left[W^{(0)}_{(j)} * W^{(0)}_{(j)}\right] * W^{(0)}_{(j)}\right\} * \dots}_{(m-1)\ times} = \delta^{j,m} \qquad (4)$$

where * - symbol designating convolution operation

With regard for (4) random value $J^{(1)}$ distribution law $W^{(1)}_{(j)}$ is equal to :

$$W^{(1)}_{(j)} = \sum_{m} P g(m) W^{(1)}_{(j|m)} + (1-P)\delta^{j,1} = \sum_{m} P g(m) \delta^{j,m} + (1-P)\delta^{j,1} \qquad (5)$$

where $g(m)$ - random value M unconditional distribution law of neutrons number born in induced fission act (so-called unconditional distribution law of secondary neutron multiplicity).

Obviously, distribution law $W^{(2)}_{(j)}$ of random value $J^{(2)}$ is:

$$W^{(2)}_{(j)} = \sum_{m} P g(m) W^{(2)}_{(j|m)} + (1-P)\delta^{j,1} \qquad (6)$$

where conditional distribution law $W^{(2)}_{(j|m)}$ is equal to:

$$W^{(2)}_{(j|m)} = \underbrace{\left\{\left[W^{(1)}_{(j)} * W^{(1)}_{(j)}\right] * W^{(1)}_{(j)}\right\}}_{(m-1)\ times} * \dots \qquad (6')$$

Thus, for any large preset number of secondary neutron generations "K", random value $J^{(K)}$ distribution law $W^{(K)}_{(j)}$ of neutrons passing through the material surface per one primary neutron of any nature is :

$$W^{(K)}_{(j)} = \sum_{m} P g(m) W^{(K)}_{(j|m)} + (1-P)\delta^{j,1} \qquad (7)$$

$$W^{(K)}_{(j|m)} = \underbrace{\left\{\left[W^{(K-1)}_{(j)} * W^{(K-1)}_{(j)}\right] * W^{(K-1)}_{(j)}\right\}}_{(m-1)\ times} * \dots$$

where:

$$\delta^{j,1} = \begin{matrix} 1 & (j=1) \\ 0 & (j\neq 1) \end{matrix} \Big\} - \text{kronecker symbol.}$$

If we are interested in distribution law of neutrons number from material surface directly per primary neutron production act of the corresponding nature, we should write:

$$W^{(k)}_{\alpha,f}(j)=\sum_{\nu} V_{\alpha,f}(\nu)\, W^{(k)}_{(j|\nu)}$$
$$W^{(k)}_{(j|\nu)}=\underbrace{\Big\{\Big[W^{(k)}_{(j)} * W^{(k)}_{(j)}\Big] * W^{(k)}_{(j)}\Big\} * \cdots}_{(\nu-1)\ times} \qquad (8)$$

where $V^{(\nu)}_{\alpha,f}$ - unconditional multiplicity distribution law of primary neutrons in (α ,n) reaction act (index "α") or in spontaneous fission act (index "f"). (Unconditional multiplicity distribution law of primary neutrons).

Evidently, according to (8) mean numbers of neutrons $\bar{j}^{(k)}_{\alpha}$ and $\bar{j}^{(k)}_{f}$ passing through the material surface per reaction (α,n) or spontaneous fission act is :

$$\bar{j}^{(k)}_{\alpha,f} = \bar{j}^{(k)} \cdot \bar{\nu}_{\alpha,f} \qquad (9)$$

where $\bar{j}^{(k)}$ - mean value of random value $J^{(k)}$ having distribution law $W^{(k)}_{(j)}$;

$\bar{\nu}_{\alpha,f}$ - mean value of primary neutrons in (α ,n)reaction or spontaneous fission act .

In its turn, from formula (7) it follows that:

$$\bar{j}^{(k)} = \sum_{j} j W^{(k)}_{(j)} = p\sum_{m} g(m)\Big[\sum_{j} j W^{(k)}_{(j|m)} + (1-p) =$$
$$= (1-p) + p\sum_{m} g(m)\bar{j}^{(k)}_{|m} = p\sum_{m} g(m)\, m\, \bar{j}^{(k-1)} + (1-p) =$$
$$= p\bar{m}\bar{j}^{(k-1)} + (1-p) \qquad (10)$$

where $\bar{m}$ - mean number of secondary neutrons in induced fission act.

Using this formula successively, we obtain:

$$\bar{j}^{(1)} = (1-p)+\bar{m}p;\ \bar{j}^{(2)} = (1-p)+\bar{m}p(1-p)+(\bar{m}p)^2;$$
$$\cdots\cdots\cdots\cdots\cdots$$
$$\bar{j}^{(k)} = (1-p)\big[1+\bar{m}p+(\bar{m}p)^2+\cdots+(\bar{m}p)^{k-1}\big]+(\bar{m}p)^{k}$$

as $\bar{m}p<1$ then:

$$\bar{j}_{\alpha,f} = \lim_{k\to\infty} \bar{j}^{(k)} \cdot \bar{\nu}_{\alpha,f} = \bar{\nu}_{\alpha,f}\,\frac{1-p}{1-\bar{m}p} \qquad (11)$$

Expression (11) is an exact expression (received for infinite number of secondary neutron generations) for mean number of neutrons per spontaneous fission act or (α ,n) reaction, so the divergence between this value and its estimation $\bar{j}^{(k)}_{\alpha,f}$ for any finite preset "K" can be considered as an error of presentation of the sought true neutron multiplicity distribution law $W_{\alpha,f}(j)$ by its corresponding estimation $W^{(k)}_{\alpha,f}(j)$. Obviously the number of secondary neutron generations "K",taken the calculation, can be chosen a priori as a rather large one to reduce this error to any small value.

Now some words about the procedure itself of mathematical convolution calculation. Obviously for discrete positively definite functions of integer-valued argument as all the distribution laws mentioned in this paper without elimination are of this kind, the operation of (m-1) -multiple convolution calculation for any K$\geqslant$1 value can be described by the following step-by-step summation procedure:

$$W^{(k)}_{(j|1)} = W^{(k-1)}_{(j)} \qquad (m=1)$$

for all m$\geqslant$2:(see expression (7))

$$W^{(k)}_{(j|2)} = W^{(k-1)}_{(j|1)} * W^{(k-1)}_{(j)} = \sum_{i=0}^{i=j} W^{(k)}_{(i|1)} W^{(k-1)}_{(j-i)}$$
$$W^{(k)}_{(j|3)} = W^{(k-1)}_{(j|2)} * W^{(k-1)}_{(j)} = \sum_{i=0}^{i=j} W^{(k)}_{(i|2)} W^{(k-1)}_{(j-i)}$$
$$\cdots\cdots\cdots\cdots\cdots \qquad (12)$$
$$W^{(k)}_{(j|m)} = W^{(k)}_{(j|m-1)} * W^{(k-1)}_{(j)} = \sum_{i=0}^{i=j} W^{(k)}_{(i|m-1)} W^{(k-1)}_{(j-i)}$$

The expressions (12) follow from the convolution mathematical definition with the only difference that the corresponding integral is substituted by finite sum because of discreteness and positively definite area of its integrand determination.

It should be noted that if one is interested in the multiplicity distribution law of neutrons from the material surface per any primary neutron production act in general, such a distribution law can be given as :

$$H(j) = \frac{1}{1+\bar{\nu}_f\alpha}\, W_f(j) + \frac{\alpha\,\bar{\nu}_f}{1+\bar{\nu}_f\alpha}\, W_\alpha(j) \quad (13)$$

where α - ratio of intensity of primary neutrons received in (α ,n) reactions to number of primary neutrons of spontaneous fission.This ratio is determined by isotope-element composition of nuclear material. The moments $\bar{j}^{s}_{H}$ of this distribution are easily determined by formula:

$$\bar{j}^{s}_{H} = \sum_{j} j^{s} H(j) \qquad s=1,2,\ldots \ (13')$$

Now some words about certain ways of determining input constants in the given model, namely, unconditional multiplicity distribution laws of primary ($V_{\alpha,f}(\nu)$) and secondary($g(m)$) neutrons and induced fission probability p.

The distribution law $V_\alpha(\nu)$ is obvious: $V_\alpha(\nu)=\delta^{\nu,1}$. The distribution law $V_f(\nu)$ is assumed to be equal to the corresponding spontaneous fission neutron multiplicity distribution law for Pu-240 isotope /3/ which usually introduces significant contribution into total intensity of spontaneous fission acts.

The unconditional distribution law $g(m)$ is represented here as "weighted" sum of individual distribution laws $g_\ell(m)$ of separate fissionable nuclides including into the given nuclear composition which can be conveniently given by the following expression /4/:

$$g_\ell(m) = \frac{m_{max}!}{m!(m_{max}-m)!}\left(\frac{\bar{m}_\ell}{m_{max}}\right)^{m}\left(1-\frac{\bar{m}_\ell}{m_{max}}\right)^{m_{max}-m} \quad (14)$$

where ℓ - index characterizing the specified fissionable nuclide type in the nuclear composition being considered;

m_{max} - upper boundary of distribution which equals to six;

$\overline{m}_\ell$ - mean number of neutrons formed in the induced fission act for ℓ – type fissionable nuclide.

If it is assumed that total cross-sections of primary and secondary neutron interactions with nuclei of all uranium and plutonium isotopes for secondary and primary neutron energy values are practically equal to each other, then the distribution law $g(m)$ can be expressed as:

$$g(m)=\sum_\ell \sigma_f^\ell \lambda_\ell g_\ell(m) \Big/ \sum_\ell \sigma_f^\ell \lambda_\ell$$

where σ_f^ℓ - nuclei fission cross-sections of ℓ - type fissionable nuclide by primary and secondary neutrons;

λ_ℓ - concentration of ℓ-type nuclide nuclei in the given nuclear composition.

Similarly the corresponding moments of distribution $g(m)$ are determined:

$$\overline{m^s}=\sum_\ell \sigma_f^\ell \lambda_\ell \overline{m_\ell^s} \Big/ \sum_\ell \sigma_f^\ell \lambda_\ell \quad s=1,2\ldots$$

The cross-sections σ_f^ℓ averaged over neutron energy spectrum and the first moments $\overline{m}_\ell$ of individual distributions $g_\ell(m)$ are commonly used nuclear constants which are sufficiently well known for all uranium and plutonium isotopes.

One should,of course, use the most exact information on these constants; the appearance of new experimental data on distribution laws $g_\ell(m)$ themselves which can be easily used in the given model instead of expression (14) is also rather important .

If there is an assurance that value α is sufficiently well determined from isotope-element composition of the given nuclear composition and if there is a possibility to have results of the examined material neutron measurements at any device of neutron-neutron coincidence counter HLNCC family, then value p can be determined from condition:

$$\frac{\begin{vmatrix} T\varepsilon^{-1} & \ell_{12}(p) \\ 2RY^{-1}\varepsilon^{-2} & \ell_{22}(p) \end{vmatrix}}{\begin{vmatrix} \ell_{11}(p) & T\varepsilon^{-1} \\ \ell_{21}(p) & 2RY^{-1}\varepsilon^{-2} \end{vmatrix}} = \alpha \overline{\nu}_f \qquad (15)$$

and as is given in /1/:

$$\ell_{11}(p)=\frac{1-p}{1-\overline{m}p}; \left(\overline{\nu}_f=2.156; \overline{\nu_f(\nu_f-1)}=3.824/3/\right)$$

$$\ell_{12}(p)=\frac{\overline{\nu}_f(1-p)}{1-\overline{m}p}; \ \ell_{21}(p)=\frac{\overline{m(m-1)}(1-p)^2 p}{(1-p\overline{m})^3}$$

$$\ell_{22}(p)=\left(\frac{1-p}{1-\overline{m}p}\right)^2\left[\overline{\nu_f(\nu_f-1)}+\frac{p\overline{\nu}_f \overline{m(m-1)}}{1-p\overline{m}}\right]$$

Here T, R - all count rates of neutrons and neutron-neutron coincidences, respectively, measured at HLNCC device;

ε, Y - detector efficiency and sampling coefficient of neutron coincidences,respectively, which are known instrument constants for the given measuring means.

In conclusion it should be noted that the solution of the sought distribution law determination task is obtained in implicit form, so as a final result ready for practical usage the version of the operating computer program realizing the given algorithm is proposed.The program text is presented in Appendix.

The author considers this program only as an example and assumes its modernization by other engaged users.

The given program version is operable for materials with any isotope-element composition at $p \lesssim 0.15$.

3. Results of Practical Usage

As a result of practical usage, the multiplicity distribution law of neutrons from surface of really existing products,made of fissionable material.In one case it was a ball with mass of ~ 83g of pure metal plutonium with abnormally high Pu-240 content (~90%) /5/ and in the second case it was a fresh assembly made of MOX material /6/ containing about 10 kg of plutonium (full mass of fuel was about 40 kg).

The usage of results of these objects neutron measurements by means of HLNCC family devices and α value estimations received on the basis of data on their isotope-element composition /5/,/6/ allow to determine according to the above-mentioned principle the p values for them and then the multiplicity distribution laws themselves of neutrons from their surfaces which are given in Table. The corresponding moments of four orders are also given there. In this Table the similar distribution laws for the given materials at hypothetic case of p=0 are presented for comparison.

These results show everything by themselves and hardly need any comments. The only note which can be done deals with the necessity of taking into account about nine generations of secondary neutrons for achieving required accuracy of these distribution laws calculations for p>0 (about some characters after decimal point for moment $\overline{\gamma}_H$).

4.Conclusion

In the present paper the analytical model and the corresponding computer program for determining the multiplicity distribution law of neutrons from the surface of object containing fissionable material with significant multiplication effect are given.

The algorithm has no limitations in part of nuclear material isotope-element composition and during the calculations as many induced fission neutrons are taken into account as is needed for achieving preset calculation accuracy.

As input data for calculation, commonly used nuclear constants such as mean number of neutrons born in the induced fission act for each fissionable nuclide composing the given nuclear composition averaged over energy spectrum and their corresponding fission cross-sections in intrinsic neutron field of the given nuclear material are used.

If there is an assurance that the ratio of primary neutrons to spontaneous fission neutrons depends only on fissionable material isotope element composition and there is a possibility to carry out additional neutron measurements of the object at HLNCC family device, the problem of determining the sought distribution is solved very simply.

The author hopes that the proposed model will be useful at developing new fissionable material monitoring methods and in this sense will successfully supplement the known Böhnel theory.

TABLE

$\dot{\gamma}$	METAL /5/		MOX FUEL /6/	
	P=0	P=0.0516	P=0	P=0.0802
0	0.655(-1)	0.657(-1)	0.295(-1)	0.301(-1)
1	0.232	0.221	0.654	0.606
2	0.329	0.301	0.148	0.138
3	0.251	0.228	0.113	0.109
4	0.101	0.103	0.457(-1)	0.544(-1)
5	0.184(-1)	0.365(-1)	0.828(-2)	0.243(-1)
6	0.240(-2)	0.196(-1)	0.108(-2)	0.140(-1)
7	0	0.112(-1)	0	0.868(-2)
8	.	0.605(-2)	.	0.530(-2)
9	.	0.330(-2)	.	0.327(-2)
10	.	0.191(-2)	.	0.211(-2)
15	.	0.145(-3)	.	0.279(-3)
20	.	0.131(-4)	.	0.441(-4)
25	.	0.131(-5)	.	0.771(-5)
30	.	0.669(-7)	.	0.143(-5)
40	.	0	.	0
50	0	0	0	0
***	**********	**********	**********	**********
$\overline{\dot{\gamma}_H}$	2.156	2.428	1.520	1.842
$\overline{\dot{\gamma}^2_H}$	5.98	8.58	3.242	6.121
$\overline{\dot{\gamma}^3_H}$	18.96	41.38	9.088	35.36
$\overline{\dot{\gamma}^4_H}$	68.45	273.04	30.47	313.71

APPENDIX

```
        PROGRAM DISTR.
        DIMENSION BP(600),V(7),PR(600,2),G(7)
        COMMON SVER(600,6)
556     DO 8 I=1,600
8       BP(I)=0.0
555     CALL ISOT (G,SN,AN,SF,AL1)
        NEL=0
        JJ=1
        NO=7
        NG=1
        GOTO 2
1       JJ=2
2       NE=7
        IF (JJ.EQ.2) GOTO 31
        V(1)=0.0
        V(2)=1.0
        DO 41 I=3,7
41      V(I)=0.0
        GOTO 38
31      V(1)=0.065
        V(2)=0.2319
        V(3)=0.3289
        V(4)=0.2514
        V(5)=0.1015
        V(6)=0.0184
        V(7)=0.0024
38      IF (JJ.EQ.2) GOTO 19
        IF (NG.GT.1) GOTO 19
71      FORMAT (1X,'I.F.MOMENTS:',5X,'M1=',F6.3,5X,'M2=',F6.3,5X,
        1'F1=',F6.3,5X,'F2=',F6.3)
        GM=G(2)+2*G(3)+3*G(4)+4*G(5)+5*G(6)+6*G(7)
        GM1=G(2)+4*G(3)+9*G(4)+16*G(5)+25*G(6)+36*G(7)
        GF1=2*G(3)+6*G(4)+12*G(5)+20*G(6)+30*G(7)
        GF2=6*G(4)+24*G(5)+60*G(6)+120*G(7)
        TYPE 71,GM,GM1,GF1,GF2
55      FORMAT (15X,5E10.3)
C       TYPE 55,(G(I),I=1,7)
        TYPE*,'                ','NEUTR.MEASUR.? (0-YES;1-NO)'
        ACCEPT*,MH
        IF(MH.EQ.0) GOTO 5
        TYPE 26
26      FORMAT (' I.F.PROBAB.=',$)
        ACCEPT*,PY
        GOTO 19
5       CALL TR (AL1,GM,GF1,PY)
19      CONTINUE
        IF (JJ.EQ.2) GOTO 433
        IF (NG.EQ.1) GOTO 22
        DO 85 I=1,7
        V(I)=G(I)*PY
85      CONTINUE
22      IF (NG.GT.1) GOTO 433
        BP(1)=G(1)*PY
        BP(2)=1+PY*(G(2)-1)
        DO 17 I=3,7
17      BP(I)=G(I)*PY
        GOTO 89
433     DO 599 I=1,NO
        BP(I)=PR(I,1)
599     CONTINUE
89      CALL SV (NO,BP)
        DO 77 I=1,600
77      PR(I,JJ)=0.0
        DO 79 I=2,600
        DO 79 MR=1,6
79      PR(I,JJ)=PR(I,JJ)+SVER(I,MR)*V(MR+1)
        IF (NG.EQ.1) GOTO 14
        IF (JJ.EQ.2) GOTO 14
```

```
      PR(2,JJ)=PR(2,JJ)+(1-PY)
14    PRS=0.0
      DO 81 I=2,600
81    PRS=PRS+PR(I,JJ)
      PR(1,JJ)=1-PRS
      IF (JJ.EQ.2) GOTO 113
113   DO 111 I=7,NO*6
      IF (PR(I,JJ).GT.1E-7) NE=NE+1
111   CONTINUE
      IF (NEL.EQ.NE) GOTO 1
      IF (JJ.EQ.2) GOTO 143
      NEL=NE
      NG=NG+1
      NO=NE
      GOTO 2
143   DO 139 I=1,600
139   BP(I)=0.0
      DO 140 I=1,NE
140   BP(I)=PR(I,2)*SF+PR(I,1)*AN
      TYPE*,'                ','OUTPUT DISTR.LAW:'
      TYPE*,'____________________________________'
      TYPE 55,(BP(I),I=1,NE)
      TYPE*,'____________________________________'
      DIS1=0.0
      DIS2=0.0
      DIS3=0.0
      AMAT1=0.0
      DO 52 I=1,NE
      SS=I
      DIS1=DIS1+(SS-1)**2.0*BP(I)
      DIS2=DIS2+(SS-1)**3.0*BP(I)
      DIS3=DIS3+(SS-1)**4.0*BP(I)
      AMAT1=AMAT1+(SS-1)*BP(I)
52    CONTINUE
      DIS=DIS1-AMAT1
      TYPE*,'MOMENTS:','J1=',AMAT1,'J2=',DIS1,'J3=',DIS2,'J4=',DIS3
      TYPE 72,AL1
72    FORMAT (5X,'*****','ALPHA=',F6.3,'*****')
      TYPE*,'COEFF.OF MULT.=',AMAT1*(1.+AL1*2.1563)/2.1563/(1.+AL1)
      TYPE*,'CONTINUE? (0-NO;ANY-YES)'
      ACCEPT*,IV
      IF(IV.NE.0) GOTO 556
      STOP
      END
C     DEFIN.OF CONVOL.____________________
      SUBROUTINE SV (NO,B)
      DIMENSION B(600)
      COMMON C(600,6)
      DO 1 I=1,6
      DO 1 J=1,600
1     C(J,I)=0.0
      DO 2 J=1,NO
2     C(J,1)=B(J)
      DO 300 MR=2,6
      DO 300 J=1,NO*MR
      DO 300 I=1,J
      C(J,MR)=C(J,MR)+C(I,MR-1)*B(J-I+1)
300   CONTINUE
      RETURN
      END
C     DEFIN.OF ISOT.COMP.________________
      SUBROUTINE ISOT (G,SN,AN,SF,AL1)
      DIMENSION PU5(7),PU8(7),PL8(7),PL9(7),PL0(7),PL1(7),PL2(7),G(7)
      TYPE 1
1     FORMAT (' %PU-238=',$)
      ACCEPT*,C1
      TYPE 2
2     FORMAT (' %PU-239=',$)
      ACCEPT*,C2
      TYPE 3
3     FORMAT (' %PU-240=',$)
      ACCEPT*,C3
      TYPE 4
4     FORMAT (' %PU-241=',$)
      ACCEPT*,C4
      TYPE 5
5     FORMAT (' %PU-242=',$)
      ACCEPT*,C5
      TYPE 6
6     FORMAT (' %AM-241=',$)
      ACCEPT*,C6
      TYPE 7
7     FORMAT (' U/(U+PU) (%) =',$)
      ACCEPT*,U
      TYPE 8
8     FORMAT (' U235/U ENRICHMENT (%) =',$)
      ACCEPT*,U7
      TYPE*,'MATERIAL? (0-METAL;1-OXIDE;0,---->-OTHER KIND)'
      ACCEPT*,AL
      S0=(1.97*C1+1.83*C2+1.31*C3+1.39*C4+1.03*C5)*(100.0-U)
      1+U*(1.26*U7+0.52*(100.0-U7))
      S1=1.97*C1*(100-U)
      S2=1.83*C2*(100-U)
      S3=1.31*C3*(100-U)
      S4=1.39*C4*(100-U)
      S5=1.03*C5*(100-U)
      S6=1.26*U7*U
      S7=0.52*(100-U7)*U
      DATA PU5/0.032,0.149,0.288,0.297,0.173,0.0536,0.00693/
      DATA PU8/0.0239,0.124,0.267,0.307,0.2,0.0687,0.00989/
      DATA PL8/0.0156,0.0937,0.234,0.313,0.234,0.0937,0.0156/
      DATA PL9/0.0031,0.0597,0.2271,0.3367,0.233,0.1263,0.0141/
      DATA PL0/0.0144,0.0883,0.228,0.312,0.241,0.0988,0.0169/
      DATA PL1/0.0103,0.0708,0.202,0.308,0.264,0.12,0.023/
      DATA PL2/0.0169,0.0988,0.241,0.312,0.228,0.0888,0.0142/
      DO 15 I=1,7
      G(I)=(S1*PL8(I)+S2*PL9(I)+S3*PL0(I)+S4*PL1(I)+S5*PL2(I)+
      1S6*PU5(I)+S7*PU8(I))/S0
15    CONTINUE
      SN=0.0
      DO 16 I=1,7
      SN=SN+(I-1)*G(I)
16    CONTINUE
      IF (U.EQ.100.0) GOTO 20
      A=(134.0*C1+0.381*C2+1.41*C3+0.013*C4+0.02*C5+26.9*C6)*AL
      S=4.73*(2.54*C1+2E-5*C2+C3+1E-8*C4+1.69*C5)
      AN=A/(A+S)
      SF=S/(A+S)
      AL1=AN/SF/2.1563
      GOTO 22
20    AN=AL
      SF=1.0-AL
      AL1=99999999.0*AL
22    RETURN
      END
C     USING OF HLNCC____________________
      SUBROUTINE TR (AL1,GM,GF1,PY)
      TYPE*,'DEVICE: EFF.(%),','DIE-AWAY TIME (MKC),','GATE (MKC)
:'
      ACCEPT*,E,TL,TG
      E=E/100.
      TYPE*,'TOTAL (1/SEC),','REAL COIN. (1/SEC):'
      ACCEPT*,T,R
      Y=EXP(-4.5/TL)*(1.-EXP(-TG/TL))
      DO 1 I=1,1000
      PY=(I-1)*0.0002
      A11=(1.-PY)/(1.-PY*GM)
      A12=2.1563*A11
      A21=GF1*A11**2*PY/(1.-PY*GM)
      A22=A11**2*(3.824+PY*2.1563*GF1/(1.-PY*GM))
      V=(T/E*A22-A12*2.*R/Y/E**2)/(A11*2.*R/Y/E**2-A21*T/E)
```

```
      IF(V.GT.AL1*2.1563) GOTO 2
1     CONTINUE
2     TYPE*,'I.F.PROBAB.=',PY
      RETURN
      END
```

5. Reference

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MEASUREMENT OF A FRESH MOX-LWR TYPE FUEL ASSEMBLY UNDER WATER

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Abstract

A fresh MOX-PWR type fuel assembly in a 17x17 configuration has been assayed under water with an adapted FORK type detector wherein fission chambers were replaced by ^{3}He tubes. Shift register electronics were used, revealing total neutrons and real coincidence rate.

The influence of pin removal was investigated at the periphery and in a central position.

The boron concentration in the water was gradually increased to a maximum value of 2200 ppm and the influence on totals and reals was examined.

1. Introduction

With the use of mixed oxides (U+Pu)(MOX) in Light Water Reactor (LWR) fuel, there is an increased tendency to store fresh MOX fuel assemblies in the reactor pool under water, in view of their loading in the reactor.

This material becomes more or less inaccessible for verification, and as there is currently no adequate tool for independent verification available, a safeguards problem arises in confirming non diversion of nuclear material.

In order to overcome this inconsistency, an agreement was made with the Euratom Safeguards Inspectorate to modify an existing FORK detector, so as to incorporate the relevant equipment, appropriate to measure the output of a non-irradiated fuel assembly.

These technical feasibilities are outlined in paragraph 2.

The system has been tested as well, in order to have an appropriate idea about the performance of the system in field conditions. IAEA inspectors were invited to participate in this test phase.

Fuel of the Venus critical facility was used, as described in paragraph 3, and full account of the experimental results is given in paragraph 4.

Throughout the text, comparison is made to results obtained in a similar exercise at Los Alamos National Laboratory (LANL).

2. Experimental Set Up and Equipment

2.1. The installation of the fuel in the water tank.

MOX fuel, originating from the VENUS critical facility, was set up in a mock-up frame simulating a real 17x17 LWR fuel assembly. This mock-up was installed in a cylindric container of 1 m diameter and a height of 1.2 m.

In order to make sure the safety of the operation, a calculation of the k_{eff} of the system has been made by the SCALE-2 computer code. A 17x17 square configuration, containing 289 pins of fuel composition as described in paragraph 3, and a pitch of 12.6 mm yielded a k_{eff} = 0.727 ± 0.004. Although the calculation yielded a fair confidence in the undertaking, the approach to criticality [1] was followed, in order to assure remaining in the subcritical state.

This procedure was followed to start the exercise, and in the follow-up, to make any change in the pin configuration, like pin removal.

2.2. The equipment: the modified FORK detector

Starting from the existing FORK II detector that is regulary used for the verification of irradiated fuel [2], a number of modifications have been made:

* the relative insensitive fission chambers (Centronic type FC167, 0.12 c/n cm^2 s) have been replaced by ^{3}He counters (Reuter-Stokes type RS-P4-0805-223, 20 c/n cm^2 s). The two pairs of ^{3}He tubes are put symmetrically in the polyethylene inserts and no Cd shielding is applied. The pairs of counters are wired separately (representing an adequate performance check), but both signals can be added and treated together, improving count rate and statistics.

* a gross gamma measurement is no longer required, so that the ionization chambers could be removed.

- it was decided to readjust the discriminator settings of the original AMPTEK A-111A's and to adjust them for the lower output pulse of the ^{3}He counters. They were moved inside the FORK II head. The lead shield for the preamplifiers was no longer required so that shield and stainless steel covers were replaced by two blind flanges.

- signal transmission was made to an HLNCC electronics shift register, and totals and reals count rates were obtained. Data treatment is made as in the traditional HLNCC experiment.

- in the final version the high voltage DC converter is mounted in the FORK II detector head itself because the multicoax connectors are rated for 500V only. During this experiment the shift register HV convertor was used.

- a general view of the electronic interconnections of the ^{3}He counters to the AMPTEK's is given in figure 1.

So that prototype was choosen that was declared having the best performance in an earlier experiment [5,6].

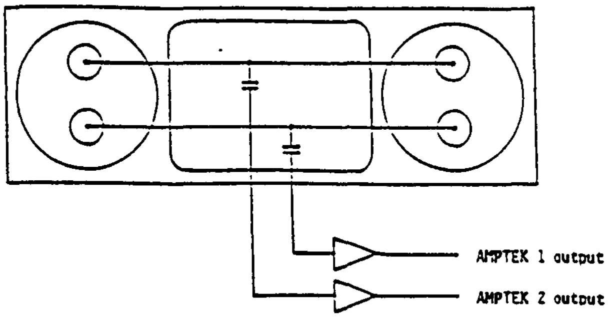

Fig.1. Electronic interconnections from the ^{3}He counters to the AMPTEK's

3. Description of the Fuel Assemblies

The fuel pins used in this experiment originate from the VENUS critical facility with characteristics as given in Table 1.

Herein Pu isotopic composition is given at analysis and measurement date, as well as the weight per rod, amounting to 7g Pu at the analysis date. Other parameters mentioned are chemical composition, linear specific weight, and physical parameters concerning fuel and cladding materials.

For this experiment a basic mock-up of 17x17 pins was used. The grid from which the pins are easily removed (pin removal experiments) and/or replaced (absorber rod experiments) has a pitch of 12.6 mm.
The mock-up was positioned in a stainless steel cylindrical tank (diameter 1 m, height 1.2 m, water level 0.8 m).

Table 1: FUEL CHARACTERISTICS

	Pu isotopics/rod			
Isotope	November 15, 1966		October 25,1990	
	(g)	(wt%)	(g)	(wt%)
Pu-238	0.005	0.068	0.005	0.058
Pu-239	5.547	79.247	5.547	80.969
Pu-240	1.204	17.197	1.204	17.538
Pu-241	0.213	3.044	0.066	0.979
Pu-242	0.031	0.445	0.031	0.455
Am-241		0.075		2.159
Isotopic Composition (wt%)	U-234		0.016	
	U-235		2.002 ± 0.005	
	U-236		0.013	
	U-238		97.970	
Stoichiometry			2.046 ± 0.005	
Chemical Composition (wt%)	UO_2		97.30	
	PuO_2		2.70 ± 0.03	
Linear spec. weight (per rod)	U		5.0 g	
	Pu		7.0 g	
Fuel diameter			0.902 ± 0.004 cm	
Fuel lenght			50.00 ± 0.50 cm	
Cladding			SS 304	
Cladding thickness			0.038 ± 0.002 cm	

4. Experimental Results

4.1. Die-away time measurements

To start with, the die-away time of the system has been determined. In a classical way, the gate length was varied over 8, 16, 32, 64 and 128 µs and the related totals and reals count rates were registered. During the whole exercise, the pre-delay was kept fixed at 4.5 µs. These data are presented in Table 2. It is interesting to note that the totals count rate is fairly constant.

Table 2: REALS COINCIDENCE RATE AS A FUNCTION OF GATE LENGHT

(HT = 1680 V, 2200 ppm Boron)		
Gate lenght (µs)	R (c/s)	T (c/s)
8	28.66	6720.84
16	50.58	6713.40
32	90.47	6700.95
64	145.02	6702.33
128	221.03	6702.84

A DEMING curve fitting to the reals count rate was made of the form:

$$y = a1 \, [\, 1 - \exp(-a2x) \,]$$

in which x is the gate length (μs) and y the reals count rate (cps).
The fitting parameters obtained are:

$$a1 = 289.98 \pm 10.66$$

$$a2 = 1.1174E{-2} \pm 7.9413E{-4}$$

From this equation the die-away time of the detector was determined as 1/a2 = 89.5 μs. Consequently, the gate length of 128 μs has been used in the measurements.

4.2. Influence of the high-voltage on the neutron detectors

The influence of the bias voltage on the neutron detector response was investigated as well. The high-voltage was varied between 1300 V and 1850 V in 50 V steps in different pin configurations. Gate length was set at 64 μs and 128 μs.
Some experiments were performed in the waterbath containing 2200 ppm Boron. The obtained totals and reals count rates are given in Table 3 and displayed in Fig. 2 .
Consequently, it was decided that the optimum detector bias voltage was at 1650 V.

Table 3 : MOX FORK HIGH VOLTAGE PLATEAU

	Gate lenght 64μs 2200 ppm Boron		Gate lenght 128μs ID12	
HV	T (c/s)	R (c/s)	T (c/s)	R (c/s)
1300	904.45	0.53		
1350				
1400	2517.09	13.84		
1450			6189.54	136.08
1500	4872.13	75.89	7478.34	263.32
1550	6055.90	113.94	7787.99	269.96
1600	6544.61	150.90	7905.31	311.39
1650	6656.44	149.40	8100.25	257.83
1680	6702.33	145.02	8428.47	311.97
1700	6775.30	152.99	8526.91	303.75
1750	7165.63	163.90	10444.69	186.19
1800	9398.90	91.99	17023.00	80.33
1850	19306.92	-160.26	25868.77	68.75

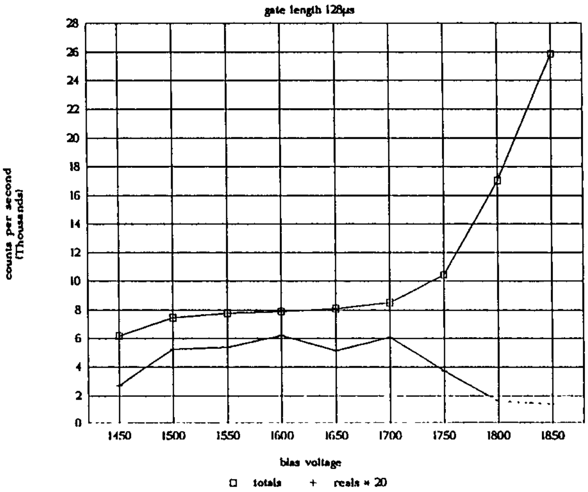

Fig.2. MOX FORK HV plateau

4.3. Calibration experiment

Measurements started from a 17x17 configuration with the 25 locations for control rods empty, as shown in Fig.3 , corresponding to commercially available assemblies.

A complete row and column of fuel pins was removed from the original set-up reducing it to a 16x16 fuel assembly, in which 20 empty positions were left , corresponding to actual 16x16 fuel assemblies [3]. The MOX FORK position was adjusted as to have the fissile mass centrally placed between the fork arms. This situation doesn't really correspond to a 16x16 fuel assembly measurement, because the pitch is not changed, and is probably to be considered as pin removal in a very pronounced way.

The same philosophy applies to a reduction to 15x15 and 14x14 assemblies, that have been formed from previous models by removing rows and columns in the appropriate way and leaving empty places as in commercial assemblies [3].
The totals and reals, as well as multiplication corrected reals are given in Table 4.

Multiplication correction was applied, using the Ensslin formalism [4], with the following values:

$$\alpha = 0.6434$$

$$\rho_{o} = 0.0043$$

	A	B	C	D	E	F	G	H	I	J	K	L	M	N	O	P	Q
1																	
2																	
3						X			X			X					
4				X										X			
5																	
6			X			X			X			X			X		
7																	
8																	
9			X			X			X			X			X		
10																	
11																	
12			X			X			X			X			X		
13																	
14				X										X			
15						X			X			X					
16																	
17																	

Fig.3. 17x17 VENUS Mock-up fuel pin location
X = empty fuel pin location
(simulating control rod thimble)

A DEMING curve fitting was made of the form:

$$y = a1\ x + a2$$

with y the reals coincidence rate and x the Pu-240eff mass loading (from table 4), given by the expression

$$\text{Pu-240eff} = 2.52\ f(238) + f(240) + 1.68\ f(242)$$

with f(i) the weight percent fraction of the appropriate Pu isotope.
The results obtained are:

$$a1 = 2.2185 \pm 5.4584E-2$$

$$a2 = -1.3949 \pm 3.2109E-1$$

with a correlation coefficient of 0.998187.
The different patterns have very close Rc normalized to Pu-240eff density:

pattern		
17x17	:	2.00 counts/s/g/cm
16x16	:	1.99
15x15	:	1.96
14x14	:	1.91

4.4. Pin removal experiment

Additional pin removal experiments were performed, such as the removal of 20 pins in the outer respectively second rows, and a removal of 16 pins from row 7, always squared around the assembly midpoint.

The experimental data concerning pin removal location are given in Table 5. From this table it becomes clear that pin removal of 7.6% (20 pins) causes a reduction in reals coincidence count rate of 6.1% in the outer rows whereas a 6.1% pin removal in the inner rows yields a 5% reduction in the count rate, clearly giving a non-linear effect.

This fact is not surprising: a pitch of 12.6 mm does not represent an optimized situation concerning the effective thermalization of neutrons. By removing pins from the second row, or even any other inside row, thermalization can take place in a more effective way, increasing the reactivity of the assembly. A general trend should be that the more central the pins are removed, the least will be the effect per pin on reals coincidence count rate.

This effect is moreover confirmed by a recent study [5,6], looking for similar effects on a 15x15 fuel assembly. In this case 6% pin removal in the outer row caused a 14% reals count rate reduction, whereas in the second row a 6% increase was seen. Although the general trend being similar, this clearly indicates the sensitivity of the result to the pitch of the fuel assembly as well as to the exact location where pins are removed.

4.5 Absorber and poison effects

The influence of absorber pins was investigated as well. AgInCd pins were used, and were introduced in the 17x17 mock-up as displayed in Fig.3 and once in combination with stainless steel pins, put alternately in the empty positions.
The results are given in Table 6.

Adding 16 AgInCd pins reduces the reals count rate by 9.4%. If 8 absorber pins are again replaced by 8 stainless steel pins, the reals count rate is reduced by 6.9%.
If the absorption process were linear, then the 8 stainless steel pins were supposed to contribute for an additional 2.6% absorption of the neutrons.
Poison rods obviously reduce totals (T), coincidence (R) and multiplication corrected coincidence (Rc) count rates.

4.6. Measurement in borated water

Up to this point the experiments were based on pin removal and absorber rod influence, but all were performed in a bath of demineralized water.

As can be expected in actual reactors, the pool water may contain more or less boric acid, that is used to absorb the neutrons from the stored fuel assemblies.

An experiment was carried out to estimate the influence of boron concentration on the MOX FORK performance.
Concentrated boric acid solutions were added to the tank, followed by homogenization and sample taking. Target boron concentrations of 250 ppm, 500 ppm, 750 ppm, 1000 ppm, 1500 ppm, 2000 ppm and 2200 ppm were in reality 503, 758, 1080, 1520, 2020 and 2170 ppm.

So we relied on sample taking and chemical analysis, or on operator declared values. If an independent NDA check of boron concentration is required, propositions are made elsewhere [5,6], consisting of either the use of a boron detector or the use of detector type 2 that contains a bare ^{3}He and a Cd wrapped ^{3}He chamber (similar to the FORK detector for spent fuel).
All measured (T & R) and calculated (M & Rc) values are considerably affected by boron. The multiplication correction procedure is the same as for unborated water, but a boron adjustment factor is determined from a calibration measurement, so that Rc = R / rMf(B).

The results of the MOX-FORK performance as a function of Boron concentration is given in table 7 and the boron adjustment factor f(B) is given in figure 4. It is clear that the reals count rate is reduced to half its value at 2200 ppm boron concentration.

Table 4 : Totals & Reals in pin removal experiment - No Boron

pattern (n° pins)	Pu-240eff (g/cm)	T (c/s)	dT (c/s)	R (c/s)	dR (c/s)	r	M	CF	Rc (c/s)	dRc (c/s)	A (c/s/g/cm)	dA
17x17(264)	6.818	16804.14	84.13	1211.52	15.12	27.55	3.23	88.88	13.63	0.17	2.00	0.02
17x17(244)	6.301	14738.39	73.78	927.26	11.54	24.05	3.04	73.08	12.69	0.16	2.01	0.03
16x16(236)	6.095	13580.71	68.01	783.78	10.72	22.06	2.93	64.58	12.14	0.17	1.99	0.03
15x15(205)	5.299	10841.17	54.24	530.86	6.20	18.71	2.73	51.07	10.40	0.12	1.96	0.02
14x14(180)	4.648	8523.99	42.73	344.37	5.74	15.44	2.52	38.85	8.86	0.15	1.91	0.03

Table 5 : Totals & Reals - geometrical positions of pin removal

pattern (n° pins)	Pu-240eff (g/cm)	T (c/s)	dT (c/s)	R (c/s)	dR (c/s)	r	M	CF	Rc (c/s)	dRc (c/s)	A (c/s/g/cm)	dA
17x17(264)	6.818	16804.14	84.13	1211.52	15.12	27.55	3.23	88.88	13.63	0.17	2.00	0.02
row 1(244)	6.301	14738.39	73.78	927.26	11.54	24.05	3.04	73.08	12.69	0.16	2.01	0.03
row 2(244)	6.301	15953.78	4.21	1202.18	8.60	28.80	3.29	94.72	12.69	0.09	2.01	0.01
row 7(248)	6.404	16509.18	4.28	1285.50	8.90	29.76	3.34	99.30	12.95	0.09	2.02	0.01

Table 6 : Totals & Reals with absorber rods

pattern (n° pins)	Pu-240eff (g/cm)	T (c/s)	dT (c/s)	R (c/s)	dR (c/s)	r	M	CF	Rc (c/s)	dRc (c/s)	A (c/s/g/cm)	dA
17x17(264)	6.818	16804.14	84.13	1211.52	15.12	27.55	3.23	88.88	13.63	0.17	2.00	0.02
16AgInCd(280)	6.818	12208.74	3.68	527.29	6.56	16.51	2.59	42.71	12.35	0.15	1.81	0.02
8AgInCd+8SS(280)	6.818	13626.78	3.89	714.56	7.33	20.04	2.81	56.31	12.69	0.13	1.86	0.02

Table 7 : Totals & Reals as a function of Boron Concentration

Boron conc. (ppm)	Pu-240eff (g/cm)	T (c/s)	dT (c/s)	R (c/s)	dR (c/s)	r	M	CF	Rc (c/s)	dRc (c/s)	A (c/s/g/cm)	f(B)
17x17 no boron	6.818	16804.14	83.13	1211.52	15.12	27.55	3.23	88.88	13.63	0.17	2.00	1.000
250ppm	6.818	12862.08	3.78	810.32	6.93	24.08	3.04	73.23	11.07	0.09	1.62	0.812
503ppm	6.818	11121.17	3.52	608.49	5.99	20.91	2.86	59.84	10.17	0.10	1.49	0.746
758ppm	6.818	9701.95	3.28	462.68	5.23	18.23	2.70	49.18	9.41	0.11	1.38	0.690
1080ppm	6.818	8752.25	3.12	390.13	4.72	17.04	2.62	44.67	8.73	0.11	1.28	0.640
1520ppm	6.818	7794.53	2.94	299.26	4.20	14.67	2.46	36.14	8.28	0.12	1.21	0.607
2020ppm	6.818	6962.22	2.78	233.78	3.75	12.83	2.33	29.90	7.82	0.13	1.15	0.574
2170ppm	6.818	6708.65	2.73	214.32	3.61	12.21	2.28	27.88	7.69	0.13	1.13	0.564

Table 8 : Totals & Reals in a positioning experiment

positioning	Pu-240eff (g/cm)	T (c/s)	dT (c/s)	R (c/s)	dR (c/s)	r	M	CF	Rc (c/s)	dRc (c/s)	A (c/s/g/cm)	dA
14x14 Right	4.648	8658.88	43.41	368.16	5.94	16.25	2.57	41.77	8.81	0.14	1.90	0.03
Symmetric	4.648	8523.99	42.73	344.37	5.74	15.44	2.52	38.85	8.86	0.15	1.91	0.03
Left	4.648	8526.56	42.74	347.75	5.76	15.59	2.53	39.37	8.83	0.15	1.90	0.03
16x16 Right	6.095	13539.24	67.81	809.96	10.90	22.86	2.97	67.99	11.91	0.16	1.95	0.03
Symmetric	6.095	13580.71	68.01	783.78	10.72	22.06	2.93	64.58	12.14	0.17	1.99	0.03
Right	6.095	13576.90	68.00	803.47	10.86	22.62	2.96	66.94	12.00	0.16	1.97	0.03

Table 9: Totals & Reals in a positioning experiment - 17x17 - Boron 1520ppm - f(B)=0.607

distance d (cm)	Pu-240eff (g/cm)	T (c/s)	dT (c/s)	R (c/s)	dR (c/s)	r	M	CF	Rc (c/s)	dRc (c/s)	A (c/s/g/cm)	dA
0	6.818	7794.53	2.94	299.26	4.20	14.67	2.46	36.14	13.64	0.19	2.00	0.03
1	6.818	7751.48	2.94	282.74	4.18	13.94	2.41	33.61	13.86	0.20	2.03	0.03
2	6.818	7706.46	2.93	289.54	4.15	14.36	2.44	35.05	13.61	0.20	2.00	0.03
3	6.818	7585.04	2.90	276.51	4.09	13.93	2.41	33.58	13.56	0.20	1.99	0.03
4.5	6.818	6823.77	2.75	230.46	3.68	12.91	2.34	30.15	12.59	0.20	1.85	0.03

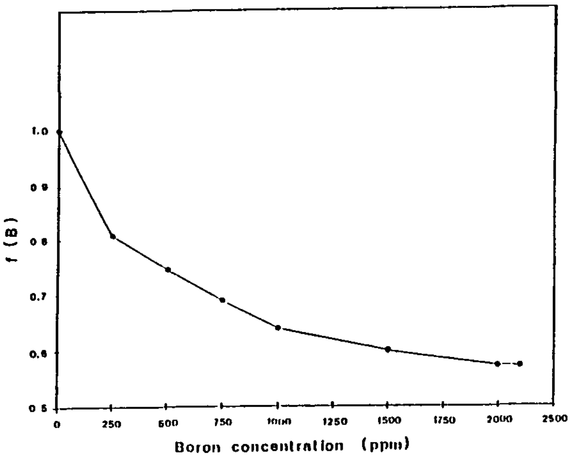

Fig.4. Boron adjustment factor

4.7 Sensitivity to mispositioning

In all previous measurements, the fuel was kept in the middle of the two detector arms, to avoid any asymmetry effect that could be caused by the equipment.

Now, because of the complex nature of the measurement, it is often difficult to keep the FORK in a stable position.
Therefore we did like to investigate the sensitivity of the measurement to incorrect positioning of the detector around the fuel bundle. The symmetry effect was tested with a 16x16 and a 14x14 fuel assembly in a pure water bath.

The results are given in Table 8. The position uncertainty within approximately 2 cm in any direction has no significant effect on the measurement results. Within statistical errors, no distinction could be made. The distance d between the assembly and the FORK backplate was varied as well between 0 cm (close contact) and 4.5 cm. The results are given in Table 9. Up to 3 cm distance from the backplate of the FORK, positioning seems to have no influence on the measurement results.
Because of the boron content of 1520 ppm the Rc values include the boron adjustment factor too (f(B) = 0.607).

5. Conclusions and recommendations

Tests have been made with the MOX-LWR FORK detector.

The effects of pin removal and absorber-poison rods in the fuel assembly have been investigated.

The removal of a number of pins corresponding to a 6% decrease in Pu content, is clearly detectable based on evaluating the multiplication corrected count rates.
The uncertainty in positioning the FORK detector does not significantly affect the verification results.

A correction coefficient can be established to account for the boron concentration in water. The multiplication corrected reals rate is more sensitive to pin removal in borated water than in unborated.

Measurements of different fuel assembly configurations show very close values of multiplication corrected reals rates normalized to Pu linear specific weight.

Poison rods obviously reduce the detector response even after multiplication correction. Based on that fact, one possible way of diversion - simultaneous removal of poison rods and a corresponding quantity of fuel - requires further investigation.

Although good agreement was found with LANL results, a different assay efficiency was found for which the explanation is not evident and that needs further consideration.

Even if the problem of verification of MOX-LWR fuel under water is not completely solved, a lot of progress has yet been made.

6. Acknowledgement

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EXPERIMENTAL STUDY OF MOISTURE EFFECTS ON PLUTONIUM FUEL MEASUREMENT FOR NEUTRON COINCIDENCE COUNTING WITH COINCIDENCE MOMENTS DETERMINATION

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Abstract

The moisture effect on plutonium measurements by neutron coincidence method was investigated experimentally using the advanced shift register electronics unit that realizes the first three coincidence moments counting. Two samples were used for experiments: one of them contains about 50 g PuO_2 powder, another - 500 g mixed dioxide fuel.

The measurement results are presented for water contents up to about 12 wt%. The coincidence moments determination was used for combined evaluation of multiplication factor M and alpha-ratio of (α,n)-neutron to spontaneous fission neutron yields. The use of the obtained M and α values for multiplication correction allowed to reduce the moisture effect on corrected coincidence count rate to less than 1% measurement statistical error level.

1. Introduction

Moisture is one of the spread matrix impurities contained in oxide plutonium fuel that causes the error of plutonium measurement by neutron coincidence counting. The two main processes are at the bottom of moisture effect on measurement results. Firstly, the presence of the moisture in the fuel increases the (α,n)-reaction neutron yield due to the water oxygen. Secondly, the neutron moderation by water hydrogen enhances the multiplication effect. Furthermore, the softening of neutron energy spectrum due to moderation usually causes the neutron detection efficiency to be increased. As a result the increase of coincidence rate is observed owing to the moisture effects.

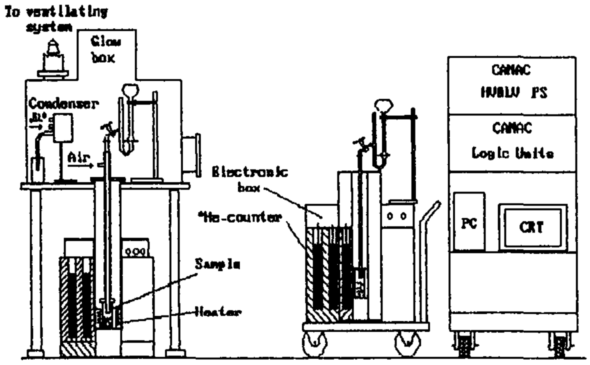

Fig. 1. Experimental facility.

The known correction technique /1/ is used usually in practice to overcome the effects of neutron multiplication on the assay results. This correction method requires a knowledge of the α-ratio of (α,n) to spontaneous fission neutron yields in the aside material. The coincidence and total count rates are two measured parameters and the ratio of count rates serves as an informative parameter of multiplication. The value of this ratio for a nonmultiplying sample is used as a base constant specified for the detector and depends on the neutron detection efficiency. Thus, it is necessary to consider the changes in the α value and the detection efficiency depending on the water concentration in the sample in order to take into account the moisture effect on the assay results.

According to Monte Carlo calculations /2/ for detector system of HLNCC type the major effects of water in the assayed samples can be accounted for by a change in the α value whereas the change in neutron detection efficiency is less important. This allows the multiplication correction to be accomplished for moist plutonium oxide samples if the moisture content is known.

A simple algorithm to account for the moisture effect is given by Stewart /3/. In this algorithm the α-value calculated for the plutonium oxide from its isotopic composition data was multiplied by a coefficient determined as a quadratic function of the known weight % (wt%) of the moisture content in a sample. The increase of detection efficiency due to moisture was considered to be negligible. For monitoring the moisture amount in samples the compound detector systems with a different effective moderator thickness can be used /4/. The principle of monitoring is similar to the two-ring measurement method based on neutron energy spectrum shift as a result of the moisture moderating properties. It is not easy to apply in practice this moisture correction method for various samples differing in form, mass and composition. In addition to, it is necessary to know the initial α-value for the dry fuel that is a source of the assay error, if the impurities presented in a sample affect the (α,n)-neutron yield.

The more perspective way for the problem solution is the analysis of neutron coincidence statistics which gives the information for multiplication correction with the unknown (α,n)-neutron contribution. This way is connected with development of measurement technique. The convenient analysis method is based on determination of coincidence statistical moments. The essence of this method is given in the report /5/. The method was applied in the work investigated experimentally the moisture influence on the plutonium oxide fuel measurements by neutron coincidence counting. The essential results of investigation will be presented in this paper.

2. Experimental equipment

Work was carried out at the experimental facility (Fig. 1) including the glove box with special equipment allowing to manipulate with open fuel containers, to supply the controlled amount of water into the container and to extract it by means of evaporation. Two neutron well detectors were used for measurements. One of them with 30% efficiency for fission neutrons was used for continuous monitoring of neutron emission from fuel sample in the process of moisture supply and removal. The monitor detector is installed immediately under the glove box so that the sealed measuring channel passing through the glove box bottom entered into the detector cavity by its lower part. The fuel container screwed tightly to the lower end of the special probe was located in the center of the detector through measuring channel. The controlled doses of water were supplied into the container through 0.5 m polyethylene tube. The water dosage was performed by means of a measuring stopcock burette providing accuracy about 0.02 ml per 1 ml of water.

The electrical heater and special condenser mounted at the top of probe and cooled by water were applied for the moisture extraction. To intensify the extraction process the dry air was passed through the container.

The second detector of high efficiency (about 40% for fission neutrons) is not connected directly with the glove box. The simple device with measuring stopcock burette was used for moisture addition. The father moisture extraction was performed in the glove box.

No means of fuel agitation for the moisture homogenization were applied.

In both detectors the neutrons are counted by He-counter tubes symmetrically embedded into the polyethylene moderator block around the central cavity in circular geometry. To reduce dead-time counting losses the counter tubes are divided into groups. The outputs of groups can be combined so that the neutron pulses from the inner and outer rings of counter tubes can be counted separately or together.

The detectors are supplied with the common electronic equipment located into the CAMAC crates. Three coincidence electronics units have been made in CAMAC standard and can be used in experiments. Two of them realize the ordinary shift-register coincidence logic and are similar functionally to the HLNCC coincidence circuit. The third electronics unit realizes the advanced version of coincidence circuit which allows the first three statistical moments of neutron coincidence to be determined.

The coincidence units are managed by program. The accumulated counting data are read through the CAMAC data-way into the computer memory for proper processing.

3. Algorithm for coincidence moments counting method

Six output counts of advanced coincidence unit are the measurement results:

. total number of detected neutrons T_o;

. accumulated sums of coincidence numbers from prompt and delayed gates of coincidence circuit $(R+A)_1$ and $(A)_1$, correspondingly;

. accumulated sums of the squared coincidence numbers from prompt and delayed gates $(R+A)_2$ and $(A)_2$, correspondingly;

. accumulated sum of the cubed differences of these numbers $(R)_3$.

The count rates for the first three real coincidence moments were determined using these six counts normalized to the counting time t:

$$R_1=(R+A)_1/t-(A)_1/t,$$
$$R_2=(R+A)_2/t-(A)_2/t-2\cdot R_1\cdot(A)_1/T_o$$
$$R_3=(R)_3/t-6\cdot R_1\cdot[(A)_2/T_o-(A)_1{}^2/T^2{}_o].$$

These count rates as well as the total count rate $T=T_o/t$ are the four measured parameters which after dead-time correction can be used for solving of the assay problem.

The problem solution is described in ref. /5/. The count rates for coincidence moments is shown to be defined via the fission rate and factorial moments of neutron multiplicity distribution. The Bohnel model /6/ was applied to make allowance for multiplication effect on the factorial moments. In point geometry and one-group energy assumption this model allowed to obtain the set of four equations relative to four unknowns: spontaneous fission rate, efficiency of neutron detection ϵ, multiplication factor M and α parameter.

However, examination of method with plutonium fuel samples showed that application of the third coincidence moment is unsuccessful due to the high statistical measurement error. Thus, the two first moments only were used to evaluate in common M and α parameters, considering the neutron detection efficiency as a constant value.

The multiplication factor M is determined by solving the cubic equation

$$AM^3 + BM^2 + CM + D = 0, \qquad (1)$$

the coefficients of which present the following expressions:

$$A=d(d_2-d_1),$$
$$B=dR_1/(Tr_o)-A,$$
$$C=(d_1-d)R_1/(Tr_o),$$
$$D=(R_1-R_2)/(4Tr_o{}^2),$$

where $d=(W_{1sp}W_{2in})/[W_{2sp}(W_{1in}-1)]$,

$d_1=(W_{1sp}W_{3sp})/(3W_{2sp}{}^2)$,

$d_2=(W_{1sp}\cdot W_{3in})/(3\cdot W_{2sp}\cdot W_{2in})$,

$r_o=G\cdot\epsilon\cdot W_{2sp}/(2\cdot W_{1sp})$,

W_{jsp} and W_{jin} - j-th factorial moments of neutron multiplicity distribution for spontaneous and induced fissions correspondingly,

G - probability to get into coincidence gate for neutrons of the same fission event.

The value of r_o parameter depends on neutron detection efficiency and therefore, in general it can be changed slightly because of the matrix effect. In constant efficiency assumption the value of parameter can be determined by direct measurement using a small "nonmultiplying" plutonium sample

$$r_o=(R_2/R_1-1)/(4\cdot d_1).$$

The multiplication factor M is found as the following solution of the cubic equation (1):

$$M=2\cdot P^{1/2}\cdot\cos(\theta/3)-B/(3\cdot A), \qquad (2)$$

where $P=B^2/(9\cdot A^2)-C/(3\cdot A)$,

$\theta=\arccos\{[BC/(3A)-2B^3/(27A^2)-D]/(2AP^{3/2})\}$.

Having determined M, the unknown value of α-parameter is found

$$\alpha=M/[R_1/(Tr_o)-M\cdot(M-1)\cdot d]-1 \qquad (3)$$

and multiplication-corrected real coincidence rate is calculated

$$R_{cor}=R_1/\{M^2\cdot[1+(M-1)\cdot(1+\alpha)\cdot d]\}. \qquad (4)$$

The foregoing algorithm was implemented in software of the neutron coincidence system with the coincidence moments counting unit.

4. Measurement of moisture effects

Preliminary experiments

Before the fuel samples measuring the preliminary experiments were performed in order to test the procedures of moisture supply and extraction, to try out the main characteristics of counting system and to choice the mode of measurement and data acquisition.

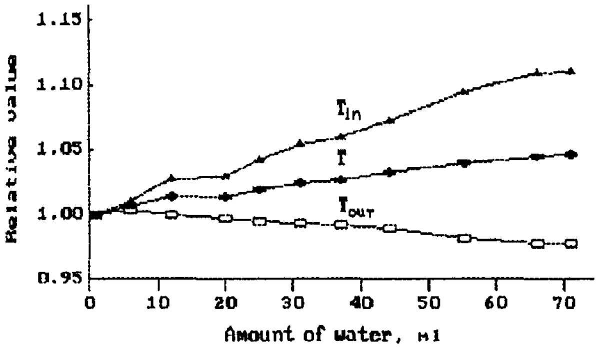

Fig. 2. Relative change of totals for ^{252}Cf depending on amount of water in ZrO_2.

As in studying the moisture effects it is necessary to measure during the long time the special attention was given to stability of detector response. The response drift in 2-3% of initial value was observed for both detectors during the first day after switching in the high voltage supply. However, after two days of operation the response drift from the reached saturation level did not exceed 0.2%.

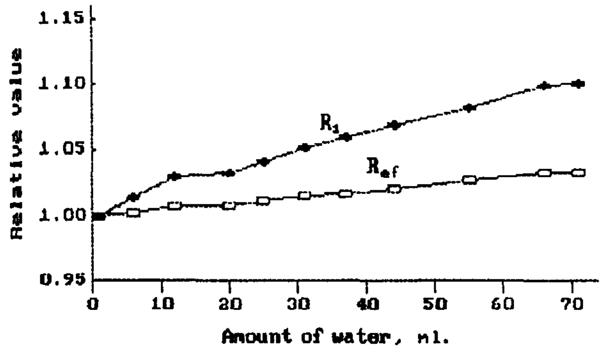

Fig. 3. Relative change of reals for ^{252}Cf as a function of water amount in ZrO_2 before and after correction for efficiency variation due to water.

The measurements were performed in multiscalar mode allowing to carry out the operative statistical processing of data in order to evaluate the random error of measurement results and response drift during the measurement.

As a rule two coincidence units were used simultaneously in the experiments. The ordinary coincidence unit was applied to measure the response of inner (or outer) ^{3}He-counter ring. The response of the whole detector was measured by the coincidence moments counting unit.

Procedures of water supplying into container and moisture extraction were preliminary tested using a container filled with ZrO_2 powder. A small californium source was fixed in the middle of the container. The neutron absorption in ZrO_2 is negligible and so the experiments with adding of water in ZrO_2 allow to study the moisture effect as a neutron moderator on the efficiency of californium neutron detection. Fig. 2 shows the relative change of the total count rates (totals) from the counter rings and from the common output of monitoring detector as a function of water amount in ZrO_2. Growth in totals T of the whole detector is occurred by increasing of totals T_{in} of the inner ring while totals T_{out} of the outer ring varies slightly and even has an opposite tendency. The two-ring measurements allow to obtain the information about the detector efficiency change due to the moisture influence. For example, totals ratio T_{in}/T can be used as an informative parameter for rough estimation of relative efficiency variation according to the relation

$$K_{ef}=1+T_{in}/T-N, \qquad (5)$$

where N is a value of T_{in}/T ratio for dry ZrO_2.

The real coincidence count rate (reals) can be corrected for efficiency variation due to moisture using this relation as it is shown in Fig. 3, where the corrected reals R_{ef} was determined as R_1/K_{ef}^2.

Experiments with fuel samples

Two fuel samples were used in experiments. The fuel is contained in the thin-walled stainless steel containers 44 mm in diameter, 145 mm in height. One of them contained PuO_2 powder (with 96% abundance of ^{239}Pu), another one - electrodeposited mixture of PuO_2 and dioxide of natural uranium. The characteristics of samples are given in table 1. The given values of effective ^{240}Pu mass and α-parameter were calculated using the known data of plutonium isotopic composition.

Table 1. Characteristics of plutonium fuel samples.

Sample	Fuel mass, g		^{240}Pu effect mass, g	α
	PuO_2	UO_2		
MTK-1	48.69	-	1.68	1.131
MTK-9	124.93	374.80	38.65	0.861

The more careful measurements of the moisture effect were performed in the detector with 40% efficiency. As it was mentioned no tools were used for agitation of moist fuel in the container. The natural homogenization of moisture in fuel was expected to be during some time after adding the every water portion. The stabilization of detector response was considered to correspond with the end of homogenization process. It was determined that the transition period doesn't exceed 20 minutes. The measurement results of this period were leaves out of account.

The observed count rates for coincidence moments after water adding into MTK-9 sample up to 12 wt% are shown in Table 2. The random error of measurement is characterized by the standard deviation of results with 10 minute measurement time. The results averaged for 17-20 measurements are given in table. One can note the significant growth in the count rates with the moisture increasing.

Table 2. The observed count rates for MTK-9 sample after adding of moisture.

Moisture		Count rates, sec^{-1}			
ml	wt%	T	R_1	R_2	R_3
0	0.0	34207± 7	4840±26	6500±52	10656±336
1	0.2	34304± 9	4850±24	6514±55	10790±522
3	0.6	34534±11	4868±29	6542±46	10712±507
4	0.8	34579±11	4875±18	6571±47	10858±621
5	1.0	34693± 8	4885±19	6588±59	11005±546
6	1.2	34743± 9	4892±29	6591±62	10629±705
7	1.4	34866±12	4905±26	6618±57	11000±543
12	2.3	35522± 8	4959±25	6701±53	11073±539
15	2.9	35747± 6	4991±21	6757±52	11108±427
20	3.8	36094± 9	5040±26	6828±56	11386±472
25	4.8	36392± 8	5083±29	6892±59	11401±494
30	5.7	36594±12	5137±23	6982±42	11560±514
35	6.5	36652±20	5144±30	6985±54	11662±644
40	7.4	36657±16	5131±25	6969±56	11586±328
45	8.3	36688±10	5149±25	7010±68	11500±606
50	9.1	36720±11	5146±17	7001±36	11484±511
55	9.9	36756± 9	5165±22	7040±66	11621±706
60	10.7	36782±11	5176±13	7054±47	11703±635
65	11.5	36797±10	5189±25	7067±52	11651±499
70	12.3	36801±13	5186±26	7076±49	11690±522

The high measurement error is observed for the third coincidence moment. For this reason the first two coincidence moments only were applied for multiplication correction in accordance with algorithm set down above. The values of multiplication factor M and α-parameter were estimated assuming that the neutron detection efficiency does not depend on the moisture amount in the sample. The parameter $r_o=0.226\pm0.001$ was determined from measurement of the small sample containing the PuO_2 thin layer the neutron multiplication in which can be neglected. The factors $d=2.06$, $d_1=0.26$ and $d_2=0.36$ in cubic equation (1) were calculated from the neutron multiplicity distribution for spontaneous and induced fissions of plutonium isotopes. The calculated values M, α and multiplication-corrected reals R_{cor} are shown in table 3.

One can see that the growing of α-value is observed mainly with the moisture increasing. The increase of α relative to its value for dry fuel reaches about 10%. The increase in multiplication factor M is less than 1%. The α-value measured for dry fuel is rather more than that calculated from plutonium isotopic data. It is necessary to note that this value is estimated in accordance with the model based on the one-group neutron energy assumption and can differ little from its actual value due to difference in detection efficiency of (α,n) and fission neutrons. However, the main reason for disagreement of calculated and determined α-value is presence of impurities in fuel affecting on (α,n)-neutron yield. According to the estimations obtained from the data about impurities in MTK-9 fuel their presence causes 10 -15% addition in (α,n)-neutron yield that is in a good agreement with the α-value determined.

Table 3. Calculated parameters and corrected reals for MTK-9 sample.

Moisture		Calculated parameters			
ml	wt%	M	α	Rcor	Ref
0	0.0	1.0428	0.959	3795 ±20	3789 ±20
1	0.2	1.0429	0.961	3802 ±21	3795 ±21
3	0.6	1.0432	0.970	3807 ±20	3797 ±20
4	0.8	1.0447	0.985	3778 ±21	3768 ±21
5	1.0	1.0449	0.991	3778 ±22	3766 ±22
6	1.2	1.0444	0.986	3795 ±19	3782 ±19
7	1.4	1.0452	0.997	3786 ±25	3770 ±25
12	2.3	1.0459	1.021	3806 ±17	3774 ±17
15	2.9	1.0468	1.032	3808 ±22	3769 ±21
20	3.8	1.0473	1.036	3836 ±21	3783 ±21
25	4.8	1.0476	1.039	3861 ±22	3796 ±21
30	5.7	1.0489	1.040	3874 ±17	3796 ±16
35	6.5	1.0484	1.037	3890 ±26	3807 ±25
40	7.4	1.0486	1.046	3873 ±27	3789 ±26
45	8.3	1.0498	1.051	3861 ±26	3775 ±25
50	9.1	1.0494	1.050	3867 ±15	3779 ±14
55	9.9	1.0503	1.054	3861 ±23	3771 ±22
60	10.7	1.0504	1.051	3869 ±21	3778 ±20
65	11.5	1.0500	1.041	3891 ±23	3798 ±22
70	12.3	1.0509	1.053	3865 ±18	3773 ±17

The multiplication-corrected reals changes little within the moisture range up to about 3 wt%. However, when a large amount of water are contained in the sample so that the fuel is supersaturated with the moisture and even a layer of water formed above the fuel column the corrected reals exceeds noticeably its value for dry fuel. Evidently it occurs due to significant increase of neutron detection efficiency caused by water. To correct this effect the measurements of totals from the inner counter ring of detector were used. The relative efficiency change was defined using the totals ratio T_{in}/T in accordance with equation (5). The value of constant N in this equation was determined as an average value of this ratio from the measurements of several samples with dry plutonium oxide fuel.

To take account of efficiency variation the corrected parameter $r_{ef}=r_oK_{ef}$ was applied in cubic equation (1) to estimate the new values of multiplication factor M_{ef} and parameter α_{ef}. The multiplication-corrected reals was determined then as

$$R_{ef}=R_1/\{M_{ef}^2K_{ef}^2[1+(M_{ef}-1)(1+\alpha_{ef})d]\}. \quad (6)$$

The value of R_{ef} calculated for MTK-9 sample is shown in the last column of table 3.

By the same way the measurements of MTK-1 sample have been carried out. The water was added into the sample by 0.5 ml portions up to 7 ml (about 12 wt%, accordingly).

Table 4. Bias percent (δR) of reals relative to its value for dry fuel before and after multiplication correction.

Sample	Moisture		Bias, %		
	ml	wt%	δR1	δRcor	δRef
MTK-9	1	0.20	0.20	0.17	0.16
	3	0.60	0.58	0.31	0.21
	4	0.79	0.73	-0.46	-0.57
	5	0.99	0.94	-0.44	-0.61
	6	1.19	1.08	0.00	-0.19
	7	1.38	1.34	-0.24	-0.51
	12	2.34	2.46	0.30	-0.40
	15	2.91	3.13	0.36	-0.53
	20	3.85	4.14	1.08	-0.16
	30	5.66	6.14	2.07	0.19
	40	7.41	6.01	2.07	-0.00
	50	9.09	6.33	1.89	-0.28
	60	10.71	6.94	1.94	-0.31
	70	12.28	7.16	1.84	-0.44
MTK-1	0.5	1.0	0.62	-0.06	-0.20
	1.0	2.0	1.26	-0.33	-0.54
	1.5	3.0	1.78	0.57	0.30
	2.0	3.9	2.54	0.54	0.19
	2.5	4.9	3.22	0.68	0.27
	3.0	5.8	3.60	0.91	0.45
	3.5	6.7	4.48	0.57	0.02
	4.0	7.6	4.54	1.21	0.56
	4.5	8.5	5.11	1.95	1.22
	5.0	9.3	5.46	1.00	0.19
	5.5	10.1	5.66	1.55	0.64
	6.0	11.0	5.42	1.92	0.92
	6.5	11.8	5.36	2.24	1.22
	7.0	12.6	4.77	1.34	0.33

The main results of moisture effect measurements are summarized in table 4, where the bias percent for reals relative to its value for dry fuel is shown as a function of moisture content before and after multiplication correction. As the results analyzed show the multiplication correction using the first two coincidence moments allows to reduce the moisture influence on corrected reals to level of measurement statistical error (less than 1%) within the moisture range up to about 3 - 4 wt%.

It should be noted that this range overlaps the area of possible values of fuel moisture in practice. The overestimated values of multiplication-corrected reals (up to about 2%) is observed when the too large quantity of water is added into the sample. The use of the inner counter ring to whole detector totals ratio to take account of the moisture influence on detector efficiency allows to remove a bias of multiplication-corrected reals within the whole range of measurements.

5. Conclusion

The use of the first two coincidence moments for multiplication correction is an effective way to minimize the moisture influence on plutonium assay in oxide fuel samples with unknown moisture content. The main advantage of this way consists in that the multiplication factor M and α-parameter for the assayed sample are estimated directly from measurement. In addition to moisture this allows to consider the matrix impurities influence on the assay results.

It should be noted that the long measuring time is necessary in order to obtain the high precision in determining of the second and in some cases the third coincidence moments. It is essential that the dead-time correction of count rates for coincidence moments is accurate over the whole count rate range. The solution of last problem is necessary to make possible the assay of large amounts of plutonium oxide fuel.

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EVALUATION OF THE U-Pu RESIDUAL MASS FROM SPENT FUEL ASSEMBLIES WITH PASSIVE AND ACTIVE NEUTRONIC METHODS

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ABSTRACT

The interpretation of passive and active neutronic measurements to evaluate the U-Pu residual mass in spent fuel assemblies is presented as follows :

- passive neutron measurements are well correlated to the plutonium mass,
- active neutron measurements give information linked to the fissile mass content of the assembly (^{235}U + ^{239}Pu + ^{241}Pu) and, using the passive neutron measurement, lead to the ^{235}U mass content of the assemblies.

1 - INTRODUCTION

Le contrôle non destructif des assemblages irradiés à l'aide de méthodes neutroniques passive et active s'avère intéressant pour l'évaluation des paramètres physiques caractéristiques de ces assemblages (taux de combustion, réactivité...) [1, 2, 3].

Ce papier présente l'interprétation de ces mesures neutroniques en termes de masse résiduelle d'Uranium et de Plutonium des assemblages irradiés.

La démarche suivie consiste à décrire successivement :

- les principes physiques des mesures neutroniques passive et active,
- les corrélations envisagées pour évaluer, à partir de ces mesures, les masses de Pu et d'^{235}U résiduelles,
- la vérification de la validité de ces corrélations sur des résultats expérimentaux.

2 - PRINCIPE PHYSIQUE DES MESURES UTLISEES

2.1 - Mesure neutronique passive

Cette mesure repose sur la détection des neutrons émis par les noyaux lourds du combustible irradié (fissions spontanées et réactions (α, n)). Le tableau ci-dessous montre la contribution des différents noyaux lourds à l'émission neutronique pour deux taux de combustion (l'assemblage considéré est un REP 17 x 17 d'enrichissement initial en ^{235}U 3.1 % et de temps de refroidissement 3 ans) :

Isotope	Contribution à l'Emission Neutronique (%)	
	TC=18000MWJ/T	TC=36000MWJ/T
^{238}Pu	3,76	0,75
^{239}Pu	1,016	0,05
^{240}Pu	5,7	0,46
^{242}Pu	0,9	0,19
^{241}Am	1,66	0,14
^{242}Cm	4,4	0,77
^{244}Cm	81	91,74
^{246}Cm	1,08	5,75

Pour les taux de combustion usuels, (> 25000 MWJ/T), le ^{244}Cm contribue de façon prépondérante (> 90 %) à l'émission neutronique du combustible.

Celui-ci étant formé à partir des isotopes du Plutonium ^{241}Pu et ^{242}Pu, une mesure de l'émission neutronique passive s'avère intéressante pour l'estimation de la masse de Plutonium.

2.2 - Mesure neutronique active

Cette technique a été développée afin d'obtenir une information directement interprétable en termes de réactivité de l'assemblage (Keff) et donc de masse fissile résiduelle [3, 4, 5, 6]. La méthode nécessite que l'assemblage soit sous eau (borée ou non).

Le principe physique en est le suivant :

- une source de neutrons est placée à proximité d'une des faces de l'assemblage (face A de la figure 1) pour créer des fissions dans le milieu,

- un détecteur de neutrons, placé de l'autre côté (face B de la figure 1) compte, pendant la présence de la source, les neutrons prompts issus des fissions provoquées par la source. Afin d'améliorer le contraste, ce détecteur est entouré d'un absorbant aux neutrons thermiques.

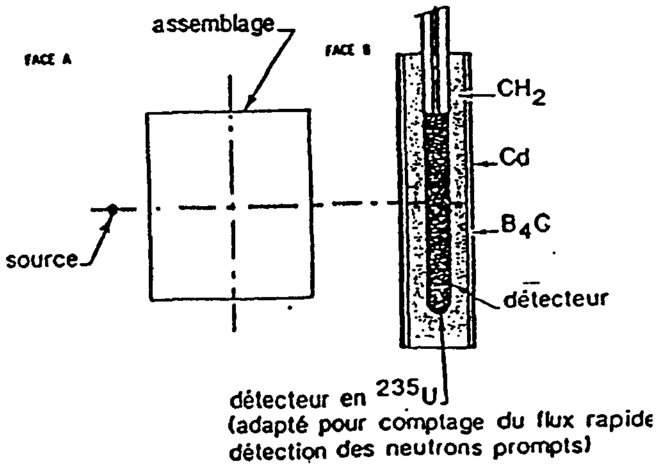

Figure 1 : Schéma de principe d'une mesure neutronique active

- du fait de l'émission neutronique passive de l'assemblage, la source est positionnée alternativement en position éloignée de l'assemblage (on mesure dans ce cas le signal "passif") puis en position proche de l'assemblage (on mesure dans ce cas le signal "actif + passif"). Cette modulation permet par la suite de distinguer ces deux signaux.

Le signal "actif" obtenu est lié à la réactivité résiduelle de l'assemblage et donc à la masse fissile (^{235}U + ^{239}Pu + ^{241}Pu).

3 - DEFINITION DES CORRELATIONS

3.1 - Corrélation émission neutronique-Masse de Plutonium

La corrélation entre la masse de Plutonium (MPu) et l'émission neutronique (E_n) peut s'écrire (cette relation est spécifique pour chaque géométrie d'assemblage) :

$$MPu = C\ f(E_n)\ [1+\varepsilon_1(EI)][1+\varepsilon_2(TR)] \quad (1)$$

avec :

C	: Constante de calibration
$f(E_n)$	: Fonction dépendant de l'émission neutronique
$\varepsilon_1(EI)$	: Fonction de correction due à l'enrichissement initial en ^{235}U
$\varepsilon_2(TR)$	: Fonction de correction due au temps de refroidissement

Une étude théorique concernant des assemblages REP 17 x 17 a permis d'évaluer les différentes fonctions décrites ci-dessus.

On considère premièrement un enrichissement initial en ^{235}U de 3.1 % et un temps de refroidissement de 1 an. Le tableau ci-après donne la masse de Plutonium en fonction de l'émission neutronique :

TC MWJ/T	MPu kg/ass	En n/s/ass
10000	2,03	1,91 10^6
18000	3,15	1,96 10^7
24000	3,80	6,34 10^7
30000	4,34	1,61 10^8
36000	4,77	3,40 10^8
40000	5,02	5,21 10^8

La figure 2 ci-dessous, qui présente ces valeurs, montre la grande sensibilité de la masse de Plutonium à l'émission neutronique :

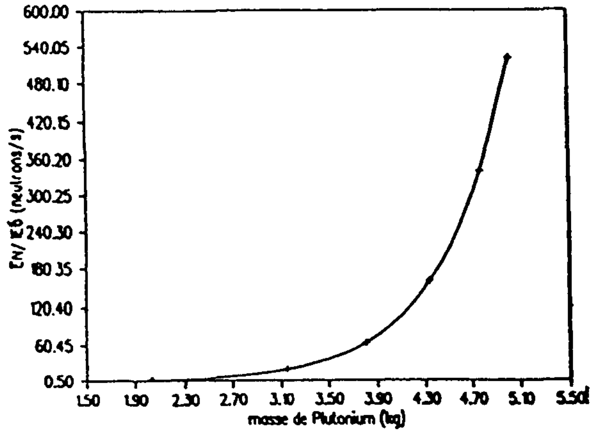

Figure 2 : Variation de la masse de Plutonium en fonction de l'émission neutronique

Ces valeurs permettent par lissage, d'expliciter $f(E_n)$:

$$f(E_n) = (E_n)^{0.153}$$

avec E_n en neutrons/s/assemblage

On considère deuxièmement un taux de combustion de 36000 MWJ/T et un temps de refroidissement de 1 an. Le tableau ci-après donne la masse de Plutonium en fonction de l'enrichissement initial en ^{235}U :

Enr ^{235}U %	1,8	2,4	3,1	3,68
m(Pu) kg/ass	4,3	4,79	4,77	4,60
Correction par rapport à 3,1 %	1,0115	1.0043	1	0,96486

Ces valeurs permettent d'obtenir la fonction de correction ε_1 due à l'enrichissement initial en ^{235}U sous la forme d'un polynôme d'ordre 2.

On considère troisièmement un taux de combustion de 36000 MWJ/T et un enrichissement initial en ^{235}U de 3,1 %. Le tableau ci-dessous donne la masse de Plutonium en fonction du temps de refroidissement :

TR an	0.5	1	3	8	15
m(Pu) kg/ass	4.79	4.77	4.71	4.58	4.53
Correction par rapport à 1 an	0.9971	1	1.0127	1.042	1.0522

Ces valeurs permettent d'obtenir la fonction de correction $\varepsilon_2(TR)$ due au temps de refroidissement, sous la forme d'une exponentielle.

Remarques

- Cette corrélation est obtenue en considérant un historique "standard" d'irradiation.

- Il est à noter que les termes de correction $\varepsilon_1(EI)$ et $\varepsilon_2(TR)$ n'interviennent que pour quelques % dans la relation (1) alors que l'émission neutronique du combustible varie fortement avec la masse de Plutonium, ce que justifie le bien fondé de la méthode.

3.2 - Corrélation mesure active - Masse de matière fissile

Le taux de comptage obtenu avec la mesure neutronique active (correspondant aux neutrons prompts de fissions) est lié de façon directe à la masse fissile de l'assemblage par une corrélation de la forme :

$$Ta = \alpha\ MF + \beta \qquad (2)$$

avec :

MF : Masse fissile résiduelle (kg)

Ta : Taux de comptage "actif" (coups s^{-1})

α β : Constantes de calibration dépendant de la géométrie des assemblages considérés et des conditions de mesure (concentration en Bore de la piscine, intensité de la source neutronique...)

La relation (2) est explicitée directement à l'aide de résultats expérimentaux.

Remarque :

La constante β correspond aux neutrons prompts de la source directement comptés. Le dispositif de mesure est optimisé de façon à minimiser ce terme.

4 - RESULTATS EXPERIMENTAUX

4.1 - Description du dispositif de mesure

Le dispositif utilisé pour les mesures neutroniques active et passive est présenté dans la figure 3 ci-après :

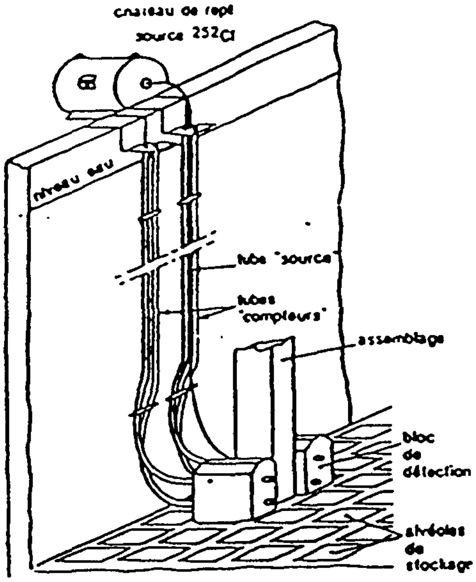

Figure 3 : Système PYTHON de contrôle des assemblages irradiés

Les deux ensembles de détection du dispositif sont positionnés dans deux alvéoles en fond de piscine et l'assemblage à mesurer est introduit entre ces deux alvéoles à l'aide du pont de manutention. Les signaux "gamma total" "neutrons passifs" et "neutrons actifs" sont obtenus simultanément. L'interprétation des mesures est effectuée en ligne par un micro-calculateur et permet de valider le taux de combustion et le temps de refroidissement et d'évaluer le facteur effectif de multiplication. On se propose ici d'utiliser les résultats des mesures neutroniques passive et active afin de tester les corrélations développées au § 3.

4.2 - Mesure neutronique passive

Ce paragraphe présente premièrement une analyse des différentes sources d'incertitudes intervenant dans l'évaluation de la masse de Pu et deuxièmement une validation de la corrélation "masse de Plutonium - Emission Neutronique" sur des résultats expérimentaux.

4.2.1 - Précision sur la masse de Pu estimée

a) Incertitude de calibration

L'expérimentation a montré que cette incertitude est inférieure au %.

b) Incertitude statistique

L'incertitude statistique sur l'estimation de l'émission neutronique $\frac{\Delta En}{En}$ est fonction du taux de combustion et est ici en moyenne de 2 % à 2 σ ce qui amène une incertitude sur la masse de Pu telle que :

$$\frac{\Delta MPu}{MPu} = 0{,}153 \frac{\Delta En}{En}$$

(cette relation est obtenue en différenciant la relation (1))

soit $\frac{\Delta MPu}{MPu} = 0{,}3\ \%$

c) Incertitude de positionnement de l'assemblage

Le fait d'utiliser la moyenne des signaux neutroniques obtenus sur deux faces opposées de l'assemblage permet de réduire cette incertitude à une valeur faible (< 1 %).

d) Incertitude due à la corrélation

Une étude paramétrique en fonction du taux de combustion, de l'enrichissement initial en ^{235}U, du temps de refroidissement et de l'historique d'irradiation a montré que cette incertitude est au maximum de 2,5 % et qu'elle est due en grande partie à l'historique d'irradiation. Elle peut donc être réduite si la corrélation tient compte de ce dernier.

e) Incertitude globale

L'incertitude globale représente la somme quadratique des incertitudes décrites précedemment soit :

$$\Delta MPu/MPu \simeq 3\ \%$$

4.2.2 - Utilisation des résultats expérimentaux

La constante de calibration de la relation (1) a été déterminée sur un assemblage de référence qe qui permet d'évaluer pour les assemblages mesurés, la masse de Plutonium estimée d'après la corrélation (1). Le tableau ci-après présente, pour quelques assemblages irradiés mesurés par comptage neutronique passif : les caractéristiques de ceux-ci, l'incertitude statistique obtenue à 2 σ (c'est-à-dire avec un niveau de confiance de 95 %) sur la mesure passive, la masse de Plutonium annoncée et la masse de Plutonium évaluée par la corrélation (1) :

Enr %	TC MWj/t	Tr ans	Emission neutronique n/s/ass	Incertitude statistique à 2 σ sur la mesure passive %	Masse Pu kg annoncée	Masse Pu corrélation kg	$\frac{\Delta MPu}{MPu}$ (%)
1.8	15400	7.5	$1.527\ 10^7$	8.5	2.95	3.12	5.45
3.25	35200	3.9	$2.144\ 10^8$	2.5	4.63	4.56	1.4
3.25	38200	0.6	$4.557\ 10^8$	0.75	4.92	5.02	2
3.45	42100	1.7	$4.851\ 10^8$	1.74	5.13	5.05	1.5
3.7	34700	0.6	$2.510\ 10^8$	1	4.58	4.50	1.7
3.7	39700	0.6	$4.198\ 10^8$	1	5.01	4.85	3

L'écart moyen d'environ 2,5 % entre la masse de Pu calculée par la corrélation et la masse réelle de Pu conforte la relation obtenue au § 3.1 et l'étude de précision du § 4.2.1.

4.3- Mesure neutronique active

De même qu'au § 4.2, on présente premièrement une analyse des différentes sources d'incertitudes intervenant dans l'évaluation de la masse fissile résiduelle et deuxièmement une validation de la corrélation "Taux de comptage actif - Masse fissile résiduelle" sur des résultats expérimentaux.

4.3.1 - Précision sur la masse fissile résiduelle estimée

a) Incertitude de calibration

L'expérimentation a montré que cette incertitude est inférieure au %.

b) Incertitude statistique

Cette incertitude, due à l'extraction du signal "actif", est en moyenne de 3 %.

c) Incertitude due au positionnement de l'assemblage

Cette incertitude a été évaluée expérimentalement à environ 2 %.

d) Incertitude due à la corrélation

Cette incertitude, liée à l'évaluation de la masse fissile résiduelle nécessaire à l'obtention de la corrélation peut être considérée comme inférieure au %.

e) Incertitude globale

L'incertitude globale représente la somme quadratique des incertitudes décrites précedemment soit :

$$\Delta MF/MF \simeq 4\ \%$$

4.3.2 - Utilisation des résultats expérimentaux

Le tableau ci-dessous présente, pour quelques assemblages mesurés par méthode neutronique active : les caractéristiques de ceux-ci, l'incertitude statistique obtenue sur la mesure du taux de comptage "actif" à 2 σ, la masse fissile résiduelle annoncée et celle évaluée par la corrélation (2) :

Enr %	TC MWj/t	Incertitude statistique à 2 σ sur τa (%)	Masse fissile annoncée (kg)	Masse fissile évaluée (kg)	$\frac{\Delta MF}{MF}$ %
1.8	15400	3.45	5.45	5.68	4
3.25	39500	3.2	6.44	6.68	3.7
3.45	41300	3.1	6.47	6.70	3.6
3.70	32400	3	8.61	8.31	3.5
3.70	38400	3	7.55	7.28	3.5
3.70	0	1.9	17.06	17.15	1

L'écart moyen d'environ 3,25 % entre la masse fissile calculée par corrélation et la valeur annoncée conforte la relation obtenue au § 3.2 et l'étude de précision du § 4.3.1.

4.4 - Combinaison des mesures active et passive pour l'évaluation de la masse d'^{235}U

Le tableau ci-après présente, pour quelques assemblages mesurés : les caractéristiques de ceux-ci, la masse d'^{235}U annoncée et celle évaluée par combinaison des mesures passive (qui donne la masse de ^{239}Pu + ^{241}Pu) et active (qui donne la masse ^{235}U + ^{239}Pu + ^{241}Pu) :

Enr %	TC MWj/t	Masse ^{235}U annoncée (kg)	Masse de ^{239}Pu+^{241}Pu évaluée (kg)	Masse ^{235}U évaluée (kg)	$\frac{\Delta M^{235}U}{^{235}U}$ %
1.8	15400	3.27	2.3	3.38	3.4
3.1	33400	3.59	2.98	3.41	5.2
3.25	35200	3.65	3.1	3.89	6.5
3.70	32400	5.36	3.19	5.12	4.5
3.70	38400	4.16	3.34	3.95	5.3

Cette évaluation montre que l'utilisation combinée des méthodes neutroniques active et passive permet d'obtenir la masse d'^{235}U résiduelle d'un assemblage irradié avec une précision de l'ordre de 5 %.

CONCLUSION

L'évaluation de la masse résiduelle d'Uranium et de Plutonium des assemblages irradiés peut s'effectuer à l'aide de méthodes non destructives.

- La mesure neutronique passive permet l'obtention de la masse de Plutonium avec une précision de l'ordre de 3 %.
- La mesure neutronique active permet d'obtenir la masse fissile résiduelle avec une précision de l'ordre de 4 % et donc, par combinaison avec la méthode passive, la masse résiduelle d'^{235}U.

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A VERSATILE PASSIVE/ACTIVE NEUTRON COINCIDENCE COUNTER FOR IN-PLANT MEASUREMENTS OF PLUTONIUM AND URANIUM*

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Abstract

A new, well-type neutron coincidence counter has been developed for nondestructive assay of a wide variety of items containing plutonium, uranium, or both. The instrument will be used in several locations at the Los Alamos National Laboratory Plutonium Facility (TA-55). Both active and passive measurements can be made using the same instrument by simply changing the well inserts. By combining results of active and passive measurements, assay of some impure plutonium-bearing items can be accomplished.

1. Introduction

We have designed, constructed, and tested a new thermal-neutron coincidence counter /1/ for general use in plants processing both uranium and plutonium. Versatility and optimum performance were primary goals of the development.

Features of the design include:

- removable sample-well inserts for passive and active modes;
- both fast- and thermal-mode inserts for active measurements;
- removable AmLi sources for combined passive/active measurements;
- AmLi sources (2) sufficiently intense (5 x 10^5 n/s each) to override passive coincidence signals for mixed U-Pu and/or impure plutonium samples;
- shielding to (1) reduce effects of plant neutron backgrounds, and (2) reduce personnel exposure;
- uniform efficiency in the sample cavity volume to minimize sample positioning effects;
- low sensitivity to sample matrix, containers, and coverings;
- passive efficiency of 37.4%;
- active efficiency of 37.0% in thermal mode and 32.7% in fast mode;
- large sample cavities to accommodate a wide range of containers; and
- inherent safety, reliability, and maintainability.

Figure 1 is an elevation sectional view of the instrument, showing details of the shielded sample well, a suspended active insert, and the lifting mechanism, which rotates to easily allow changing inserts. The insert-lifting mechanism has several unique features including both up- and down-limit switches and a quickly actuating motor brake. The lift was designed to enhance both safety and ease of operation.

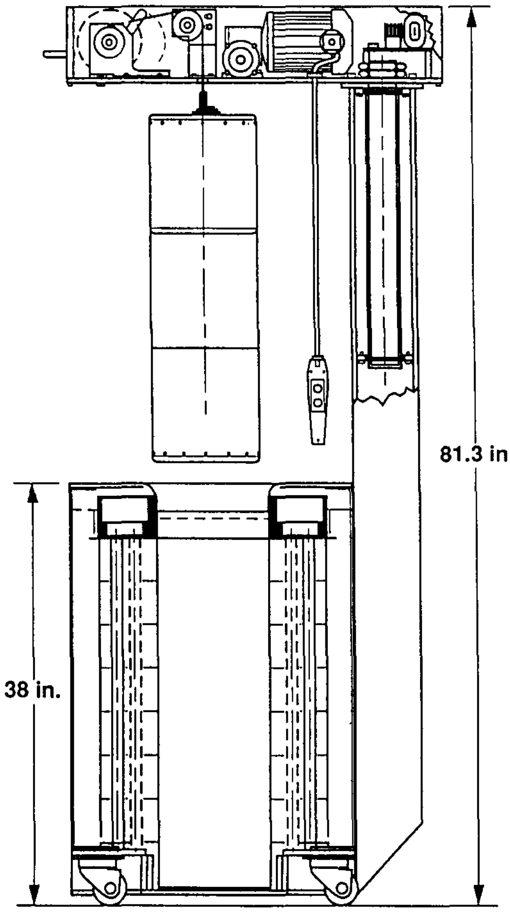

Fig. 1. Elevation sectional view of the Passive/Active Neutron Coincidence Counter (NCC).

Figure 2 is a photograph of the complete Passive/Active NCC system, showing the counter body, the lifting mechanism with the passive insert suspended, and the two active inserts resting on the floor. Also shown in the photo is a JSR-11 coincidence electronics package and the MS-DOS personal computer with peripherals. A general purpose neutron coincidence counter software package will be used with the system.

Figure 3 displays details of one of the active inserts on the left and the passive insert on the right. The ^{252}Cf source holder is shown with the passive insert.

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Fig. 2. Photograph of the Passive/Active NCC system.

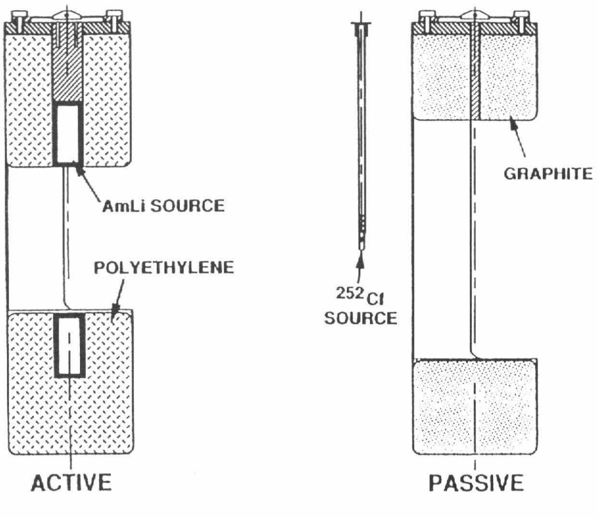

Fig. 3. Active and passive inserts for the Passive/Active NCC.

The passive insert contains graphite end plugs (each 178 mm or 7 in. thick) for optimum reflection (and minimal absorption) of neutrons originating in the plutonium-bearing sample. Also, the top plug allows placement of a ^{252}Cf source in the center of the sample cavity for routine measurement-control checks. The passive cavity will hold sample containers up to 216 mm (8.5 in.) in diameter and 406 mm (16 in.) high.

The active inserts hold polyethylene end plugs (each 273 mm or 10.75 in. thick) that include receptacles for AmLi sources immediately above and below the sample cavity. Each source is easily removed from its polyethylene end plug using handling rods. Empty AmLi source shields (tungsten "bottles") are available for passive/active measurements. The first active insert has cadmium surrounding the sample cavity (fast mode). The second insert has no cadmium (thermal mode). The active cavities will accommodate sample containers up to 216 mm (8.5 in.) in diameter and 229 mm (9 in.) tall.

The neutron moderator body is an annulus of polyethylene containing 48 ^{3}He proportional counters (tubes) placed in two concentric rings. Monte Carlo /2/ calculations were used to optimize ^{3}He-tube placement and moderator design. This approach resulted in a measured efficiency of 37.4% for the passive insert. For the active inserts, we measured 32.7% efficiency in fast mode and 37.0% in thermal mode using a centered ^{252}Cf source. The high-efficiency design yields very good detection sensitivities for both uranium and plutonium measurements.

Intended routine uses for the counter include passive measurements of plutonium-bearing samples, active thermal-mode measurements of samples with low ^{235}U masses (<50 g ^{235}U), and active fast-mode measurements of samples with high ^{235}U masses (>50 g ^{235}U).

2. Inherent Performance Characteristics

One of the primary goals of the design was to achieve nearly constant axial efficiency profiles for both passive and active modes.

Figure 4 shows measured axial efficiency profiles of totals, reals, and multiplication-corrected reals /3/ for a PuO_2 sample (LAO2–171 g of plutonium) in the passive-insert cavity. This is a differential efficiency profile, showing efficiency variation for a small sample placed at different axial positions in the cavity. The multiplication-corrected reals efficiency is uniform to within ±1% from 50 to 350 mm (~2–14 in.) above the cavity floor. This efficiency profile is nearly equivalent in uniformity to that of

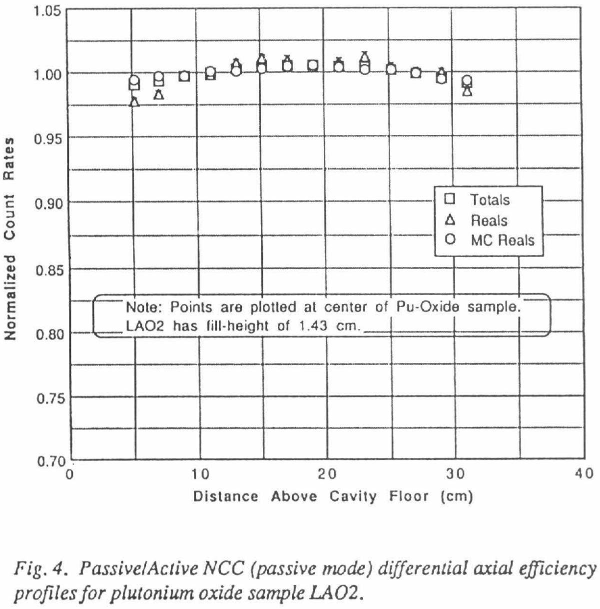

Fig. 4. Passive/Active NCC (passive mode) differential axial efficiency profiles for plutonium oxide sample LAO2.

the Flat-Squared Counter, /4/ which has the same cavity diameter, a 17% taller cavity, a single ring of 24 ^{3}He tubes, and a cadmium-polyethylene insert to flatten its profile. The Flat-Squared Counter is a passive counter only. We evaluated the cadmium/polyethylene insert idea for the Passive/Active Counter using the MCNP code. This approach was not chosen because (1) the improvement in axial efficiency shape was only marginal (for the cadmium compared to the no-cadmium case), (2) the centered source reals efficiency was lower by 13%, (3) the active-mode performance was compromised, and (4) the mechanical complexity was greater.

Efficiency profiles were also measured for both the active-fast (AF) and active-thermal (AT) modes using a ^{235}U-metal disk. Figures 5 and 6 show the results. In contrast to Fig. 4, these are integrated profiles, showing the effects of sample fill-height. Figure 5 shows that in AF mode, response is lowest in the cavity center, but is uniform to within ±5% for samples with fill-heights between 2.5 and 20 cm. Figure 6 shows that in AT mode, response is uniform to within ±1% for samples with fill-heights between 2.5 and 20 cm. These profiles compare quite favorably with those of the Active-Well Coincidence Counter or AWCC. /5/

Other pragmatic, performance oriented goals of the design were to achieve low sensitivity to the sample matrix, steel container thickness, and polyethylene covering bags.

To characterize the matrix sensitivity of the instrument with the passive insert, we measured a ^{252}Cf source inside identical cans filled with several common matrix materials. Figure 7 shows the results. The new counter is approximately a factor of two less sensitive to the matrices sand, ash, lead shot, and iron shot than is the Flat-Squared Counter. For polyethylene blocks and chips, the matrix sensitivities of the two counters are approximately the same.

In passive mode, we measured separately the effects of steel and polyethylene shells surrounding the LAO2 sample. Results are shown in Figs. 8 and 9.

Figure 8 shows relative efficiencies for totals, reals, and multiplication-corrected reals vs thickness of the external steel container. The two standard steel containers (double encapsulation) for sample LAO2 are ~0.4 mm thick. Results of Fig. 7 are normalized to unity for the case with no external container. The effect of external steel on the multiplication-corrected reals is essentially nil for radial thicknesses between 0 and 5 mm.

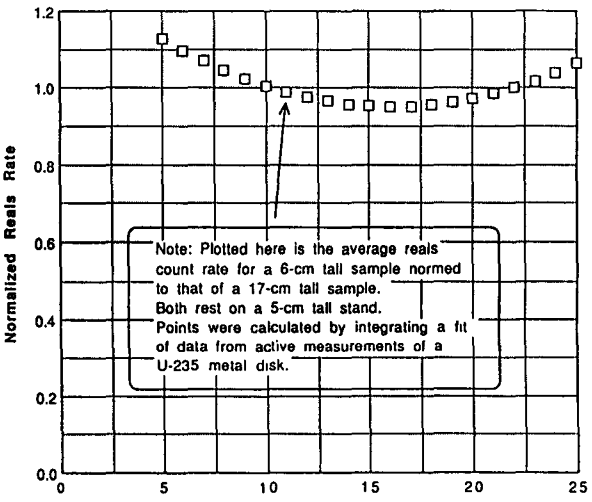

Fig. 5. Passive/Active NCC (active-fast mode) integrated axial efficiency profile (from ^{235}U metal disk measurements).

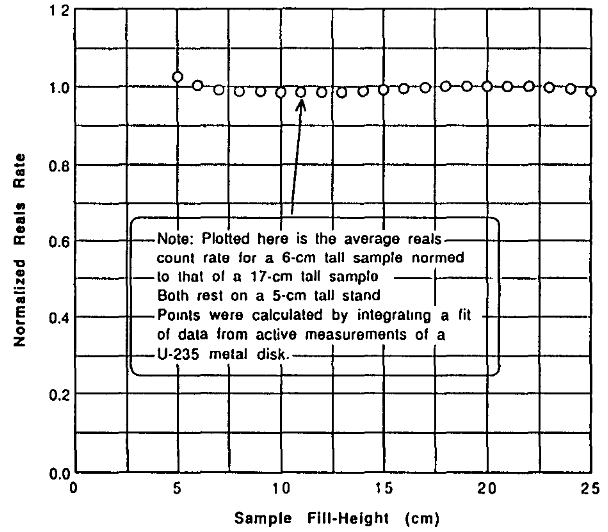

Fig. 6. Passive/Active NCC (active-thermal mode) integrated axial efficiency profile (from ^{235}U metal disk measurements).

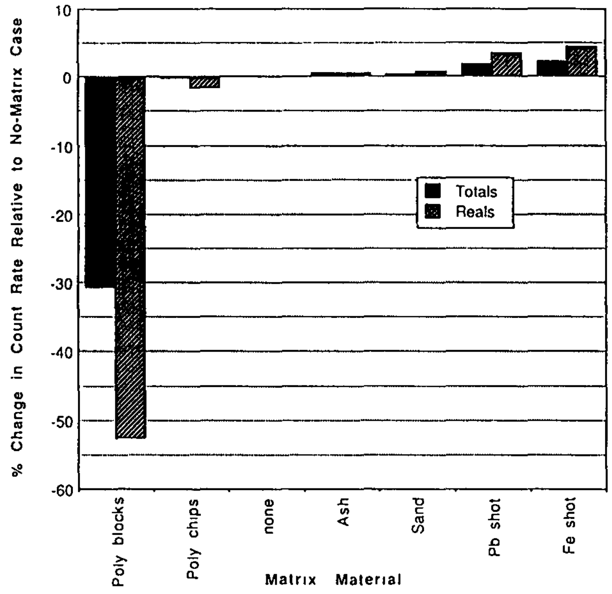

Fig. 7. Passive/active NCC (passive mode) sample matrix sensitivity comparisons

Figure 9 shows relative efficiencies for totals, reals, and multiplication-corrected reals as a function of external polyethylene thickness. Results of Fig. 8 are normalized to unity for the case with no external poly. The effect of the external poly on the multiplication-corrected reals is 1% or less for radial thicknesses between 0 and 12 mm.

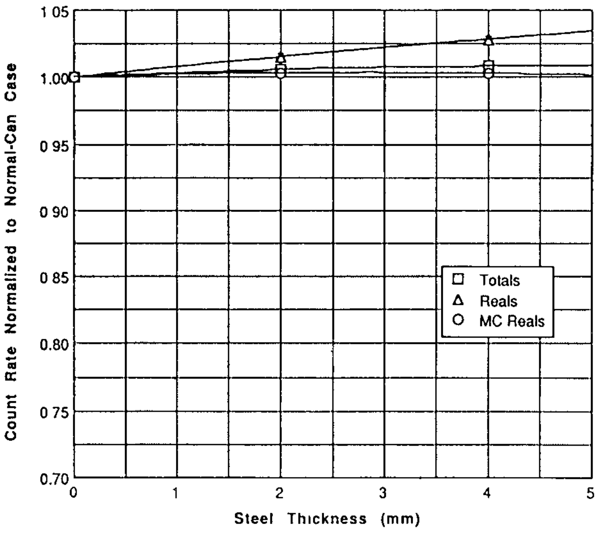

Fig. 8. Passive/Active NCC (passive mode) external steel container sensitivity for plutonium-oxide sample LAO2.

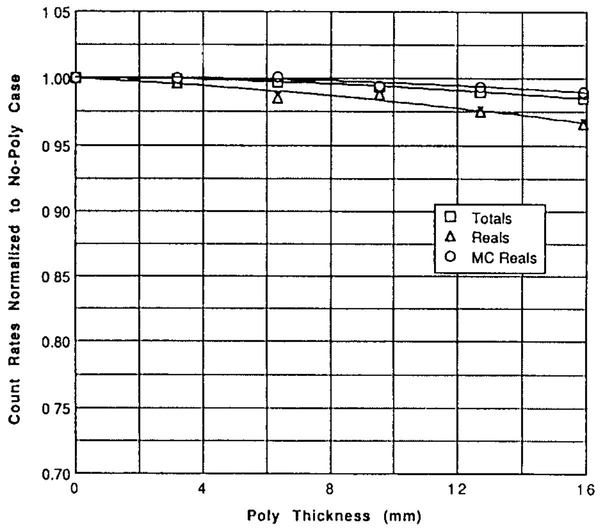

Fig. 9. Passive/Active NCC (passive mode) external polyethylene sensitivity for plutonium oxide sample LAO2.

The reals counting-rate-loss correction /6/ is made in software using a deadtime parameter that varies with the measured total count rate ($\tau = a + bTm$, where $a = 0.714$ µs and $b = 0.363 \times 10^{-6}$ µs^2). These values were taken from a least-squares fit using pairs of measurements of individual, calibrated ^{252}Cf sources.

3. Calibration Curves and Detection Limits

The Passive/Active Counter is meant for routine assay and/or verification measurements in a plant environment. Capabilities include passive measurements of plutonium-bearing samples (~5 mg to several kg ^{240}Pu$_{\mathrm{eff}}$), active thermal-mode measurements of samples with low ^{235}U masses (~2–50 g ^{235}U), and active fast-mode measurements of samples with high ^{235}U masses (~100 g to several kg ^{235}U).

Figure 10 shows the passive-mode calibration data and fitted curves for nearly pure PuO$_2$. The measured data were obtained using the LAO Series of standards (~10 to ~150 g ^{240}Pu$_{\mathrm{eff}}$). Both uncorrected and multiplication-corrected reals curves are shown. Notice the corrected data show less scatter from the fitted curve than do the uncorrected data. Generally, scatter in the uncorrected data arises from the effects of bulk-density and/or plutonium-isotopics variations on multiplication. The LAO series of standards has nearly uniform plutonium-isotopics (~17 wt% ^{240}Pu$_{\mathrm{eff}}$). From radiography and weighing, the largest of the LAO series has a bulk density of ~2.5 g/cc.

Figures 11 and 12 show the active-thermal mode and active-fast mode calibrations, respectively, for uranium oxide standards that have nearly the same oxide masses, but widely varying enrichments (low-enriched and UISO Series). /7/

Detection limits and typical mass calibration ranges for the three operating modes of the Passive/Active Counter are summarized in Table I. Detection limits quoted are for single 1000-s measurements. Here, detection limit is defined as the mass value yielding a relative precision in the measured reals rate of 1/3 or 33%.

The passive-mode detection limit was calculated using a general formula that includes the effects of counting time, calibration (reals vs mass), and ambient background rates (both reals and totals). The formula is similar to one given in Ref. 8. Background rates recorded at one of the Los Alamos ground-level safeguards measurement development laboratories were used in the calculation. The measured reals background rate was 0.5 counts/s (primarily resulting from infrequent cosmic-ray-induced neutron bursts in the counter body) and the measured totals background rate was 150 counts/s (primarily from neutron sources in shielded storage locations in the laboratory). With the lab free from sources, the measured totals rate was 30-40 counts/s. Using 40 counts/s for the background totals rate, the calculated detection limit is 1 mg ^{240}Pu$_{\mathrm{eff}}$.

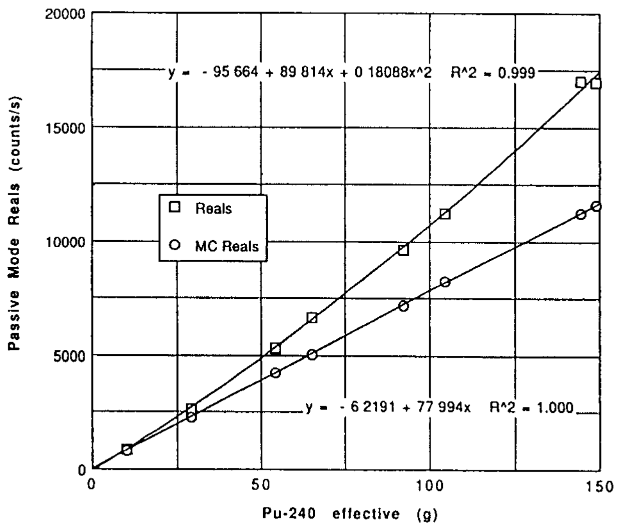

Fig. 10. Passive/Active NCC (passive mode) plutonium oxide calibration (10–150 g ^{240}Pu$_{\mathrm{eff}}$).

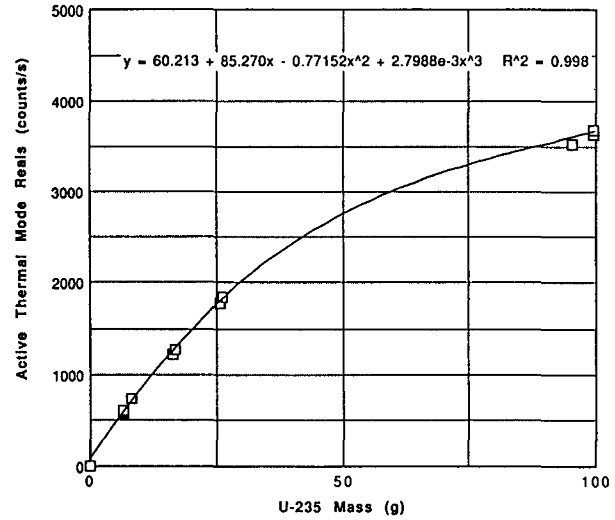

Fig. 11. Passive/Active NCC (active-thermal mode) uranium-oxide calibration (10–100 g ^{235}U).

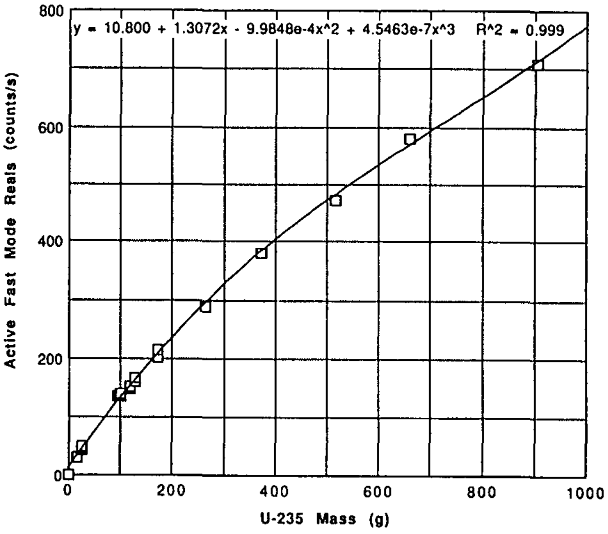

Fig. 12. Passive/active NCC (active-fast mode) uranium oxide calibration (10–1000 g ^{235}U).

TABLE I. Passive/Active Counter Detection Limits and Calibration Ranges

Measurement Mode	1000-s Measurement Detection Limits	Mass Calibration Range
Passive	1–2 mg $^{240}Pu_{eff}$	~5 mg to few kg $^{240}Pu_{eff}$
Active-Fast	50 g ^{235}U	~100 g to several kg ^{235}U
Active-Thermal	1 g ^{235}U	~2 g to 100 g ^{235}U

Active-mode detection limits were calculated using a simple formula that includes the effects of counting time and calibration (reals vs mass).

4. Combined Passive and Active Measurements

One of the long-term application goals for the Passive/Active NCC is evaluation of combined active and passive measurements for pure and impure plutonium-bearing items as well as MOX samples. To date, we have obtained experimental data for pure and impure oxide standards. Before presenting those data, we will briefly review some related work.

Reference 9 reports AWCC measurements of large plutonium oxide and metal standards. It was concluded that by using the net active real coincidence count rate (active minus passive), the ^{239}Pu content of large plutonium oxide and metal samples could be measured with a precision of 3-6% using a total counting time of 1000 s.

The standard method for application of the Neutron Coincidence Collar /10/ involves using the net active reals for ^{235}U/length assay of light water reactor (LWR) fuel elements. The passive reals rate is sometimes used for ^{238}U/length assay, although the reals count rate is low and therefore influenced by room-background-induced fissions.

Reference 11 reports both passive and net-active calibrations (with multiplication corrections) for the Fast Neutron Coincidence Counter (FNC or random driver) for plutonium oxide and metal standards. The random driver uses plastic scintillators (rather than polyethylene-moderated ^{3}He proportional counters) for neutron detection. Lead shielding is used in the FNC to reduce gamma-ray background. It was concluded that the FNC would yield 2-4% precision in 500-1000 s counting for plutonium oxide and metal samples.

In an informal Harwell report, Lambert describes some of the basic principles of plutonium assay with the AWCC. The passive reals rate is related to the mass of spontaneous-fission isotopes (^{238,240,242}Pu), and the active-minus-passive reals rate is related to the mass of the induced-fission isotopes (^{239,241}Pu) for low-burnup plutonium. Lambert concluded that the ^{239,241}Pu content of kilogram quantities of low-burnup plutonium could, in principle, be assayed using the net reals rate from active and passive measurements made by the AWCC. No experimental data were reported.

We measured the Los Alamos LAO and PEO Series of PuO_2 standards in all modes of the Passive/Active NCC. From chemical analyses and extensive analyses of neutron coincidence counting data, the LAO series is known to be nearly free of low-Z impurities (for example, F, Na, Mg, and Cl) that increase the (α,n) neutron production beyond that expected from pure PuO_2. Similarly, the PEO Series is known to contain significant and varying amounts of impurities and therefore excess (α,n) neutron production.

The LAO Series was used for calibration in all measurement modes. In passive mode, the multiplication-corrected reals (R_c) assay was used. Table II. (Passive column) shows the differences between passive assay and destructive analysis values for both the LAO and PEO series. These data are also shown in Fig. 13. Because the LAO Series was used for calibration, the LAO assay results are excellent: the average difference is +0.12 ± 0.63%. However, for the PEO Series, the average difference is +8.83 ± 3.51%.

These large differences for the PEO Series arise from using a value for α [ratio of (α,n) neutrons to spontaneous fission neutrons] that is calculated for pure PuO_2, and therefore too low for the impure samples. For these passive measurements, the measurement times varied between 600 and 2000 s. Using simple propagation of the errors in the totals and reals, the resulting precision in R_c ranged between 0.05 and 0.1%. From this, it is concluded that 0.25% precision in R_c can be obtained in only 100-s counting time for passive-mode measurements of items similar to the LAO and PEO standards. Compared to the standard High-Level Neutron Coincidence Counter (HLNCC-II) /12/ that has an efficiency of 17.5%, and the Flat-Squared Counter with an efficiency of 24.4%, the Passive/Active Counter can obtain the same passive counting precision in one-fourth and one-half the counting times, respectively.

By combining results of both passive and active measurements, another measured parameter is obtained that, in principle, allows solving for all three sample unknowns [spontaneous fission neutrons, (α,n) neutrons and induced-fission or multiplication neutrons]. Several data-analysis approaches were evaluated, but the one selected uses results of the active measurement to correct the passive-mode results for sample multiplication. A geometry and cross-section-dependent model of multiplication and an iterative procedure are part of the approach. Spectrum- and geometry-averaged macroscopic nuclear cross-sections were calculated using the MCNP code. Here again the LAO series was used for calibration of both measurements and calculations. Results are shown in Table II (passive/active column) and also in Fig. 14. These results were obtained using the active insert with cadmium (active-fast mode). Compared to the passive-mode results for the LAO standards, the passive/active results show about the same bias (almost nil) and greater scatter (+0.14 ± 2.21%). This scatter is probably from incomplete knowledge of fill-heights for all LAO standards. A single radiograph of LAO8 was used to determine fill-heights for all LAO cans. This information was then built into the calibration. For the impure PEO standards, the results are excellent; the average difference between assay and destructive-analysis values is +0.08 ± 0.80%. Measurement times for the passive measurements varied between 600 and 1800 s, with one measurement of 5400 s for the smallest sample. Measurement times for the active measurements were much longer. These varied between 1000 and 50 000 s, with two measurements of 100 000 s for small samples. Precisions were also calculated for two 1000-s measurements, one active and one passive. It is estimated that for two 1000-s measurements, the scatter in the assay-declared differences would be 3-4%. This accuracy could be improved by increasing the strengths of the AmLi sources. Most of the scatter in the assay values shown in Table II is from causes other than counting statistics.

Also in Table II, compare the two columns of α values. The pure PuO_2 α column contains values calculated from nuclear data for pure PuO_2. The passive/active inferred α column contains values predicted by the passive/active data analysis algorithm.

TABLE II. Comparison of Conventional Multiplication-Corrected Passive Assays and Combined Passive/Active Assays of Pure (LAO) and Impure (PEO) PuO_2

Sample	$^{240}Pu_{eff}$ (g)	Pure PuO_2 α	Passive/Active inferred α	Passive (A-D)/D, %	Passive/Active (A-D)/D, %
LAO1	10.098	0.427	0.471	+0.93	-0.12
LAO2	29.296	0.431	0.472	+0.28	-3.37
LAO3	54.359	0.426	0.463	+0.70	-2.81
LAO4	65.115	0.425	0.408	-0.88	+0.40
LAO5	92.188	0.433	0.399	-0.63	+2.46
LAO6	104.537	0.426	0.401	+0.49	+2.62
LAO7	144.365	0.423	0.411	-0.06	+1.07
LAO8	149.126	0.423	0.414	+0.12	+0.85
			Average	+0.12	+0.14
			Std. Dev.	0.63	2.21
PEO1	7.418	0.622	0.870	+12.54	-0.38
PEO2	14.836	0.622	0.817	+11.75	-0.88
PEO3	29.672	0.622	0.788	+11.14	-0.02
PEO4	43.339	0.625	0.704	+6.71	+1.50
PEO5	64.998	0.615	0.722	+7.20	-0.04
PEO6	81.374	0.622	0.677	+3.66	+0.28
			Average	+8.83	+0.08
			Std. Dev.	3.51	0.80

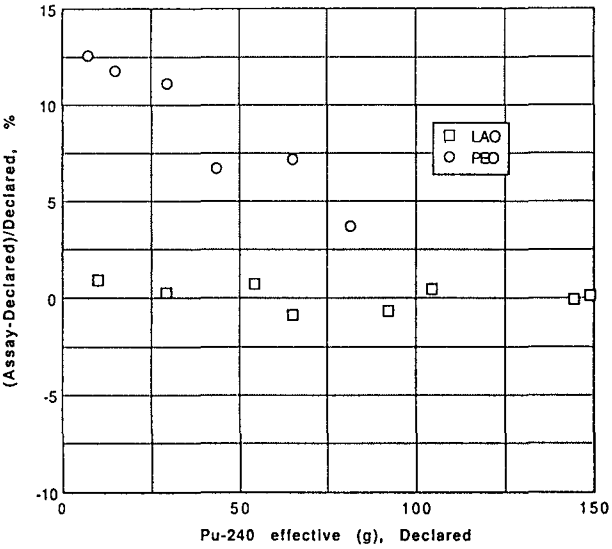

Fig. 13. Passive/Active NCC (passive mode) multiplication-corrected reals assay accuracies for pure (LAO) and impure (PEO) plutonium oxide.

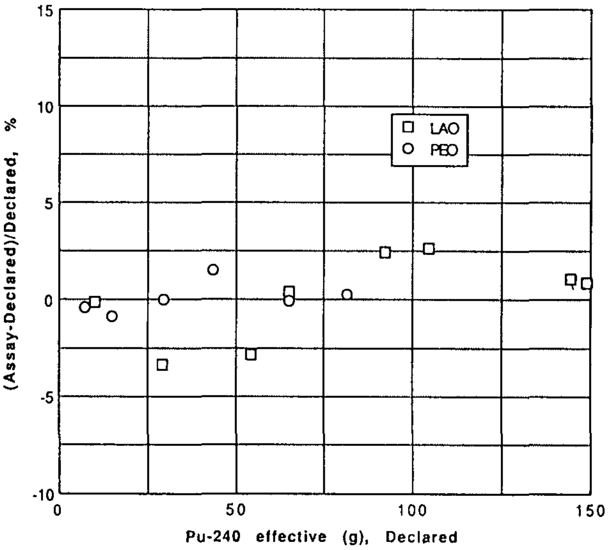

Fig. 14. Passive/Active NCC (combined passive and active-fast modes) multiplication-corrected reals assay accuracies for pure (LAO) and impure (PEO) plutonium oxide.

5. Conclusions

The new Passive/Active NCC is suited for general use in plants processing both uranium and plutonium. Optimum performance, safety, versatility, and reliability were primary goals of the development. The counter provides nondestructive assay of a wide variety of items containing plutonium, uranium, or both. Design, construction, testing, and initial evaluation have been completed. The instrument will be installed at the Los Alamos National Laboratory Plutonium Facility (TA-55).

By combining results of active and passive measurements, assay of impure plutonium-bearing items, and MOX samples can be accomplished. We have determined this new approach is capable of measuring the ^{240}Pu(eff) in impure plutonium-oxide to 2-4% in two 1000-s measurement periods. Also planned is an evaluation of combined passive/active measurements of plutonium-bearing residues and MOX. Initial data suggests the combined passive/active approach is a reasonable alternative to calorimetry and/or multiplicity counting for some impure plutonium-bearing items.

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NDA TECHNIQUES APPLIED TO A MAJOR TRANSFER OF NUCLEAR MATERIAL

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Abstract

Pu-containing materials of varied compositions have been transferred from what had been their various storage places for up to 25 years to a more central storage place. There was a requirement for independent NDA measurements to check that the material being transferred corresponded with the values recorded when the materials were first stored. The methodology devised for doing this operation is described together with the ways in which the various difficulties were dealt with.

1. Introduction

During a period of more than 20 years of research, development and manufacture of nuclear fuel a considerable quantity of plutonium bearing residues had accumulated at a laboratory in the UK. It has now been decided that the material, mainly of mixed Pu and oxides, but varying in nature from fuel plates to glove box sweepings, should be unpacked from its previous containers and repacked into stainless steel cans for transfer to centralised long term storage until they could be incorporated into future reprocessing campaigns.

The objective of the NDA measurements is to either confirm the 'book value' of the Pu or to recommend that a 'new value' be declared.

2. Background

Most of the material originates from fuel manufacturing campaigns during the 1960s and 1970s. It consists of pellets (whole or fragmented), powders, metals, alloys and various types of Pu-bearing wastes from the manufacturing processes. There are mixed oxides (MOX) in which plutonium is mixed with uranium in proportions varying from almost 100% Pu to much less than 1% Pu.

The records for the materials list the Pu and total weights corresponding generally to a date around the time of chemical analysis during the associated manufacture, a material order number (MO) which is expected to imply a common Pu isotopic composition, and other descriptive information.

Careful procedures were set up to determine the gross weight of material being taken out of the original containment and to compare this with the recorded book values. The material was then loaded into the long term storage cans, ensuring as far as possible that a can contained only one type of material. Each stainless steel can was assigned a book value equal to the sum of the fractions by weight of the original book values of its constituents.

During 1986 and 1987 a gamma spectrometer had been used to determine the isotopic composition of sample containers from most MO numbers. Analysis of these results indicated that several MO batches contained materials with significantly different isotopics.

3. NDA Measurements

3.1. Pu Mass by Passive Neutron Coincidence Counting

The body of the Passive Neutron Coincidence Counter (PNCC) used in the main series of measurements was manufactured by Jomar and utilised 18 Helium-3 tubes of active length 50.8 cm operating at 4 atm. The processing electronics, which used the 'shift register' technique were of Harwell design/1/ and manufacture, as was the software.

The PNCC measures the rate of spontaneous fissions, and since Pu240 is the main source of these, a value can be obtained for the Pu240 effective mass/2/. Knowledge of the isotopic composition of the Pu allows its mass to be obtained from the Pu240(eff) mass.

The PNCC gives values for the singles count rate (TOTALS) and the coincident count rate (REALS). A "non-multiplication"(NM) calculation can be made:

$$\text{Pu240eff} = \text{A0} + \text{A1}*\text{REALS} + \text{A2}*\text{REALS}*\text{REALS} \quad (1)$$

where A0, A1 and A2 are constants. This gives a reliable estimate for small non-multiplying samples-of-any-geometry, but larger samples (i.e. those with significant neutron multiplication) require calibration with samples of similar type, geometry and Pu density.

By using an inherently more precise and sample geometry-independent algorithm which uses both the TOTALS and REALS, a multiplication corrected REALS rate (MULT-CORR REALS) for the sample can be calculated. This algorithm requires, in addition to the information listed above, an estimate of the (α,n) production in the material. The α emission rate depends on the isotopics of the Pu, while a factor f in the calculation would be set at 0 for Pu metal and 1 for Pu oxide or MOX. The measured Pu240(eff) is given by a linear relationship, whose constants depend comparatively little on the type of standards used to determine them:

$$\mathrm{Pu240eff} = C0 + C1*(\mathrm{MC\ REALS}) \qquad (2)$$

Both NM and MC calibrations were made for the PNCC for the following materials:

Material Type	Pu240(eff)range (g)
Oxide Powder(PO)	25-200
Oxide Pellets(PE)	40-200
Very Small Oxides(VSO)	0.4-22

Since very few specimens had the Pu in metallic form no Metal calibration was made. However the mean of the oxide pellet and powder MC calibrations, but with constant f set of zero, appeared to work well.

3.2 Pu Isotopic Composition by Gamma Spectrometry

The Gamma spectrometer used for most of the measurements consists of a High Purity Germanium (HPGe) detector, 200 mm^2 in area and 10 mm thick, whose output is fed, via a Canberra 1510 Integrated Signal Processor to a Canberra Multi-Channel Analyser S100 board in an IBM PS/2 computer. Spectra are stored on hard disc for later analysis using the Multi-Gamma Analysis (MGA) code of Gunnink and Ruhter/3/. MGA uses the low energy, intense peaks from the Pu and is claimed to accuracies within 1% with measurement times of only a few minutes.

It should be noted that as the Pu242 cannot be measured using this technique the code incorporates the facility to either insert a value by hand or to calculate a value using internal algorithms.

3.3 Pu Mass by Calorimetry

The calorimeter was of an isothermal type similar to that developed by Mason et al/4,5,6/. This measures the heat evolved from the radioactive decay of the Pu isotopes and Am241. Calorimetry has the advantage that contaminants (e.g. oxygen, fluorine etc.) do not interfere with the measurements and uncertainty in the Pu242 content has negligible effect. However it is quite sensitive to the accuracy of the Pu238 measurement. Table 1 shows an example of the power from a typical sample.

Isotope	Wt.per. 100g of Pu	Power (mW/g of Isotope)	Specific Power(mw/ 100g of Pu)
Pu238	0.048	5.6716	0.273
Pu239	84.560	0.01929	1.631
Pu240	14.296	0.07098	1.012
Pu241	0.887	0.0339	0.030
Pu242	0.208	0.0011	0.000
Am241	1.314	1.1423	1.500

Table 1.

4. The Procedure for NDA Measurements

The initial vetting of cans was carried out with the PNCC because this required only about 10 minutes counting time, at least an order of magnitude less than that for a typical calorimeter measurement,

The PNCC measurement procedure requires the entry from the keyboard of the sample identifier, the calibration to be used (i.e. PE, PO, ME or VSO), the isotopic batch identifier, Pu and Am isotopic percentages with date of applicability and 'declared' Pu mass with date of applicability. From this data the 'declared' Pu240(eff) is calculated at the date of the PNCC measurement and printed. Following the measurements, the ratio (Measured Mass/Declared Mass) is computed for both the NM and MC calibrations.

The date of applicability of the 'declared' Pu mass is obtained from the records where possible, but otherwise it is set at one year after the date of chemical separation of Pu and Am which is calculated by MGA from the measured Pu241 to Am241 ratio.

The PNCC program running on the computer generates both a printed record and data files for each measurement. Another program produces a formal Assessment Form for the sample.

4.1 PNCC Acceptance Criteria

Because the accuracy to be expected for a PNCC measurement depends on various factors, each specimen was given the score appropriate to its rating on each of the attributes A to D in Table 2. These are recorded on the Assessment Form and added together to give a value S.

If the MC ratio for the measurement lies between 1-S and 1+S the PNCC is considered to have confirmed the Book Value, and the Assessment Form is signed to indicate this.

When the MC value lies outside the range a review procedure is set in motion. Frequently a γ ray measurement is carried out on the can to provide new isotopics for a recalculation of the PNCC measurement. If the MC ratio is still outside the acceptance limits a sample with high Pu content might then be measured calorimetrically, while for a low Pu can an NM ratio falling between 1-S and 1+S might justify acceptance of the book value.

5. Measurement Experience

The wide variety of the materials measured during the exercise raised quite a few points of interest.

5.1 Problems with Mixtures

The presence of materials from different MOs in the same can gives rise to problems. The first occurs if an original container of inhomogeneous material is divided among several cans. When repacked this can lead to an inaccurate value for the quantity of Pu in each new can. In addition if the isotopics for any components are in error the values for the whole can will be in error, and although spectrometer measurements were sometimes made at several places on such a can this was not very satisfactory as thorough mixing of the various components seldom occurred.

PNCC Scoring System

A. The Quality of Material.
  A1. Pu02 or MOX (Powders, Pellets or Spheres) Pu (Metal, Alloys). (Score 0.01)
  A2. Intermediate quality (Score 0.02)
  A3. Treated sludge, sweepings, ashed tissues etc. (Score 0.05)
B. Chemical Quality of Material
  B1. Pu02, MOX or Pu metal with no extra major alpha-n (Score 0.01)
  B2. Pu02, MOX or Pu metal with suspected extra alpha-n, Pu nitrate or carbide etc. (Score 0.03).
C. Quality of Isotopic Information.
  C1. Several samples measured, no evidence of inhomogeneity. (Score 0.01)
  C2. Several samples measured, evidence of inhomogeneity, (Score 0.05).
D. Quality of Fit to Calibration.
  D1. 15g < Pu240eff < 300g (PO, PE or ME calibration used). (Score 0.01)
  D2. 1g < Pu240eff < 15g (VSO calibration used). (Score 0.02)
  D3. Pu240eff < 1g (VSO calibration used) or Pu240eff > 300g (PO, PE or ME calibration used. (Score 0.05)

Table 2

5.2. Calorimetry

Calorimetry is very effective for the precise measurement of Pu largely because its result does not depend on the chemical or physical form of the samples. However it cannot compete with the PNCC in terms of measurement speed. For example, a can containing of the order of a kilogram of Pu and little else could be measured in an hour or so but a MOX sample with low Pu concentration would need to be left in the calorimeter for many hours. For this reason the calorimeter was mainly used to resolve cases where there was a possibility of a sizeable deviation from the book value.

A problem occurred in the use of the calorimeter due to its unavoidable location in a environment with wildly fluctuating mains supply, and it was not unusual for argon arc welders to be operated in the vicinity, causing the instrument to trip out. The problem was partially cured by the fitting of electrical filters but needs further attention.

The calorimeter has a prediction routine which forecasts the equilibrium result. A project is in hand to improve the algorithm of this so that measurement times may be reduced.

5.3 Uncertainties Due to Isotopics

Although modern mass spectrometry and gamma spectrometry usually give good results there are a number of ways in which isotopic errors contribute to the scatter of the ratios measured in this exercise.

Uncertainties in the isotopic percentages will of course contribute to the uncertainties in Pu measurements. For the PNCC for example the measurement uncertainty in the Pu240 fraction will propagate almost directly into the PNCC results. In addition, because the Pu242 is not measured in gamma spectrometry its estimation will initially contribute an error although this is unlikely to be as great as 1% of the Pu mass for any of the materials in the exercise.

Calorimetric measurements have the advantage that virtually all the heat comes from isotopes that can be measured by gamma spectrometry, but Table 1 indicates that errors in determination of Am 241 will be particularly significant.

5.4 A Problematic Batch of Cans

There were two groups of cans containing material involving stainless steel (S/S) with the same MO number but labelled 'Plates' and 'Powders' respectively. Although the PNCC ratios for the Plates were all close to unity, those for the Powders showed much more variation and a mean of about 0.8. After confirming the isotopics and realizing that there were too many cans of Powder for calorimetry, the first approach was to use the first 15 cans of Powders to produce a new "incestuous" calibration for the PNCC. When this was used to recalculate the measurements for the whole batch of cans most of the ratios came between about 0.9 and 1.1.

Although this showed that the problem with the powders was systematic it did not explain it. An enquiry among long-serving staff suggested that the Plates had been made by sintering with S/S powder sub-millimetre diameter balls of Pu oxide that had first been coated with adhesive and then stirred in S/S powder. Some of the balls had broken in the process, gradually clogging up the S/S coating system, and had been disposed of as 'Powder'. If this were true there should be far more Pu atoms in the powder with S/S component atoms as closest neighbours than in the plates, and the (α,n) yield from the Powder should be less than from the Plates. The MC calculation of the PNCC uses the (α,n) yield multiplied by factor f, where f is normally set at unity for Pu in an oxide environment. Changing the value of f can allow for environments giving different (α,n) yields. Trial and error led to giving f a value of 0.716 for the Powders. Figure 1 is a plot of the resulting MC ratios against Pu Book Value for 45 cans of Powder. It will be seen that the MC results have been brought reasonably close to unity over the Pu mass range. A calorimetric measurement of one can was carried out to help justify the procedure. It somewhat fortuitously gave a ratio of 0.962 for a can whose PNCC ratio was 0.955 using the chosen f value.

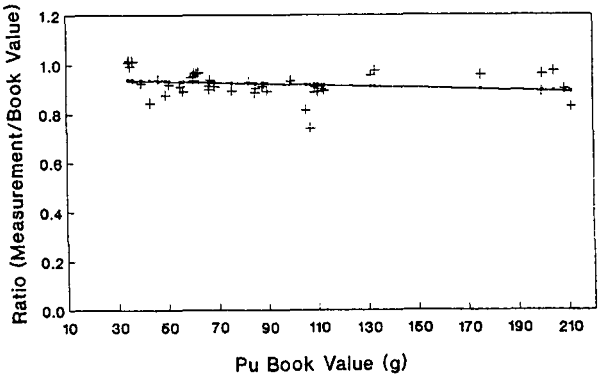

Fig.1. PNCC ratios of (Pu Mass/Book Value) for 43 'Pu Stainless Steel Powders' plotted against Pu Book Value.

5.5. Comparison of PNCC and Calorimetry

Fig.2 shows as * the ratio (Measured Pu mass/Book Value) for each of the Calorimeter measurements plotted against Pu Book Value. In all the cases where the PNCC produced MC ratios these are plotted as +.

8 of the 16 Calorimeter Measurements can be seen to have ratios within 2% of the Book Value suggesting that the Calorimeter is giving results to within 2% accuracy.

Calorimetric ratios significantly away from unity are believed to indicate errors in the Book Values.

In general the PNCC tends to support the Calorimeter ratios but gives less close agreement with Book Values that are believed to be good. In addition, the PNCC had failed to give an MC value at all for 3 of the cans.

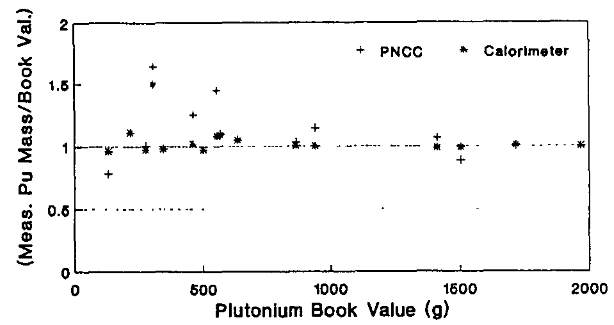

Fig.2. Ratios of (Pu Mass/Book Value) obtained with the Calorimeter and the PNCC plotted against Pu Book Value.

6. The Results of the Measurements so far

A total of 351 cans of wastes have been measured so far. A recommendation to use the Book Value has been made for 290 of the cans, New Values have been recommended for 4 of the cans, a Batch of 48 cans is still awaiting checking by calorimetry, and 12 other results are still under review.

Using the methodology of section 4.1 the Acceptance Range for the measurements varied from can to can, but the average was 0.93 to 1.07. Of the decisions to use the Book Value:

163 were based on 161 MC results of the PNCC falling within Acceptance Range and 2 of them falling just outside narrow ranges.

25 were based on NM results of the PNCC falling within the Acceptance Range.

9 were based on Calorimeter measurements.

70 were the Pu Stainless Steel Powders discussed in Section 5.4.

13 were a batch of cans, with material of common origin, giving some MC results outside the Acceptance Range but with a ratio of 1.02 for the batch.

7 cans without an acceptable PNCC result but for which the amount of discrepancy did not justify further effort.

7. Conclusions

This exercise appears to be fulfilling its intention quite well but there have been some problems and some successes. This experience allows a list of recommendations to be drawn up for any future operation of this type:

(a) History All possible information on the material should be obtained in advance of the measurements.

(b) Preparation If at all possible isotopic measurements should be carried out before material has been removed from its original storage.

A scheme for recording the results and for judging the acceptability of book values should be set-up before measurements begin. The measuring instruments should be tested in advance at the actual place of measurement and under the working conditions that are likely to occur.

(c) Flexibility A PNCC cannot give assurance on all types of residues; a Calorimeter provides very useful information in difficult cases. Gamma spectroscopy can check anomalies in the isotopics of batches of material.

Even with a preset set of rules there is considerable room for judgement in deciding when New Values are needed.

The progress so far with this exercise has only been possible with the help of many people, to whom our thanks are offered.

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A COMPACT K-EDGE DENSITOMETER FOR URANIUM

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Abstract

A compact K-edge densitometer has been designed for use by safeguards inspectors to verify the uranium concentration in product solutions from a reprocessing plant. The densitometer is based on multi-energy transmission measurements near the uranium K-absorption edge using a single isotopic source (^{57}Co) in conjunction with a uranium converter foil. The compact set-up is easily coupled to standard high-resolution spectroscopy systems. The measurement accuracy achieved in a counting time of 1000 seconds is about 0.2-0.3 % at concentration levels typical for product solutions.

An approach to determine the ^{235}U enrichment from aliquots of less than 5 ml with an accuracy of better than 1 % within the same measurement time is also discussed.

1. Introduction

Both the uranium concentration and the ^{235}U abundance in uranium product solutions are of interest for safeguards-inspection purposes. Two NDA instruments are described in this paper that allow the measurement of these parameters with due regard to inspection specific conditions.

A simplified version of a K-edge densitometer has been developed for use by safeguards inspectors to verify the uranium concentration in product solutions from a reprocessing plant. The densitometer meets the following design goals:

- to be easily adaptable to existing portable safeguards spectroscopy equipment,
- to keep the required additional hardware, including the radiation sources, at a minimum,
- to reduce the required sample sizes to small volumes,
- to provide simple and reliable operation for field applications, and
- to ensure a measurement accuracy of better than 0.5 % at U concentrations typical for product solutions in counting times of about 1000 s.

The assay sytem for the ^{235}U abundance is based on a passive gamma measurement of the characteristic radiation emitted from the decay of ^{235}U. Similar design criteria will also apply to this instrument. The envisaged measurement accuracy in this case was

- 1 % relative for ^{235}U enrichments >0.7 % within the same counting time of 1000 s.

The evaluation of the ^{235}U enrichment in small samples from passive gamma measurements requires the U element concentration as input value which in turn is determind by the aforementioned K-edge densitometer assay. Hence the requested result - here the ^{235}U abundance - is determined from the combination of two different NDA methods.

2. K-egde densitometer

Principles

K-edge densitometry makes use of the element-specific discontinuity of the photon absorption cross section that occurs when the photon energy exceeds the binding energy of the electrons in the K-shell of the particular element. In case of uranium the K-edge jump is found at the energy of 115.6 keV (see bottom part of Fig. 1). From photon transmission measurements T_l and

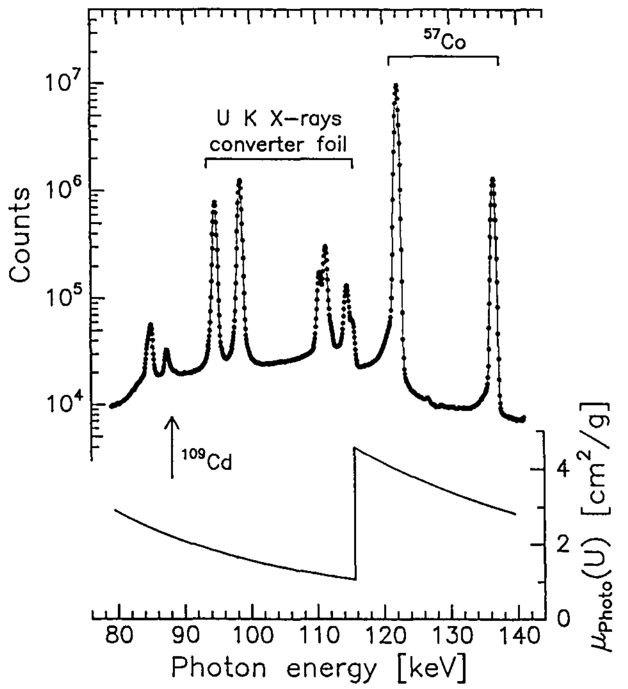

Fig. 1: Interrogating photon spectrum of the compact K-edge desitometer (top) and mass attenuation coefficient of uranium (bottom) versus photon energy.

T_2 below and above the K-edge, respectively, one gets the uranium concentration ρ_U in the sample by

$$\rho_U = \frac{ln(T_1/T_2)}{\Delta\mu \cdot d} \qquad (1)$$

where $\Delta\mu$ is the difference of the uranium mass-absorption coefficient at the energies of the interrogating photons below and above the K-edge, and d is the pathlength of the beam through the sample cell.

The element-specific effect is largest and the sensitivity to variations of the sample matrix is minimized if the energies of the interrogating photons tightly bracket the energy of the U K-edge. The problem is to find suitable photon sources that fulfill this condition. Two approaches are mentiond here.

1. The Hybrid K-edge densitometer /1/ that uses a continuous photon spectrum around the K-edge from the bremsstrahlung of an X-ray tube.

2. The Los Alamos/Tokai K-edge densitometer /2/ for plutonium. It utilizes discrete gamma lines from two different radioactive sources in a revolving source holder. The energies of 121.1 keV (^{75}Se) and 122.1 keV (^{57}Co) tightly enclose the plutonium K-edge energy of 121.8 keV.

This favorable situation is not given for U K-edge densitometry due to the lack of suitable gamma lines below the U K-edge. The 88 keV line of ^{109}Cd (indicated by an arrow in Fig. 1) could be used along with the 122.1 keV line of ^{57}Co in a double-source K-edge densitometer for uranium. However, the wide energetic distance of the former gamma line to the U K-edge leads to a reduced instrument performance.

The approach realized in the present compact K-edge densitometer was proposed some time ago /3/. It uses multi-energy transmission measurements by means of a single radioisotopic source (^{57}Co), which in turn is used to generate additional radiation in the form of K-X rays in a uranium converter foil located in front of the ^{57}Co source. In this way 5 different photon energies - the X-rays at 94.6 keV ($K_{\alpha 2}$), 98.4 keV ($K_{\alpha 1}$), 111.3 keV ($K_{\beta 1,3}$) of uranium and the ^{57}Co gamma rays at 122.1keV and 136.4 keV - are available for transmission measurements below and above the U K-absorbtion edge energy (see Fig. 1)

The K-edge densitometer adapter

A small mechanical assembly has been build that contains the ^{57}Co source (~1-5 •10^8 Bq), the uranium converter foil (~ 0.2 g/cm^2 of depleted uranium), the beam collimators, the cavity for the sample vial and some shielding parts. In order to save weight this compact K-edge adapter is made of aluminium and plastic materials, only the beam guiding parts and the radiation shielding are made of tungsten. Fig. 2 gives a simplified schematic view of the assembly. The essential parts of the adapter have a total length of 5 cm only.

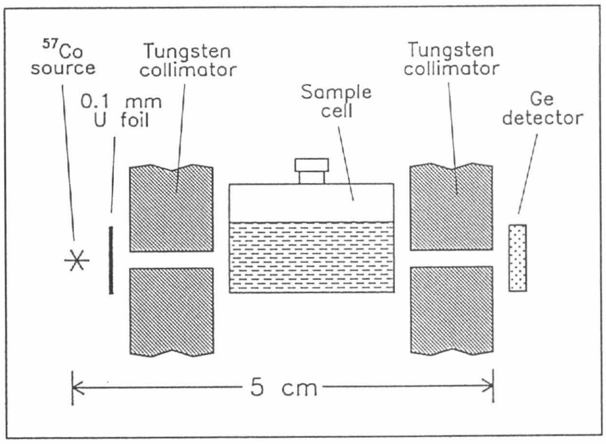

Fig. 2: Schematic view of the compact K-edge desitometer.

Towards the detector the assembly is extended by a guiding tube that is slipped over the detector endcap. The inner plastic part of the guiding tube is exchangeable to allow the adaption to detector caps with varying diameters. It is also equipped with a small clamp for fixing the K-edge adapter to the detector head.

The overall dimension of the compact K-edge adapter is about 15 cm x 8 cm ø, its weight amounts to less than 800 g. Therefore no stands or supports are required. Almost any Ge detector with vertical endcap can easily be converted to a K-edge densitometer simply by plugging the adapter to the detector´s endcap and fixing it by the clamp.

The cavity of the densitometer accepts one standard high-precision glas cell with 2 cm length, no provision is forseeen for an automated sample changer.

The photograph in Fig. 3 shows the K-edge densitometer adapter plugged to a standard Ge detector. The source shielding in front of the adapter, the sample cavity with a U sample and the guide tube with clamp can be identified.

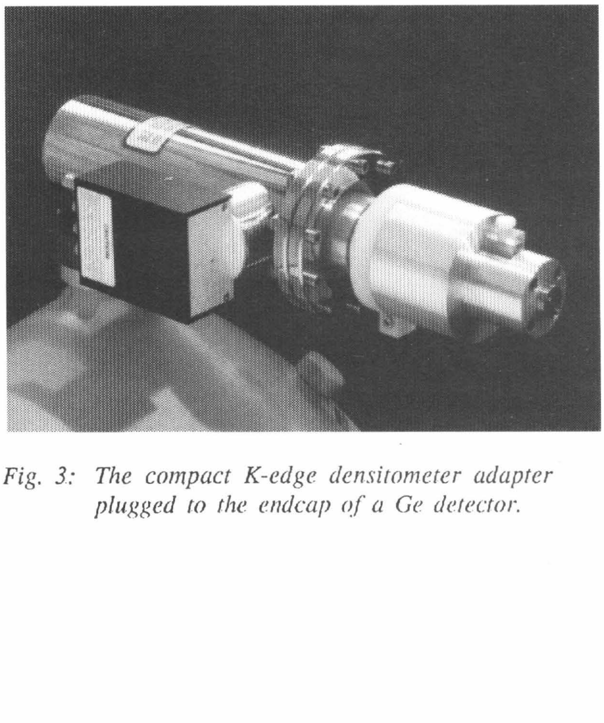

Fig. 3: The compact K-edge densitometer adapter plugged to the endcap of a Ge detector.

Sample fluorescence effects

In the compact K-edge densitometer described here the ^{57}Co lines and the U K X-rays generated in the uranium converter foil are used as interrogating photons. When Uranium is analysed in this set-up - as we do - then the ^{57}Co radiation will also introduce K X-ray fluorescence in the uranium sample material. This sample fluorescence adds to the interrogating U X-ray lines used for the determination of the photon transmission, and thus disturbs the assay result. This effect has been investigated by substituting the uranium converter foil by a lead foil exhibiting approximately the same attenuation for the ^{57}Co gamma rays.

The results of these measurements are given in Fig. 4. The percentage contribution of the sample fluorescence to the total observed peak counting rate in the U $K_{\alpha 2}$ and U $K_{\alpha 1}$ lines is shown in Fig. 4a for two different collimator geometries as a function of the uranium concentration in the sample. "Narrow" collimation stands for both the primary collimator (between source and sample) and the secondary collimator (between sample and detector) with 10 mm length and 3 mm diameter. In case of the "wide" collimation the diameter of the primary collimator was enlarged to 4 mm.

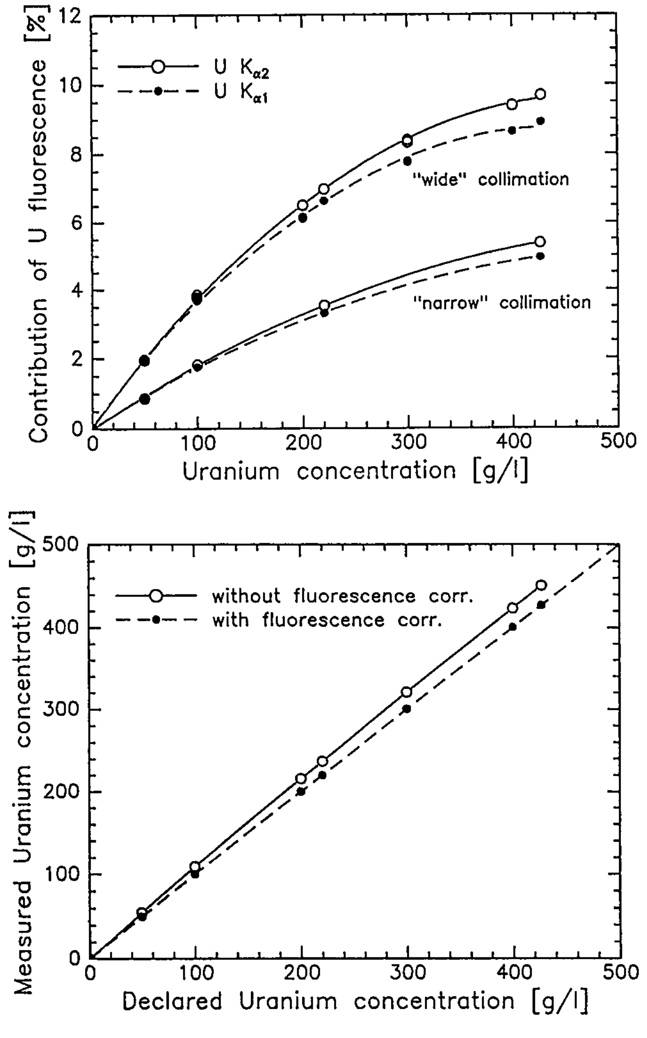

Fig. 4: Effect of sample fluorescence: a) % contribution to interrogating uranium K-X ray peaks, b) instrument response without and with sample-fluorescence correction (using same $\Delta\mu$).

Fig. 4a demonstrates that the sample fluorescence effect critically depends on the collimation used. For the "narrow" collimation it is about a factor of two smaller than for the "wide" one. Therefore, when using an extremely high collimated beam the sample fluorescence effect can be brought down to a negligible level. This is, for example, realized in the Hybrid K-edge densitometer /1/ where the solid angle of the collimation is about two orders of magnitude smaller than in the present case. This extreme narrow-beam approach however is not applicable to the compact K-edge densitometer due to the limited source strength of commercially available ^{57}Co sources. The collimation selected is therefore a compromise between the desired high counting rate of the interrogating radiation to arrive at short measurement times on one hand, and an acceptable distortion by the sample fluorescence on the other hand. In the present set-up the collimation described above as "narrow" collimation has been implemented resulting in a maximum sample fluorescence contribution of about 6 % to the U $K_{\alpha 2}$ peak at high uranium concentrations. Note that the effect slightly decreases with increasing energy of the U K X-rays.

The impact of the sample fluorescence on the assay result as a function of the U concentration is shown in Fig. 4b for the most unfavorable case of Fig. 4a (U $K_{\alpha 2}$ peak, "wide" collimation). The curves have been evaluated according to eq. 1 without and with sample-fluorescence correction using the same $\Delta\mu$ value in both cases. The sample fluorescence appears as positive bias that increases nonlinear with the U concentration.

The sample fluorescence for a given collimation geometry is a unique function of the U concentration. Once the effect is known and can be parameterized, a nonlinear logarithmic function of the U concentration ρ_U can be added to eq. 1. This leads to an implicit function of ρ_U that is solved by iteration.

A more practical approach is the piecewise linear interpolation of the slightly non-linear instrument response function. An "effective" $\Delta\mu$ value in eq. 1 has to be established from the measurement of suitable calibration standards that enclose the U concentration range of interest. The "effective" $\Delta\mu$ value is then valid only for this limited range of uranium concentrations.

Sample matrix effects

In K-edge densitometry the transmission values required for the evaluation of the U concentration from eq. 1 are obtained in general by the measurement of both the sample with the unknown uranium nitrate solution and a reference nitic acid solution with a HNO_3 molarity identical to that of the U sample under assay.

In this way the influence of sample matrix effects is widely eliminated.

However, if the composition or the density of the matrix in the uranium sample and in the reference sample are different, a small positive or negative bias will be observed in the assay result depending on the difference of the matrix composition in both samples. This is due to the fact that the mass attenuation coefficient of the sample matrix is not constant over the energy range of the interrogating photons. Fig 5 shows the virtual uranium concentrations simulated by different sample matrix compositions. The simulated U content is given for 1 mol, 3 mol and 8 mol HNO_3 samples and is evaluated for the photon pairs 95 /136 keV and 98 /122 keV, respectively, used for the determination of the transmission below and above the U K-edge. The 3 mol HNO_3 sample serves as reference, the values are taken from repeated measurements of the acid samples with good statistical precision.

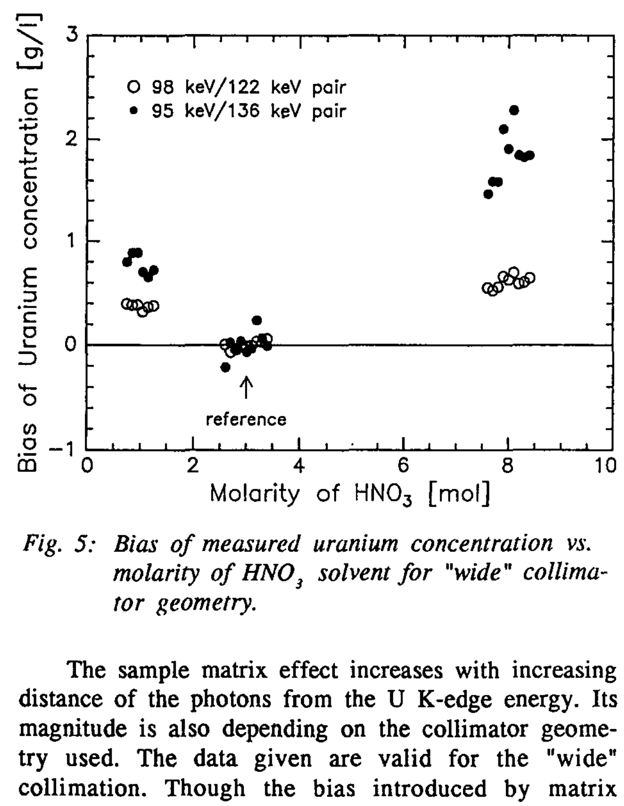

Fig. 5: Bias of measured uranium concentration vs. molarity of HNO_3 solvent for "wide" collimator geometry.

The sample matrix effect increases with increasing distance of the photons from the U K-edge energy. Its magnitude is also depending on the collimator geometry used. The data given are valid for the "wide" collimation. Though the bias introduced by matrix variations is small in case of product solutions with high U concentrations and generally low and almost constant acidity of the solvent a verification of the matrix effect and a possible correction for it from the known molarity of the samples under assay is recommendable if ultimate measurement accuracy is desired.

Measurement precision and accuracy

Fig. 6 shows the relative deviations of the K-edge densitometric assay results from the declared values of the DA chemical analysis. The results are given for the "narrow" collimation geometry. Four samples with uranium concentrations of 50 gU/l, 100 gU/l, 220 gU/l and 427 gU/l in 3 mol HNO_3 solution have been analyzed. The values shown are taken from a series of measurements to determine the optimum thickness of the U converter foil.

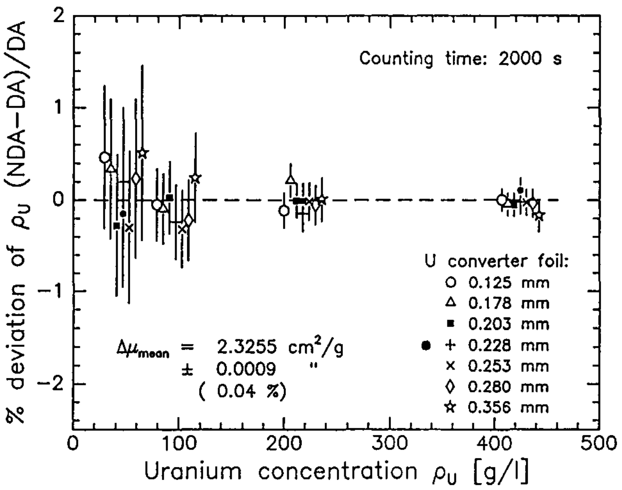

Fig. 6: % deviation of measured U concentration from declared DA values versus U concentration.

The data evaluation was performed according to eq. 1 with prior correction for sample fluorescence. The figure demonstrates an excellent linearity of the response of the assay system over a wide range of U concentrations after proper correction for sample fluorescence. The error bars given represent the measurement precisions achievable in 2000 s counting time. The calibration constant $\Delta\mu$ for the transmission pair 98/122 keV obtained from these measurements is also given in the figure along with its associated error at the level of 1 standard deviation. The evaluation of other transmission pairs from the multi-line interrogating radiation will definitely improve the precision of the assay result.

From the measurements we conclude that the target accuaracy of better than 0.5 % in 1000 s counting time for U product solutions can be met. The observed precision for 400 gU/l samples was better then 0.2 % within this counting time for a single transmission-pair evaluation (98/122 keV). Reliable values for the achievable accuracy of the compact K-edge densitometer cannot be given at present because only a limited number of samples have been analyzed (15 in total). The planned field test will give more valid information about the instrument performance under field conditions.

3. ^{235}U abundance assay

Principles

The ^{235}U abundance in uranium solutions can be determined by passive gamma-ray measurements. During the decay of the radioactive ^{235}U isotope characteristc gamma lines are emitted. A typical gamma- and X-ray spectrum observed from natural uranium is shown in Fig. 7. Four strong gamma lines with energies at

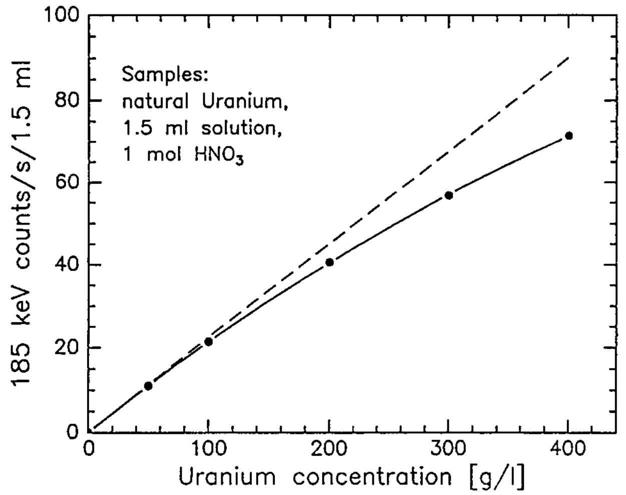

Fig. 9: 185 keV counting rate versus U concentration for 1.5 ml natural uranium solution.

for the attenuation of the 185 keV radiation remain unchanged.

The registered 185 keV counting rate is of course a function of the total uranium concentration. Fig. 9 shows the 185 keV counting rate observed from 1.5 ml natural uranium in 1 mol HNO_3 solution at various concentration levels. The deviation from linearity is caused by the increasing self-attenuation effect with increasing uranium concentration in the sample. This nonlinearity necessitates a careful calibration of the system versus the U concentration.

It can also be deduced from Fig. 9 that for 1.5 ml of a 400 g/l solution of natural uranium - which is a typical value for product solutions - a 185 keV counting rate of about 70 cps is expected. Thus a counting precision of better than 0.5 % is achievable in 1000 s counting time.

The gamma counting rate of interest is also affected when the sample matrix composition and density changes between measurements. This effect has been investigated by determining the instrument response for pairs of samples with equal uranium concentration but diffent matrix molarity: 1 mol / 8 mol HNO_3 and 4 mol / 6 mol HNO_3, respectively. The matrix attenuation of the 185 keV line given in units of (% per mol) at various U concentration levels is displayed in Fig. 10. The effect is small - about -0.2 % per mol difference at all U concentrations. For strongly varying matrix and highly accurate assays this mol-correction is recommended.

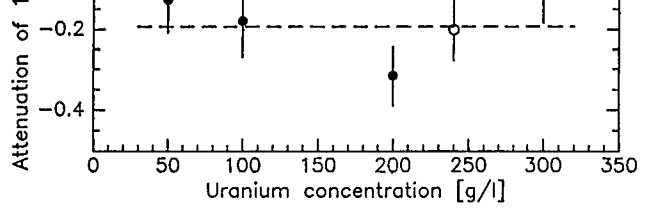

Fig. 10: Attenuation of 185 keV photons in a 1.5 ml uranium solution per mol HNO_3 versus U concentration.

Another crucial parameter is the sample volume in the measurement cell. The sample liquor has to be very carefully filled into the test tube using a high-precision pipette. Any volume variation directly proceeds into the assay result. For a sample volume of 1.5 ml and and envisaged accuracy of better than 1 % the pipetting error should not exceed about ± 5 - 10 µl.

Calibration

The relation between observed 185 keV counting rate N and the ^{235}U abundance Enr in its simplest form is given by

$$Enr = \frac{N}{(a \cdot \rho_U - b \cdot \rho_U^2) \cdot (1 - c \cdot (mol - mol_{ref}))} \qquad (2)$$

where a, b and c are calibration constants, ρ_U is the uranium concentration (value from the compact K-edge measurement), mol and mol_{ref} are the HNO_3 molarities of the sample under assay and of the reference sample, respectively.

At least three calibration samples are required to establish the constants in eq. 2. Two samples CAL1 and CAL2 with identical matrix and ^{235}U abundance, but with different U concentration are needed to evaluate the constants a and b in eq.2. A third calibration sample CAL3 is required to determine the mol-correction constant c. It should exhibit the same U concentration and ^{235}U abundance as one of the samples CAL1 or CAL2, but the HNO_3 molarity should be significantly different. The use of natural uranium is recommendable for all three calibration samples.

^{237}U interference

The ^{237}U isotope is always present in freshly separated reprocessed uranium. Since its predecessor ^{241}Pu is removed during the separation process ^{237}U will die away with a half life of 6.75 days. ^{237}U emits two strong gamma lines within the energy region of interest for ^{235}U assay with energies at 164.6 keV and 208.0 keV. There is no direct interference expected for high resolution gamma detection systems because these gamma lines are well separated from the 185.7 keV line from ^{235}U used for the enrichment assay. A problem however may arise from the high activity of ^{237}U for freshly separated high-burnup uranium. No such type of material was available for testing, therefore we can give only an estimate for the expected effect.

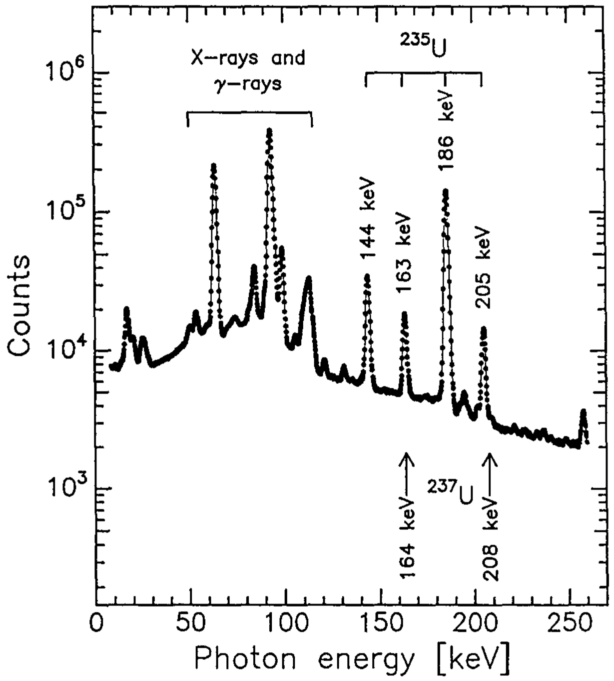

Fig. 7: Passive gamma- / X-ray spectrum of 1.5 ml natural U solution measured with a Ge well detector.

144 keV, 163 keV, 185.7 keV and 205 keV serve as a fingerprint for the presence of ^{235}U. The strongest line at 185.7 keV is normally used for ^{235}U abundance determinations. Due to the lack of suitable gamma lines from the major isotope ^{238}U near to the energy region of the characteristic ^{235}U gamma rays a direct ratio measurement is not possible. Instead the absolute number of 185 keV gammas emitted from the sample per second is determined that is a direct measure of the number of ^{235}U atoms present in the sample. In practice the 185 keV counting rate before registered in the detector is subject to various attenuation effects and has to be folded with the detector efficiency. These effects will be discussed in the following subsections.

Equipment

From the inherently low emission rates of the 185 keV radiation in low enriched uranium it is easily verified that the ^{235}U enrichment determination, when performed on a few-ml samples with standard planar or coaxial Ge detectors, will yield poor counting statistics and precision within acceptable counting times. Counting rates as low as about 10 cps have been observed from 5 ml of natural uranium solutions.

However, the measurement situation significantly improves when the samples are counted in a well-type detector exhibiting approximately a 4π geometry. A schematic view of an assay system using a Ge well detector is given in Fig. 8.

The detector used in the present exercise had a well diameter of 14.5 mm, the total depth of the well was 35 mm, the active zone inside the detector was 22 mm deep. The sample cells for the uranium liquor were standard test tubes with an outer diameter of 11.5 mm and an inner diameter of 9.8 mm. In order to reduce the influence of small variations of the test-tube dimensions the whole sample volume should be kept inside the active detector well as indicated in Fig. 8. In the present counting set-up this condition leads to a maximum recommended sample volume of about 1.5 ml.

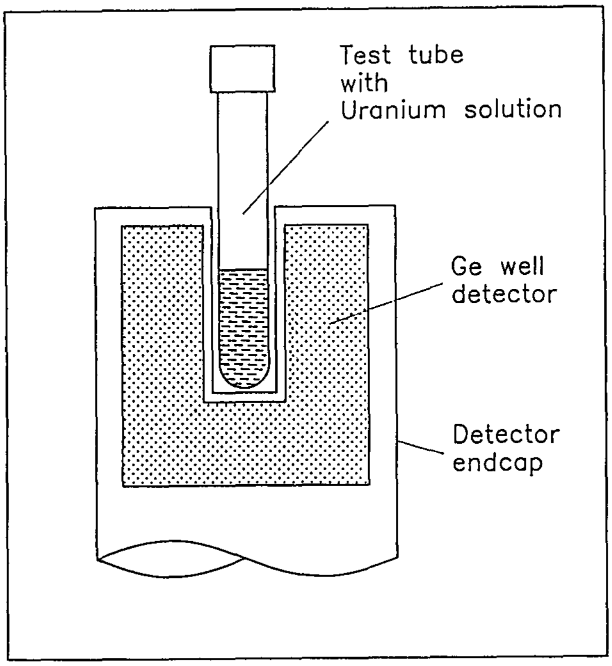

Fig. 8: Schematic view of the ^{235}U enrichment assay system.

Crucial parameters

The observed 185 keV gamma counting rate used for the determination of the 235U abundance depends on a number of parameters that influence the assay result which are listed below:

- ^{235}U isotope abundance
- total U concentration
- matrix composition and density
- sample volume
- – sample geometry (diameter, form of test tubes)
- – wall thickness of test tubes
- – position of the sample
- – detector efficiency

The last four factors marked by a minus sign can be kept well under control by using always the same type of well specified test tubes, by a careful design of the sample-cavity adapter and by calibration, respectively.

The first parameter in the above list is the wanted quantity. Keeping all other parameters constant and varying only the ^{235}U abundance will result in a strictly linear response of the assay system as a function of the ^{235}U abundance, because all other factors responsible

In the worst case of freshly separated high-burnup uranium the radioactive decay rate of ^{237}U may exceeds that of ^{235}U by a factor of 180. The 208 keV peak is the about 70 times higher than the 185.7 keV peak of interest. The more stringent problem is the total detector counting rate. From a total counting rate of about 1000 cps for a 1.5 ml natural uranium solution with 400 gU/L registered in our experiment we extrapolate to a total detector counting rate of more than 100 kcps for the above mentioned worst case material. This excessively high counting rate can hardly be handled by conventional counting electronics without serious degradation of the system performance.

Possible countermeasures are either the reduction of the sample mass at the cost of longer counting time or the insertion of a suitable absorber into the detector well to reduce the low energy gamma- and X-rays that contribute a significant part to the total counting rate (see Fig. 7). If any or which of these countermeasures has to be taken depends on the conditions found in the field test. Two weeks after separation the total counting rates will reach an acceptable level in any case.

Acknowledgements

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THE USE OF THE HYBRID K-EDGE DENSITOMETER FOR ROUTINE ANALYSIS OF SAFEGUARDS VERIFICATION SAMPLES OF REPROCESSING INPUT LIQUOR

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Abstract

Following successful tests of a hybrid K-edge instrument at TUI Karlsruhe and the routine use of a K-edge densitometer for safeguards verification at the same laboratory, the Euratom Safeguards Directorate of the Commission of the European Communities decided to install the first such instrument into a large industrial reprocessing plant for the routine verification of samples taken from the input accountancy tanks. This paper reports on the installation, calibration, sample handling procedure and the performance of this instrument after one year of routine operation.

1. Introduction

In November 1989 the new French reprocessing plant UP3 started operations. During detailed discussions held over a period of several years before plant start-up between its operator, Cogema, the French government and the Euratom Safeguards Directorate on the implementation of safeguards at this facility it was agreed to install a Euratom owned and operated hybrid K-edge densitometer for independent verification measurements of samples of input and process liquors. The number of such samples was estimated at approximately 250 per annum, the target accuracy was set at 1 %.

Traditionally this type of sample has been sent off-site for chemical analysis at one of the European Commission's Safeguards Analytical Measurement (ECSAM) laboratories. Transport of these samples requires sample conditioning such as dilution, spiking and drying of weighed aliquots by operator's staff under observation by the inspectors. This conditioning procedure thus requires operator's and inspector's resources, increases their dose rate, reduces the inspector's confidence in the authenticity of the sample, and it may introduce additional sources of error reducing the overall accuracy of the verification measurement. Additionally, transport of nuclear materials, albeit in very small quantities, is subject to increasing problems with licensing, shipment approval and public acceptance. This may lead to substantial delays between the date of sampling and the date at which the analytical results reach the inspectors. Thus the goal of timeliness may not be met, and follow-up in case of differences may be difficult if not impossible. Finally, transport of these samples has become very costly.

Following successful tests of a hybrid K-edge instrument at the European Commission's Joint Research Centre Institute for Transuranium Elements, and the routine use of a K-edge densitometer for safeguards verification measurements at the same laboratory, the Euratom Safeguards Directorate decided to implement this instrument for the routine verification of samples of input and process liquors in the large new reprocessing plants.

2. System Description

Principal Layout

The principal instrument layout of the first infield installation at UP3 is similar to the configuration of the prototype system, which has been successfully tested at the European Institute for Transuranium Elements, Karlsruhe /1/.

The combined K-edge densitometry (KED) and X-ray fluorescence (XRF) measurements for the uranium and plutonium assay in the highly radioactive dissolver solutions require a compact and efficient counting geometry, a high-intensity photon source, and tight beam collimation for both measurement channels. The standard counting configuration of the Hybrid Instrument is shown in Fig. 1. The required primary radiation - a photon continuum for KED at very high intensity for both XRF and KED - can only be obtained from a powerful X-ray unit.

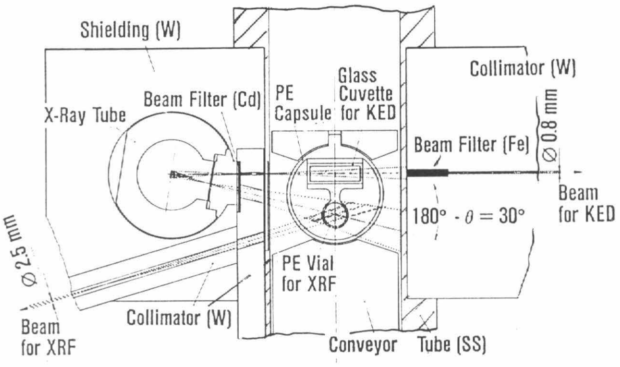

Fig. 1 : Principal counting geometry adopted for the Hybrid Instrument.

The previous concept of using two separate sample vials for the measurements - a glass cuvette for KED and a PE vial for XRF - is also applied in the present installation. An improvement compared to the previous prototype system has been achieved by reducing the distance between the focal spot of the X-ray tube and the samples from 8 cm to 6.5 cm. This modification further reduces the sensitivity to the self-radiation from the input solutions.

Mechanical Installation

For operation with undiluted input solutions the Hybrid Instrument must be installed next to a well-shielded box for sample handling. The coupling of the instrument to any type of shielded box is usually achieved by means of a rigid and tight tube of stainless steel, in which the samples are transferred from the interior of the box to the instrument outside the containment. This principal configuration, which maintains and ensures a safe sample containment at any time of operation, and which leaves the complete equipment outside the box in an accessible area, greatly facilitates its installation and adaptation to existing shielded facilities.

A suitable place for instrument installation at UP3 was identified in the HA laboratories, where a series of shielded boxes are located. The boxes are utilized by the operator for part of his analytical work on the input solutions. The given boundary conditions at the proposed location offered the possibility to design a relatively simple and compact mechanical set-up for instrument adaptation to the backside of one of the shielded boxes.

The actual mechanical assembly of the Hybrid Instrument at UP3 is shown in Fig. 2. The stainless steel tube seen on the photograph, about 60 cm long with an outer diameter of 8 cm, is dimensioned to fit into an existing flange of the box. The adaptation to the box is indicated on the plan of installation given in Fig. 3. The mechanical assembly is mounted on a small table right at the periphery of the box shielding. It is surrounded by a layer of 10 cm of lead to provide shielding against the high-energy gamma radiation from the input solutions.

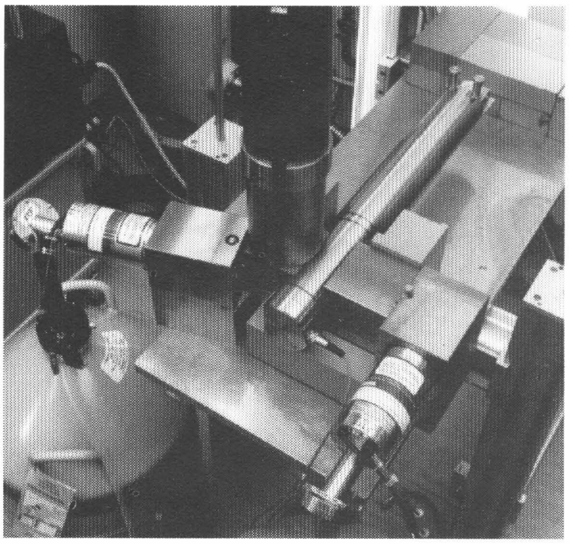

Fig. 2 : Photograph of the mechanical assembly prior to installation.

A conveyor inside the stainless steel tube carries the samples from the loading position inside the box to the position of measurement in the instrument. The displacement of the conveyor over the relatively short distance of about 50 cm is done manually. A micro-switch is actuated when the sample conveyor has reached its correct position for a measurement.

Equipment

The different instrument components required to run the Hybrid Instrument are commercially available units. The block diagram in Fig. 4 identifies the components, how they are interconnected, and where they are located at the site of installation.

The equipment located behind the shielded box comprises the mechanical set-up, two planar HpGe-detectors (200 mm^2 x 10 mm) for XRF and KED and the X-ray tube with associated power modules. The X-ray tube - a metal-ceramic tube with tungsten target - is operated at 150 kV/15 mA, which corresponds to about 75% of its maximum power rating of 3 kW. An air-cooled water circulator provides the cooling for the X-ray tube.

The equipment located in the working area in front of the shielded box comprises an electronics cabinet containing the NIM modules for the detectors, the X-ray control unit and the micro-switch control unit. The NIM modules for the two detector systems are contained in a single NIM bin. The

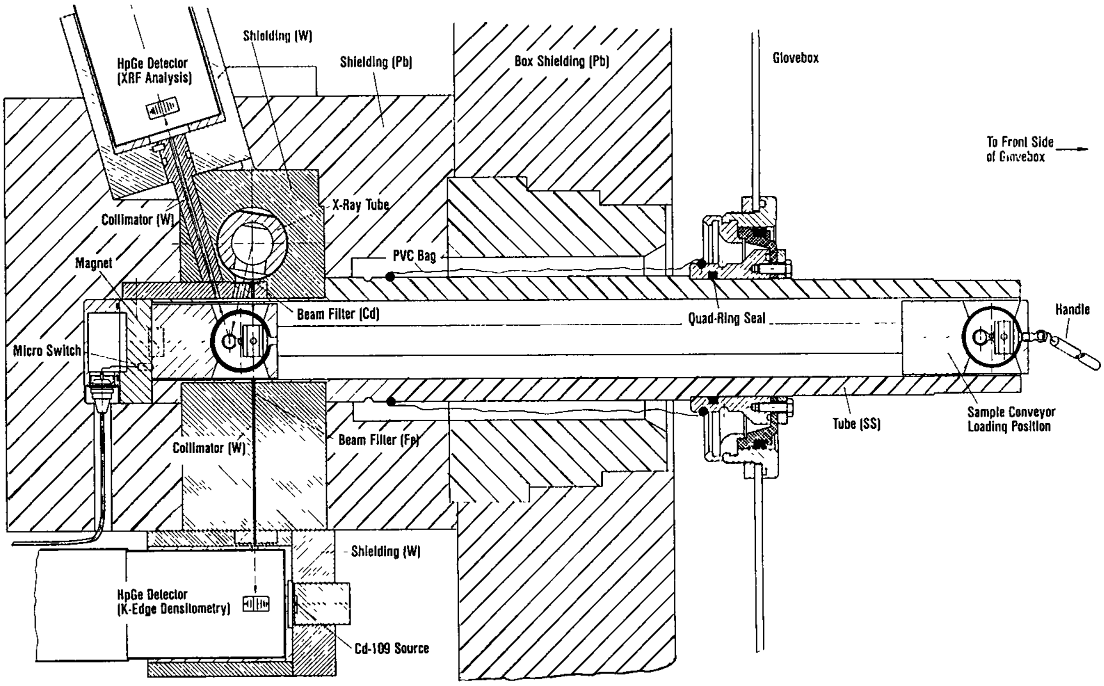

Fig. 3 : Plan of the instrument installation at UP3.

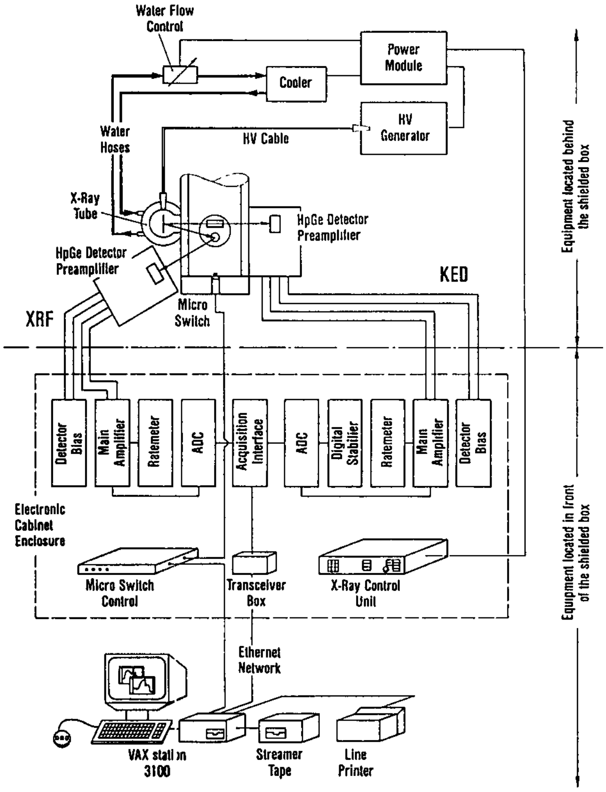

Fig. 4 : Block diagram of the instrument components.

respective modules are identical for both detectors, except that the branch for the KED detector also includes a digital stabilizer set on the reference peaks from a ^{109}Cd source.

An Ethernet-based spectroscopy workstation with associated periphery is used for data acquisition and analysis. It includes an Acquisition Interface Module (AIM) with built-in IEEE 802.3/802.2 conforming Ethernet (LAN) Interface connected to a Digital VAX station 3100. The latter runs under Digital Micro VMS Operating System and includes Micro VMS workstation software providing multi-windowing capabilities. The dedicated software packages for spectrum analysis and data evaluation are written in FORTRAN.

Test Samples for Measurement Control

Two test samples for instrument monitoring were prepared at TUI /3/. The test sample for KED consists of a uranium solution (218 g U/l) sealed in a glass cuvette of the same type as used for the input solutions. The stability of the U/Pu ratio measurement from XRF is controlled from measurements on a (U, Pu) MOX-pellet. The latter sample, welded in stainless steel, contains about 5 % Pu with a ^{239}Pu abundance of 98.3 %. Both test samples are mounted into a frame and located in a stainless steel capsule as shown in Fig. 5. The capsule with the test samples is loaded into the sample conveyor in the same manner as the capsule used for the input solutions.

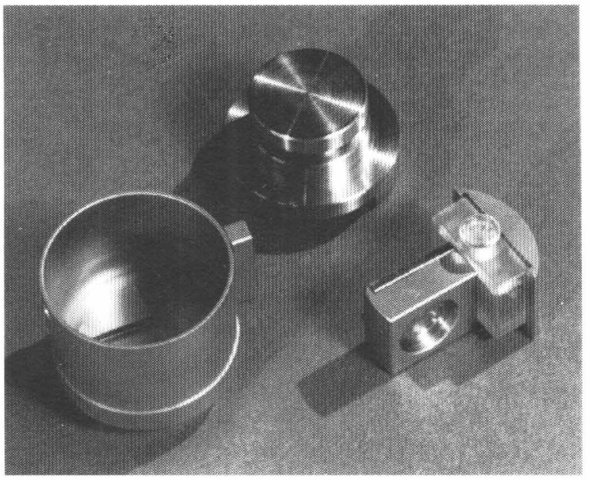

Fig. 5 : Stainless steel capsule with test samples for measurement control.

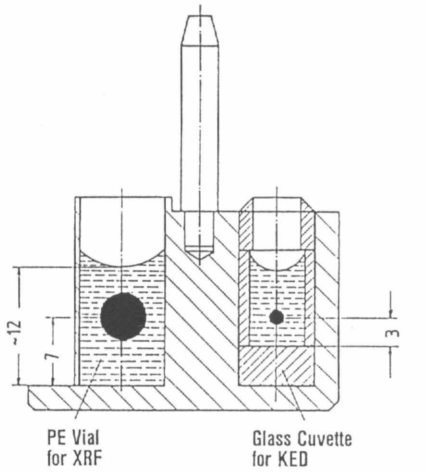

Fig. 6 : Sample vials for input solutions. Black dots indicate position and size of incident X-ray beams.

3. Operating Procedure

Samples of liquor taken from the input accountancy tank arrive directly in the shielded box to which the hybrid K-edge is attached. Their arrival, subsequent transfer to a special storage rack and any other manipulations are observed by a video camera looking down into the shielded box through a leaded window. This camera is connected to a front end video motion detector, and any activity detected in the zones of interest is recorded on a time lapse recorder. The inspector usually chooses 3 sample bottles at random from the 20 or so bottles which arrive in total before each transfer from the accountancy tank into the process.

When the inspector is ready to carry out the verification measurement he first initiates an automated warming-up sequence for the X-ray tube, whose duration depends on the time passed since the last operation of the tube. He then asks the operator's staff to place the control standard, which is normally stored in the sample rack under video surveillance, into the sample conveyor and to transfer it into the measurement position. After a combined measurement of 1000 s each for the K-edge and XRF chain the control measurements are checked and entered into an instrument quality control log.

If the instrument is found to be working properly, the operator's staff then transfer approximately 2 ml of input liquor into the K-edge vial and 1 ml into the XRF vial from the sample bottle chosen by the inspector. The samples vials, shown in Fig. 6, are placed in a special capsule which is closed by a lid to avoid spillage, and the capsule is moved into the measurement position with the conveyor system. When the control light for the micro-switch indicates correct positioning the inspector starts the verification measurement, which normally consists of 3 repeat acquisitions of 1000 s live time each. Because of the containment and surveillance measures applied to the instrument environment the inspector may leave the laboratory during the measurement. The measurement results are entered into a special log.

They are compared to the results of the operator's chemical analysis done by Isotope Dilution Mass Spectrometry (IDMS) as soon as these have been made available to the inspector. If both results agree within the known measurement uncertainties, the sample and the other bottles taken from the same batch are returned to the operator. If the operator-inspector differences in either the uranium or plutonium concentration are larger than certain thresholds, a second sample may be measured.

4. Calibration

Preliminary Calibration

A preliminary instrument calibration was carried out in November 89 at Karlsruhe, prior to the shipment of the instrument to La Hague. A set of synthetic U/Pu-solutions, prepared by the Nuclear Technology Division of the European Institute for Transuranium Elements (TUI), Karlsruhe on a gravimetric basis from U metal standard material NBS 960 and Pu metal standard material NBS 949e, were used for this purpose. The reference materials were mixed to yield samples with a U/Pu ratio of about 100.

The reference solutions served to calibrate the XRF channel of the instrument for a correct U/Pu ratio measurement. A reliable calibration of the KED channel, which is designated for the measurement of absolute uranium concentrations, was not possible at that time. There existed indications that the calibration solutions, while keeping them in the vials for measurement, had slightly increased their concentration due to evaporation. Since time constraints prior to instrument shipment did not allow to characterize new reference solutions, the instrument was initially run with a calibration factor determined for the prototype system at TUI /2/. It was expected that this calibration factor for an instrument of similar design should also apply to the present instrument within an uncertainty of about 0.1 to 0.2%.

Final Calibration

In July 1990, after about 7 months of instrument operation, a fairly extensive final calibration was carried out with a larger set of reference solutions. The synthetic calibration solutions, again prepared from the same NBS reference materials mentioned above, were established and characterized at TUI and then shipped to La Hague. They included mixed U-Pu solutions at different concentration levels and U/Pu ratios to calibrate the instrument for input solutions. Further, there were also separate uranium and plutonium solutions available with concentrations down to 0.1 g/l to calibrate the XRF channel of the instrument for the measurement of other verification samples at lower U and/or Pu concentration.

Fig. 7 shows the results of the calibration runs for KED. The single calibration factor $\Delta\mu$ to be determined from the calibration physically represents the difference of the total photon mass attenuation coefficient for uranium at energies $E_- = 113.25$ keV and $E_+ = 117.26$ keV bracketing the K-absorption edge. The calibration factor was determined as weighted mean value from the calibration points to $\Delta\mu = 3.3374$ with a standard error of 0.04%. This value differs by 0.13% from the preliminary calibration factor $\Delta\mu = 3.3331$ determined for the prototype instrument at TUI /2/.

The fission product elements in a dissolver solution cause a small negative bias to the uranium assay by KED. The magnitude of the bias depends on their concentration, which in turn is proportional to the burnup of the fuel. For burnup values between 30 and 40 GWd/t the necessary bias correction is about 0.15%. This correction is automatically applied whenever the system detects fission products through their gamma radiation. The diagnostics is provided by the XRF detector, which is more sensitive to this radiation because of the larger collimator diameter in the XRF channel.

The XRF calibration factor $R_{U/Pu}$, which relates the measured $UK\alpha_1/PuK\alpha_1$ peak area ratio to the U/Pu element ratio, was determined from the calibration runs with a standard error of 0.09%. A small non-linearity was observed for this conversion factor at high total heavy element concentrations (>250 g/l). This non-linearity was approximated by a second-order polynomial.

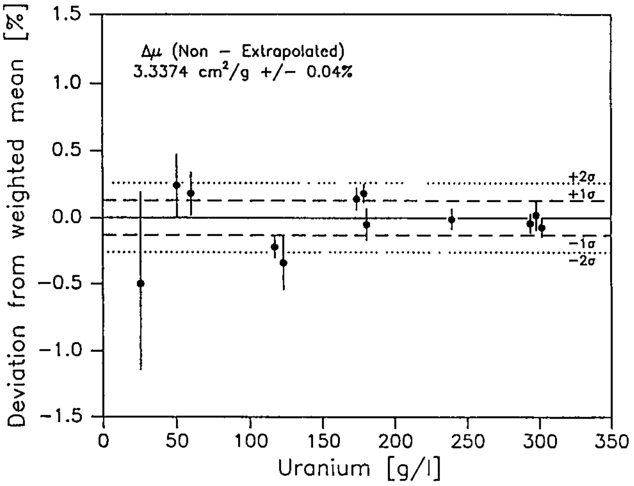

Fig. 7 : Determination of the calibration factor $\Delta\mu$ for KED.

The option to measure absolute U or Pu concentrations at lower levels by XRF alone is not yet fully implemented. The calibration runs taken for this mode of analysis are currently being used to standardize this type of measurement.

5. Performance

Instrument Stability

The short and long-term behaviour of the instrument response in both measurement channels (KED and XRF) was monitored from regular control measurements on the respective test samples. Fig. 8 shows the results of control measurements for KED, which initially were performed on a test sample made from a uranium metal foil. Unexpectedly, large systematic trends of up to 2% were observed from those measurements as shown in the left plot in Fig. 8. The large fluctuations, far beyond the measurement precision, were attributed to a possible local inhomogeneity of the foil thickness. This assumption has later been verified. As a consequence, the metal foil was then replaced in July 90 by the sealed uranium solution. The results of control measurements performed on this sample over a 9-month period (Aug. 90 - April 91) are shown on the right-hand side in Fig. 8. The relative standard deviation of 0.30% for the differences from the mean value is not significantly larger than the precision of a single measurement (0.24%). The moving average ($n = 5$) plotted in the figure indicates small instrument drifts of about ±0.3% at maximum for this period.

Similar control charts as for KED were produced for the XRF channel from measurements on the MOX test sample. Two sets of data, obtained from two MOX pellets of the same type but of slightly different U/Pu ratio, exist for the periods from 11/89 - 07/90 and 08/90 - 04/91 (Fig. 9). The relative standard deviations of the differences

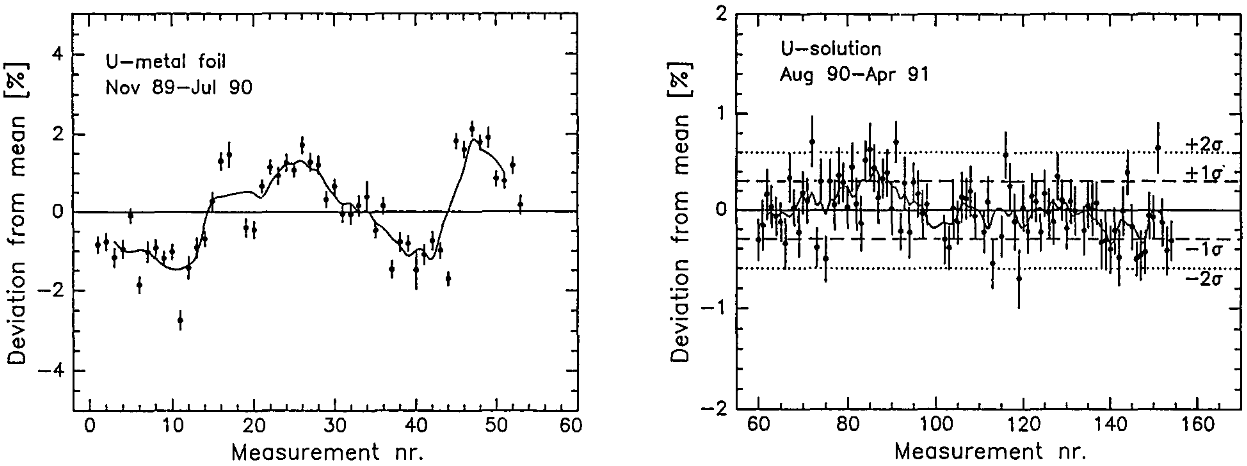

Fig. 8 : Control measurements for KED performed on a uranium metal foil (left), and on a sealed uranium solution (right). Solid line: moving average (n = 5).

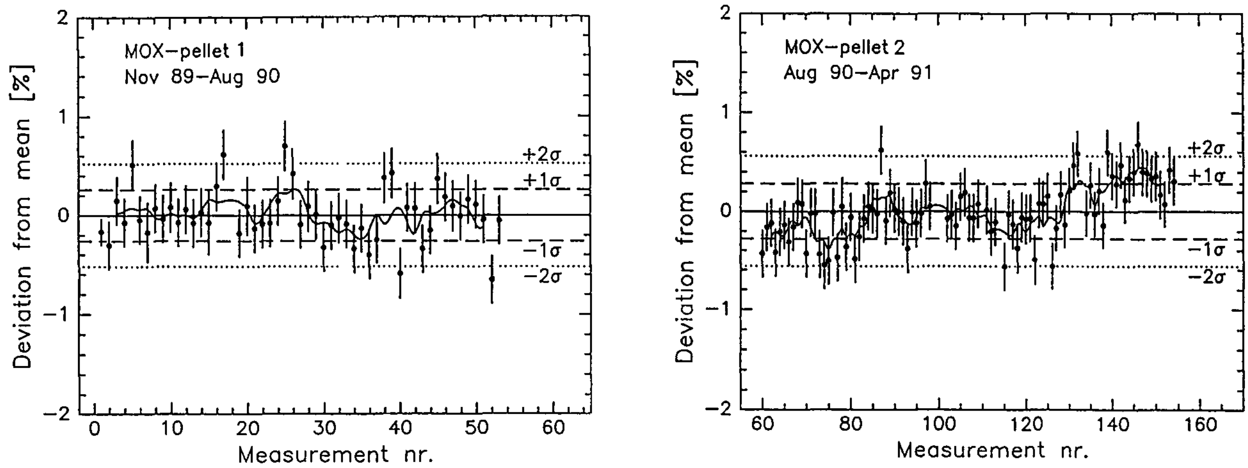

Fig.9 : Control charts for the U/Pu ratio measurement from XRF. Test measurements performed on MOX pellets. Solid line: moving average (n = 5).

from the mean values (0.26% and 0.28%) are slightly larger than the precision of 0.24% for a single measurement. Small drifts of up to 0.3% are observed for both test periods.

Up to now the instrument behaviour for both the KED and XRF measurement is only being monitored. The small systematic trends observed have not yet been used for a readjustment of calibration. This may be done in future.

Results for Input Liquors

During the first 12 months of operation two different types of samples were measured with the hybrid K-edge instrument. A first group of samples was taken from input liquor after the separation of fission products, the second group consisted of normal input samples containing fission products. The relative inspector-operator differences for the U and Pu concentration, and for the U/Pu ratio are shown in Figs. 10 and 11 for the two campaigns. Wash batches with very low U and/or Pu concentrations are not shown and have not been included in the evaluation. The average relative differences and the standard deviations of the relative differences are shown as dashed and dotted lines in the figures. Only very few results do fall outside the 2σ-range (6 out of a total of 195 data points). Relative differences deviating by more than 3σ from the average are not observed.

Table 1 summarizes the results for the two measurement campaigns on input liquors without and with fission products. Mean values and standard deviations of relative inspector-operator differences were calculated for each campaign once including all data, and once excluding differences

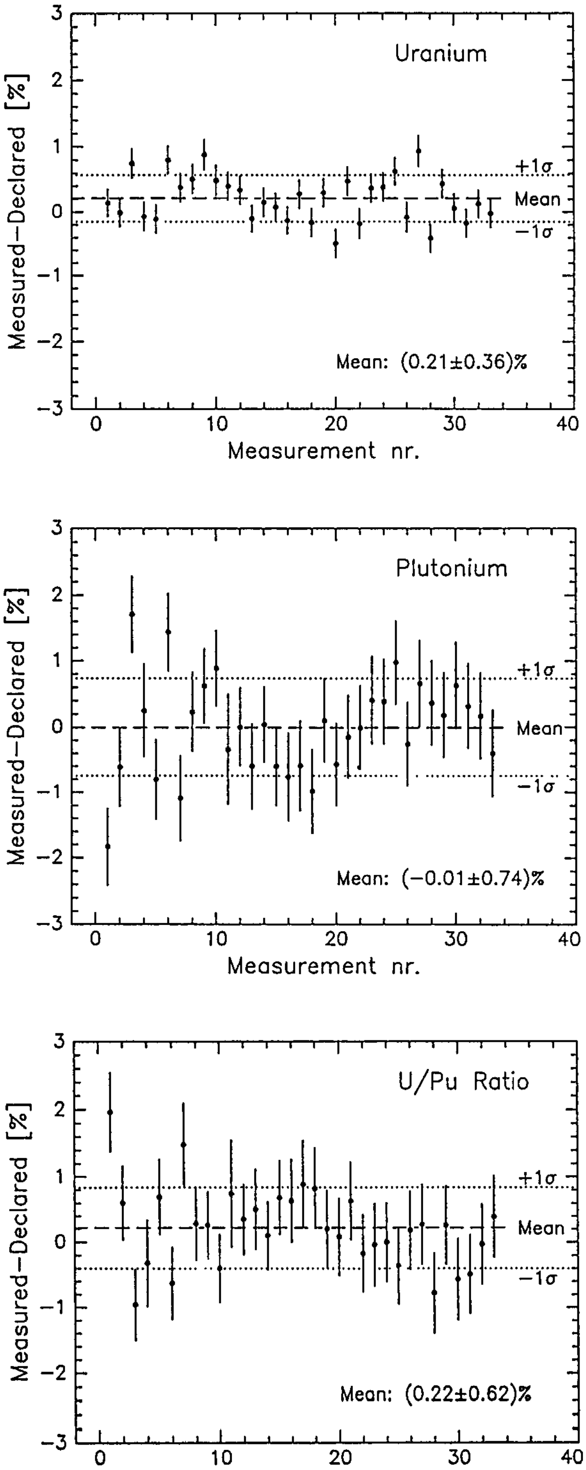

Fig.10 : Results from 1st measurement campaign on input solutions after separation of fission products.

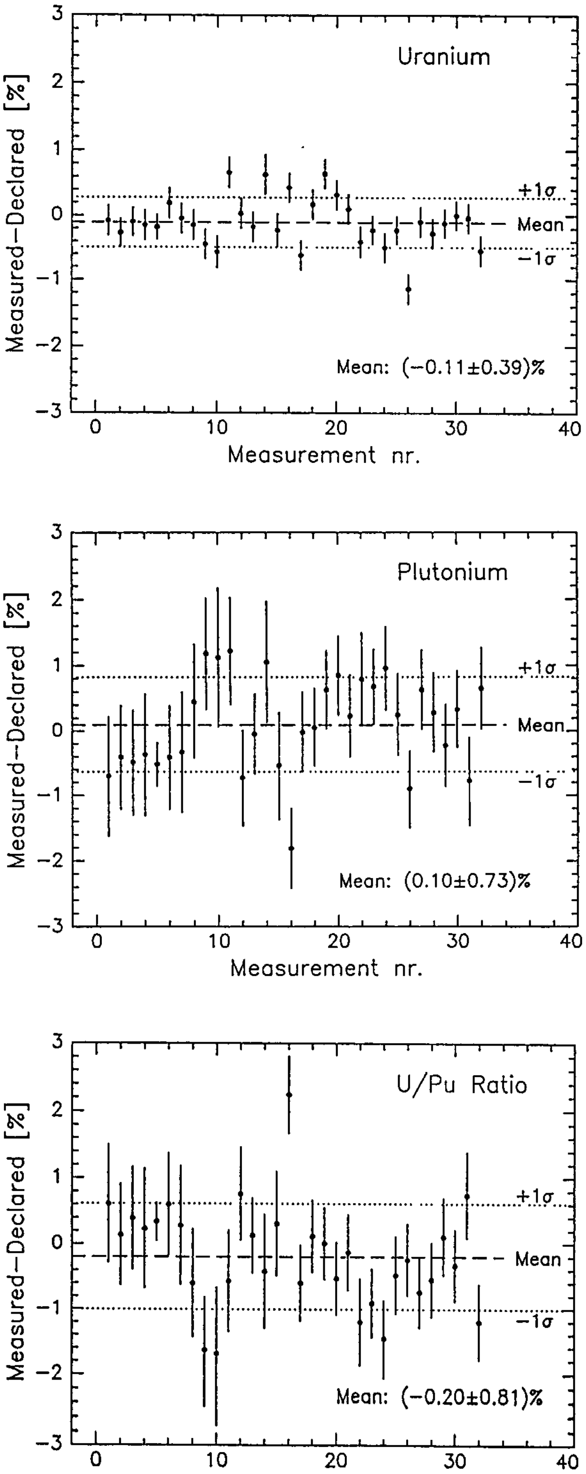

Fig.11 : Results from 2nd measurement campaign on original input solutions with fission products.

Table 1 : Mean Values and Std. Deviations for Relative Inspector-Operator Differences.

Data Base	Mean Rel. Diff. ± Std. Dev. (%) U-Conc.	 Pu-Conc.	 U/Pu-Ratio
		Input Liquors without FP	
All (n = 33)	0.21 ± 0.36	-0.01 ± 0.74	0.22 ± 0.62
Diff. > 2σ Excluded	0.21 ± 0.36 (none)*	-0.00 ± 0.61 (2)	0.16 ± 0.55 (1)
		Input Liquors with FP	
All (n = 32)	-0.11 ± 0.39	0.10 ± 0.73	-0.20 ± 0.81
Diff. > 2σ Excluded	-0.07 ± 0.35 (1)	0.18 ± 0.65 (1)	-0.31 ± 0.68 (1)

( )* Number of Excluded Results.

which deviate from the average by more than 2 units of the standard deviation calculated for the full set. The number of excluded data, ranging between zero and 2, are given in brackets.

The results confirm the error estimates for the operator's chemical analysis as well as the random and systematic error variances calculated and estimated for the K-edge measurements. The presence of fission products does not lead to a deterioration of instrument performance. The standard deviation and the average of the relative differences in U concentration are smaller than those observed for analysis by IDMS. The same is true for the average values of the relative differences in Pu concentration and in U/Pu ratio. The standard deviations of the relative differences in Pu concentration and in U/Pu ratio are better than the average performance values for paired comparison of inspector and operator analyses done by IDMS.

6. Conclusions

A hybrid K-edge instrument was installed at a shielded cell of the Cogema laboratories at La Hague as the result of a very successful cooperation between Cogema, the French government, Euratom Safeguards Directorate, KfK and TUI Karlsruhe. This instrument has been successfully employed for routine safeguards verification analysis of samples of input liquor taken from the accountancy tank of the large new reprocessing plant, UP3, for more than one year. For the first time in the history of Safeguards, inspectors can independently verify nuclear material concentrations in input accountancy tanks of reprocessing plants as early as two hours after sampling with an accuracy of 0.2-0.3 % for uranium and 0.6-0.7 % for plutonium. Instrument performance has been consistently better than expected from ESARDA target values. Considering the costs for transport and chemical analysis of samples of this kind the instrument has already paid for itself. In future the accuracy of measurements may be further improved by increasing measurement time and/or counting rates (fast pulse processors) and by using the smoothed results of the control measurements for bias correction. Process samples with lower nuclear material concentration will be measured with this instrument using the X-ray fluorescence chain on its own. Cogema have also purchased and installed a hybrid K-edge for their own analytical requirements at La Hague.

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USE OF CDTE DETECTORS FOR THE VERIFICATION OF NUCLEAR MATERIAL

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Introduction

High Purity Germanium Detectors (HPGe) and NaI(Tl) detectors are the workhorses used for the verification of nuclear material using gamma spectrometry. For some applications, however, these detectors are not optimal. HpGe detectors need to be cooled by liquid nitrogen. NaI detectors have only a moderate resolution.

CdTe detectors have properties which place them in the middle between above detector options. They exhibit a better resolution as NaI detectors but do not require liquid nitrogen for cooling. They have a higher intrinsic energy efficiency as above detectors. These properties allow the design of miniature detection systems which are portable and can e.g. be used to access nuclear fuel items which would otherwise have to be moved (isolated) for the verification. These features make the CdTe detectors attractive for different verification tasks in International Safeguards.

The IAEA has for many years been paying attention to the development of CdTe detectors. Considerable progress was made within Technical Support Programme Tasks of several Member States - France. Germany and the Soviet Union - to develop customized detection systems and electronics for use of CdTe for specific measurement tasks where other gamma detectors are difficult to use.

An overview is given below on the properties and the usage of CdTe detectors.

Properties of CdTe detectors Developed for the IAEA

Two different types of CdTe detectors are available - hemispherical and planar detectors. Hemispherical detectors do not require pulse shape discrimination (PSD) and can be used at very high countrates - up to several 100000 cps. Planar detectors can only be used in conjunction with a PSD module which corrects for the different rise times of the pulses at the output of the preamplifier.

Under the French Support Programme large area planar detectors and highly sensitive hemispherical detectors have been developed. Under a joint French - German Support Programme Task the PSD electronics has been developed.

Under a Soviet Support Programme Task and a Research Contract high countrate hemispherical detectors and other specialized detectors have been developed.

An overview on the properties of the detection systems is given in the tables 1 and 2.

Typical gamma spectra for different detectors and sources are shown on the Figures.

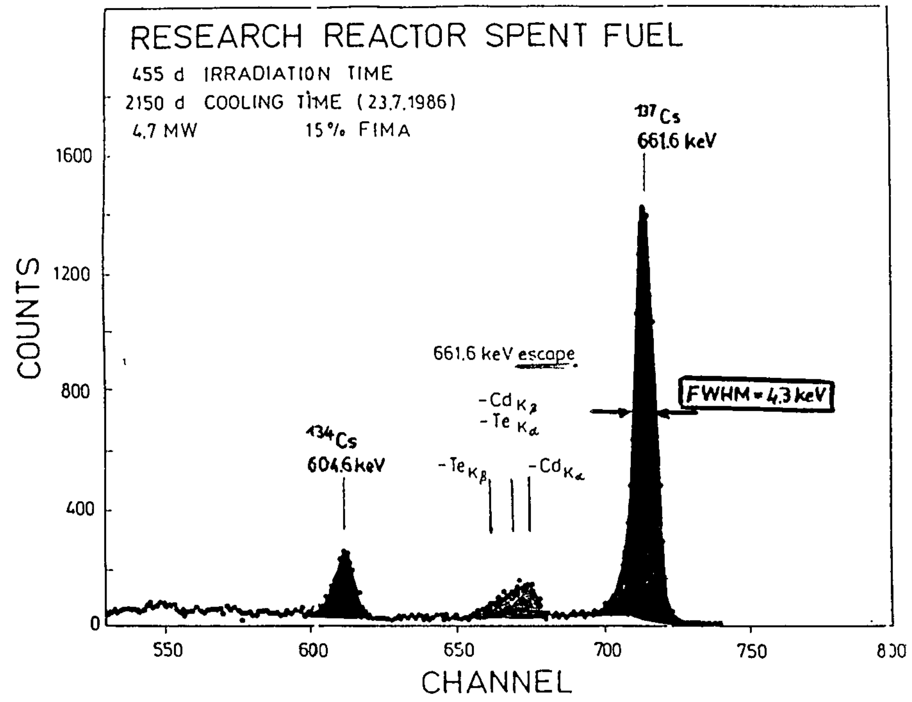
Table 1 Planar CdTe detectors

Type of planar detector	Features	Possible use
Structure metal - semiconductor	Typical volume of single detector 5x5x1-2mm, mosaic structures possible; excellent resolution can be obtained using pulse shape discrimination (PSD)	Verification of fresh LEU and MOX replacing NaI detectors in some application cases; Attribute testing of spent fuel with short cooling time
Stucture p-i-n	Size of single detector up to 1 sqcm possible, mosaic structures possible, effective thickness 0.2-0.5 mm; good resolution can be obtained using a mini Peltier cooling device	As above

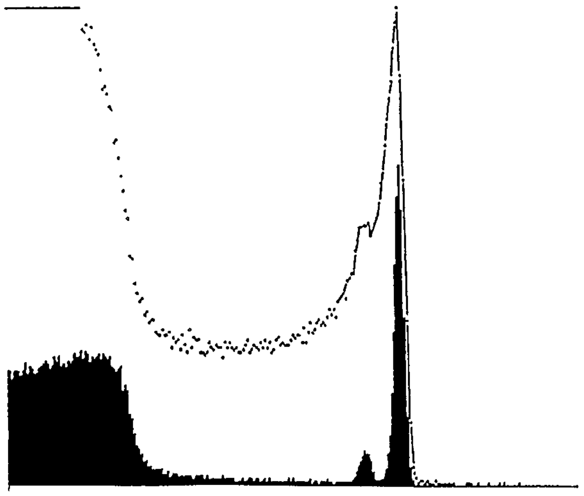
Table 2 Hemi-spherical detectors

Type of detector	Features	Applications
Dual hemispherical detection system	High sensitivity for in situ verification of the initial enrichments of fresh LWR assemblies	In situ verification of fresh fuel assemblies
Large volume hemispherical detectors	Highly sensitive detectors with a volume of up to 1 ccm	Applications which require a high sensitivity of the detector
Hemisherical detectors with integrated preamplifier coupled to mini peltier cooling device	Allows measurements with good resolution up to 60 degrees C; integral package - detection probe can be well shielded	Measurements in a high temperature environment, e.g. in a spent fuel pond; partial defect test of spent fuel if combined with GRAND-I/Fork
High countrate system with seperate second stage of preamplifier	Allows measurement of peak area of Cs-137 radiation of spent fuel up to a countrate of several 100 kcps	CANDU bundle verification; verification of difficult to access spent fuel items
High countrate system with integrated preamplifier (under evluation)	Same properties as above but integral preamplifier-detector allowing a long distance between detector and MCA	Verification of difficult to access nuclear material

GAMMA SPECTRUM OF SPENT FUEL TAKEN WITH A PLANAR CDTE DETECTOR AND A PULSE SHAPE DISCRIMINATION SCHEME

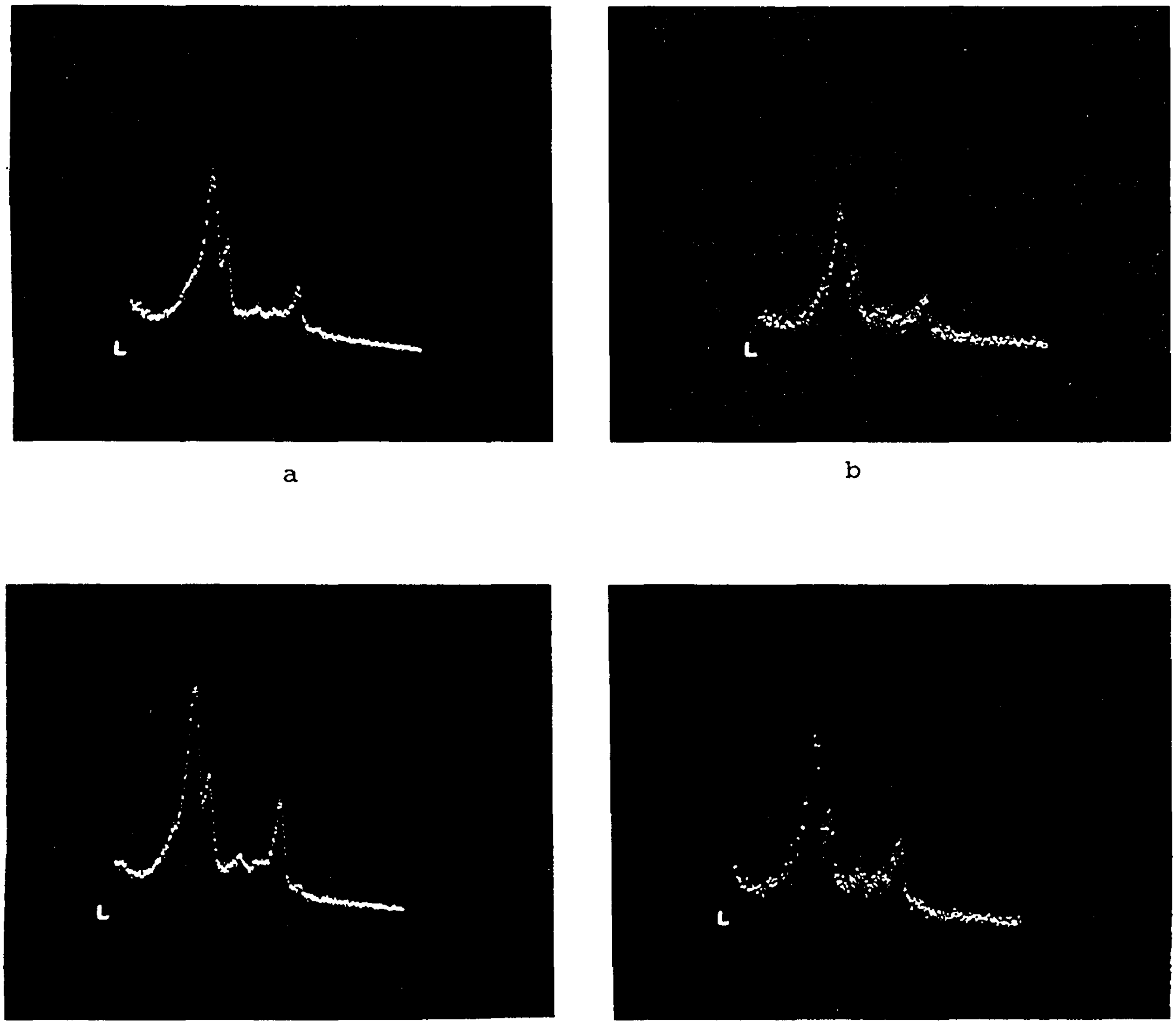


TIME WINDOW 6 NANOSEC, 80 % COUNTING LOSSES IN THE PHOTO-PEAK DUE TO THE PSD



IMPROVEMENT OF THE PERFORMANCE OF A PLANAR CDTE DETECTOR USING PULSE SHAPE DISCRIMINATION (PSD); CS-137 SOURCE, GAMMA ENERGY 662 KEV

Gamma Spectra of WWER-440 Fresh Fuel Assemblies Taken by a CdTe Detector Inserted into the Central Instrumentation Hole



a b

c d

Legend

a: Enrichment 1.6%, 1000 seconds livetime
b: Enrichment 1.6%, 100 seconds livetime
c: Enrichment 3.6%, 1000 seconds livetime
d: Enrichment 3.6%, 100 seconds livetime

Description of Some Application Cases of CdTe Detectors

Verification by Spent Fuel Specific Attributes of CANDU Bundles Stored in Stacks of Trays (access from the side with at least 100 mm gap between stacks)

Short description of the verification task:
Verify several thousand CANDU bundles stored in stacks of trays with a random low sample size.

Solution:
Use of a well shielded and collimated miniature CdTe detection probe moved along the side of the stacks. The bundles are verified by a bundle-gap modulation of the magnitude of the Cs-137 gamma peak area.

Use of other detector options:
If HPGe or NaI were used, the spent fuel inventory would have to be moved - a technically extremely difficult, expensive, and time consuming procedure.

Verification of CANDU Bundles Stored in Baskets

Short description of the verification task:
Verify CANDU bundles stored in baskets after interbay transfer.

Solution:
Same solution as described above.

Use of other detector options:
Use of HPGe was considered but would have led to about 5 times more costly solution with a higher level of intrusion (partial isolation of the bundles required). Gross gamma silicone detectors were used before, but the effectiveness of the method was lower.

Verification of CANDU Bundles in Dry Storage Canister

Short description of the verification task:
Verify inaccessible CANDU spent fuel bundles stored in a dry storage canister; a monitoring channel with a diameter of 2.5 cm is available.

Solution:
A unique pattern of the Cs-137 gamma radiation along the axis of the canister is obtained by using a miniature CdTe detection system.

Use of other detector options:
HPGe or NaI detectors cannot be used because they are not small enough.

Verification of Spent Fuel Assemblies Stored in Wells

Short description of the verification task:
Verify by attributes spent fuel assemblies stored in dry channels in concrete accessible only from above through a narrow channel

Solution:
A miniature CdTe detection system is inserted from above into the well. A Cs-137 spent fuel signature is obtained.

Use of other detector options:
HPGe or NaI detectors are too large to be sufficiently shielded and inserted into the well. The assemblies would have to be removed for the verification.

Verification of On-Load Reactor Spent Fuel Stored in a Double Layered Vertical Storage Geometry

Short description of the verification task:
Verify spent fuel bundles of an on-load reactor hanging vertically above each other in two layers in a storage rack allowing access from above through a 100 mm gap only

Solution:
A shielded and collimated CdTe detection system is inserted into the gap and rotated. The bundles can be verified by a modulation of the Cs-137 gamma signal as a function of the rotation angle.

Use of other detector options:
NaI or HPGe detectors can not be used because of their high sensitivity and the limited space for shielding

Verification of Fresh LWR Fuel Assemblies

Short description of the verification task:
In situ verification of the initial enrichment of LWR fresh fuel assemblies.

Solution:
A CdTe detection probe with a diameter of 8.2 mm is inserted into an instrumentation channel of the fuel assembly. It is not needed to lift or remove the assembly from its storage location.

Use of other detector options:
NaI detectors can only be used if the assemblies are accessible from the side. This is not always the case.

In Situ Verification of UO-2 Powder in Drums

Short description of verification task:
UO-2 power stored in drums needs to be verified. The number of DA samples taken can be reduced if the homogeneity of the powder in the drum can be checked by NDA.

Solution:
A stainless steel pipe with a diameter of 9 mm can be inserted into the powder at any location. A CdTe detection probe is inserted into this pipe. In this way the enrichment can be checked at any location inside the drum.

Use of other detector options:
NaI detectors are too big, Small diameter BGO detectors are an alternative but have a much lower resolution.

Other application cases which are still in early stage of the development:

- development and use of large area high sensitive CdTe detectors for attribute testing of fresh MOX and LEU assemblies
- development of a detection system to be used for the unattended monitoring of spent fuel transfers using spent fuel specific signatures (fission products);
- development and investigation of a high performance X-ray CdTe detector to be used for CANDU bundle verification with severe space limitations
- combination of CdTe detector with the GRAND-I/Fork to achieve a partial defect test capability for spent fuel measurement on LWR spent fuel.

A GAMMA-SPECTROSCOPY SYSTEM FOR BURNUP MEASUREMENTS OF NUCLEAR FUEL

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Abstract

A high-speed gamma-ray spectroscopy system for verification of nuclear fuel of the BWR type has been constructed. It uses a gamma-ray collimator, commercial high-speed NIM electronics, a fast PC and especially designed software and PC interface. It allows accurate determination of e. g. ^{154}Eu in presence of strong activities of ^{134}Cs and ^{137}Cs. With this technique isotopic correlations may be used for independent determination of fuel parameters.

1. Introduction

Gamma-ray spectroscopy is a well-known method for non-destructive verification of the burnup of nuclear fuel, see e. g. /1, 2/. Usually the ^{137}Cs activity is measured and the average intensity of this line (662 keV) is assumed to be proportional to the average burnup of the assembly. Also gamma radiation from ^{154}Eu is of interest for several reasons: It is sufficiently long-lived to be essientially independent of the power history of the reactor. Since its intensity is approximately proportional to the square of the burnup, the burnup of an assembly can be obtained in an alternative way to direct measurements of the cesium radiation, by forming the intensity ratio to the cesium intensity. Further, the energy of the most prominent peak (1275 keV) has a higher penetrability than the cesium radiation, which implies a larger potential for examination of the interior of the fuel assembly for missing fuel rods. On the other hand, in normal BWR fuel the europium activity is one to two orders of magnitudes weaker than the cesium activity, which requires that accurate measurements have to be performed at high counting rates.

We here present a spectroscopy system that allows gamma-ray scanning of spent nuclear fuel at counting rates exceeding 80 kcps into the computer memory.

2. Equipment

2.1. Detector arrangement

At all BWR power plants and interim storage facilities in Sweden and Finland, an arrangement consisting of a horizontal collimator and a specially designed elevator for scanning the fuel elements are installed /2/. An overview of the equipment is shown in fig. 1.

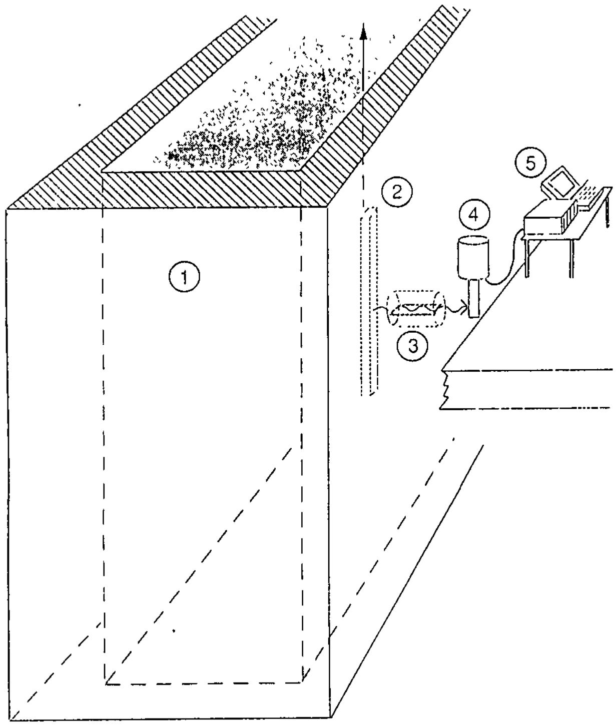

Fig. 1 - Schematic view of the equipment of the interim storage CLAB in Sweden. 1). Fuel handling pool. Depth; 13 m, thickness of walls; 1 m concrete. 2). Fuel assembly in measuring position. During measurements, the assembly is moved in the vertical direction. 3). Horizontal collimator, approximately 1 m of iron. 4). High-purity germanium detector. 5). Electronics and computer for data acquisition.

The collimator is shaped as a horizontal slit with a height that can be varied between 1 and 5 mm. The opening of the collimator is such that the detector views the full width of the measured assembly. In order to enhance the radiation in the 1 MeV region relative to the large background of scattered radiation, a 4 mm lead filter is normally used.

The detector used is a 40% HPGe n-type detector. In order to optimize the line shape, the detector is irradiated radially /3/.

2.2. Electronics

Fig. 2 shows a block scheme of the spectrometer system. The detector is equipped with a transistor reset preamplifier which, together with an Ortec 973U gated integrator amplifier, allows a throughput of over 100 kcps with an energy resolution of about 3 keV FWHM. The ADC is a ND 582 with a fixed conversion time of 1.5 μs and a total busy time of 2.4 μs.

The data are stored and analyzed in a PC via a specially designed fast interface card /4/. The essential part of the card is a FIFO circuit which de-randomizes the data transfer to the computer. With the ND 582 the transfer time to the FIFO is 0.6 μs. Normally a fast PC, like the Toshiba 5200 laptop which has a 386 processor with a 20 MHz clock frequency, is used for data collection and analysis. With a buffer size of ten events in the FIFO, data can be transferred into the computer memory at a rate exceeding 115 k events per second with essentially no deadtime loss before saturation occurs.

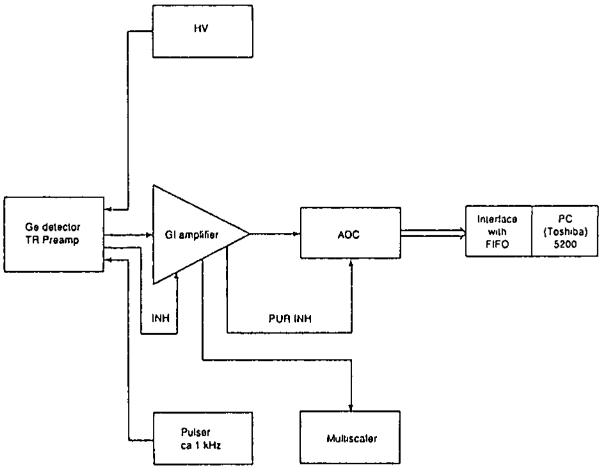

Fig. 2 - Block scheme of the spectrometer system.

In high count rate systems, the deadtime may be considerable and it is essential that it is adequately corrected for. In the present system this is done with the pulser method (see fig. 2). A pulser peak is placed at the high-energy end of the spectrum, where the background is negligible. The data taking code automatically evaluates the peak area and corrects the spectrum accordingly. For a spectrum recorded during 3 seconds at 50% deadtime and a pulser rate of 2 kHz, the correction factor will be accurate to better then 2%.

2.3. Software

The gamma-ray scanning of a fuel assembly is performed by sequential measurement of a number of spectra while the assembly is moved vertically at constant speed in front of the collimator. A special code, "SKIDAS", was written in C for sequential storing of spectra. A gross lay-out of the code is given in fig. 3.

Before starting the data taking, the number of spectra N and the recording time per spectrum T are given together with data on the gamma-ray peaks of interest in the measurement and the pulser frequency used. The code then sequentially record N spectra and store them on disc. Three spectrum areas are used for this and the switching between the areas is performed with no loss of counts. For controling the procedure, two other spectrum areas are used to display the sum of all spectra and an optional number of histograms

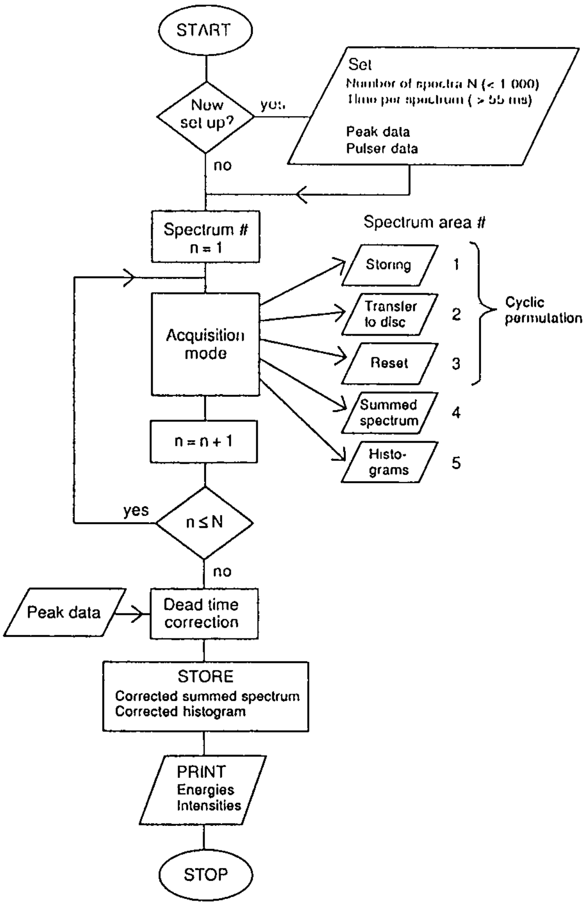

Fig. 3 - Flow diagram of the soft ware used.

showing the intensity of selected gamma-ray peaks as a function of the spectrum number. Fig. 4 displays three such histograms showing the intensities of the ^{137}Cs peak, the ^{154}Eu peak and the ^{60}Co peak as the complete length of a fuel assembly was scanned with N=70 and T=3 seconds. The sharp peaks in the ^{60}Co histogram, originate from the rod holders.

After completion of a scan, all spectra are automatically corrected for deadtime. A corrected summed spectrum is created and stored together with histograms corrected for dead time. Finally the energies and intensities of the peaks of interest are evaluated from the corrected summed spectrum and printed.

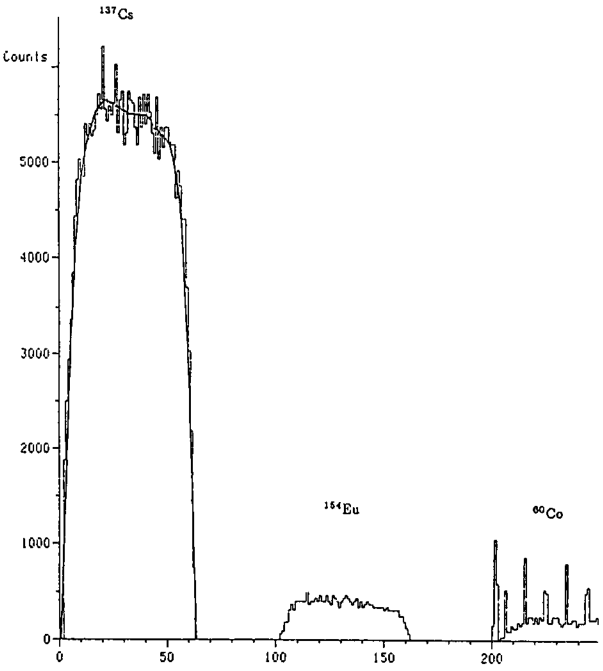

Fig. 4 - Intensity distributions along an assembly with BU=33 GWd/tU and cooling time=39 months.

2.4. Results

The high-rate capacity of this system is such that input rates exceeding 200 kcps can be handled. However, during typical measuring conditions, including pile-up rejection, an input rate at 70 kcps gives a throughput into the computer of about 40 kcps with an energy resolution of 2.6 keV. When a resolution of over 3 keV can be tolerated, the throughput into the computer can be doubled with a dead time of about 50%.

Fig. 5 is an example of a dead time corrected spectrum obtained with the present detector system. The total input rate was for this case 70 kcps and the burnup and cooling time were 33 GWd/tU and 39 months, respectively. With a measuring time of 200 seconds per corner for this assembly, a statistical error of the ^{137}Cs peak of 0.2% is achieved and the corresponding error for the ^{154}Eu peak is 0.8%.

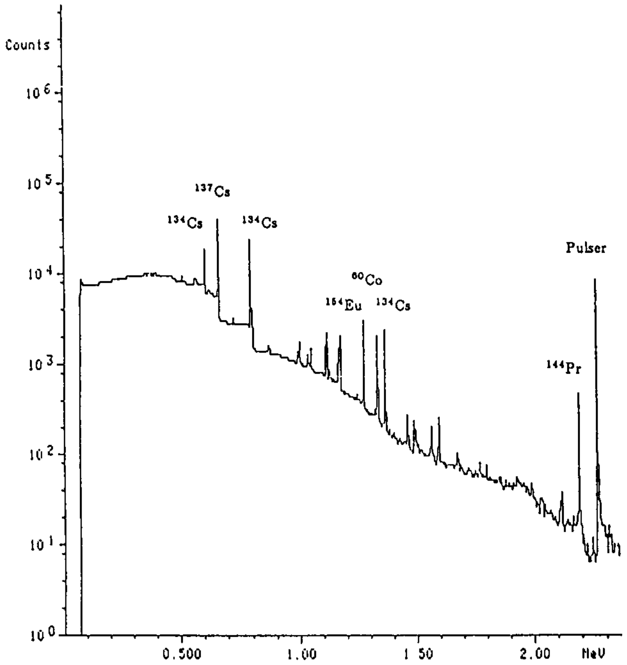

Fig. 5 - Energy spectrum for the same assembly as in fig. 4.

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A NEW SPENT FUEL ATTRIBUTE TESTER FOR IN SITU VERIFICATION OF LIGHT WATER REACTOR SPENT FUEL

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Abstract

A new spent fuel attribute test method has been developed. It allows in-situ verification of Light Water Reactor spent fuel assemblies in wet storage ponds. The method is essential for the verification of spent fuel assemblies with long cooling time when Cerenkov Viewing Devices cannot be used any more. The method uses in the most cases spent fuel specific gamma ray signatures allowing the establishment of the presence of irradiated nuclear material.

1. Introduction

The number of spent light water reactor fuel assemblies is rising from year to year. Only a small percentage of the assemblies undergoes reprocessing. The larger part is stored in recently constructed facilities - long term or away from the reactor storages. The spent fuel assemblies can stay for several decades in a wet type long term storage facility. When the age of the fuel assemblies approaches 10 years it becomes more and more difficult to verify them using Cerenkov Viewing Devices. Other available spent fuel verification methods, however, require at least partial raising of the spent fuel assembly from its storage location in the rack. Fuel movements are, however, increasingly undesirable when the spent fuel ages. This circumstance has prompted the development and implementation of a new spent fuel attribute test method. This method and the device are named Spent Fuel Attribute Tester (SFAT). Since 1991 is authorized for inspection use at the IAEA.

2. Basic Principle of the Method

The basic principle of the method (see Fig. 1) is not new. An airfilled pipe is positioned vertically above the spent fuel assembly to verify. The gamma radiation, emerging from the irradiated pellets of the fuel zone and penetrating the top structure of the assembly, is channeled through an air filled pipe to a suitable gamma detector. If the system is properly designed and only unscattered, full energy gamma rays are recorded, the influence of the other fuel assemblies surrounding the item to be verified, can be made neglectable (near neighbor effect). A similar method has been used to verify spent fuel assemblies stored in multi-element bottles (MEB). In that case, however, only the Co-60 radiation can be utilized since the 662 keV gamma radiation of Cs-137 does not sufficiently penetrate the lid of the MEB.

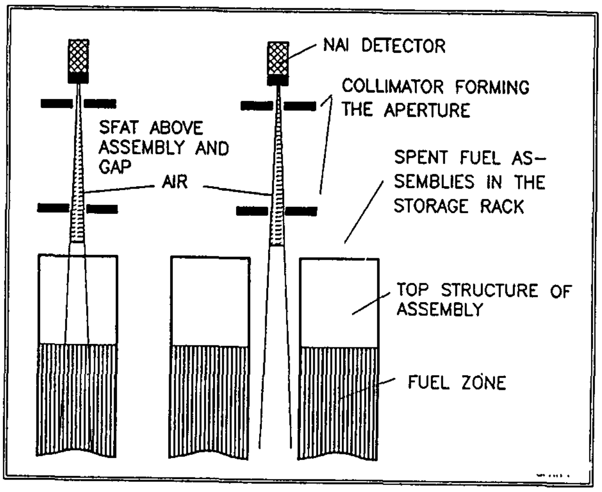

Figure 1 - PRINCIPLE OF THE METHOD

In an early study /1/, a submergible pipe with a gamma detector attached was investigated. The results, however, were not convincing since a gross gamma signal was used. In that case, the near neighbor effect could not be excluded to the extent necessary to establish a reliable verification method.

The device described in this paper uses a NaI(Tl) detector to observe the gamma rays. The aperture of the collimator is designed so that only gamma rays directly from the item to be verified plus scattered gamma rays from its surrounding can reach the NaI detector. The scattered gamma rays, however, are excluded by evaluating only events which are contained in the full energy peak of the Cs-137 gamma spectrum. The performance of this method can be checked by positioning the device above the gap between two adjacent fuel assemblies - as shown on Fig.1. The Cs-137 gamma peak will disappear and only the background of scattered gamma rays remains. The minimization of this background of scattered gamma rays is one of the main development targets since the its presence reduces the sensitivity of the methods and leads to a longer measurement time.

3. Technical Description

The Spent Fuel Attribute Tester (SFAT) consists of the following components (see Figs. 2 and 3):

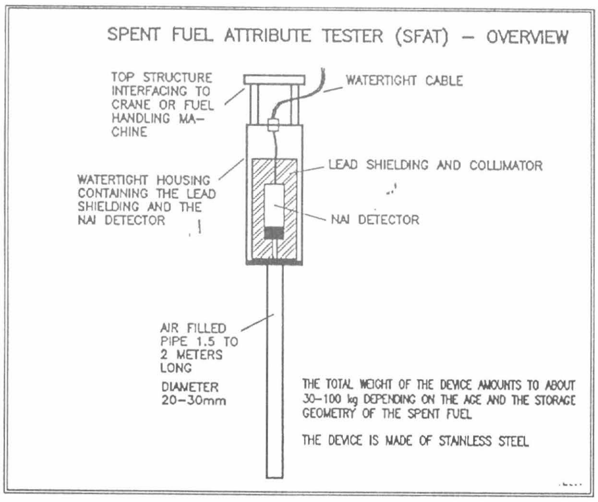

Figure 2 - COMPONENTS OF SFAT

3.1 Top structure

The top structure interfaces with the fuel grapple in case the device is handled by the operator's fuel handling tool. In this case an actual upper tie plate and fuel assembly handle is used (see Fig. 4). Otherwise a clasp and eye arrangement is used (see Fig. 3).

3.2 Detector Housing and Shielding (see Fig. 5)

The detector is lead and copper shielded and encased in a watertight, stainless steel housing. This lead shielding has the following functions: 1) To reduce the background countrate caused by contaminants in the water and by the "cloud" of scattered low energetic gamma rays reaching several meters above the spent fuel assemblies; and 2) to form, by the hole in its lower end, together with the air filled pipe a collimator which determines the aperture of the SFAT.

The size and weight of the lead shielding depends on the application case. The lead shielding can be made smaller and lighter (e.g. 10-20 kg) if long cooled spent fuel has to be verified, but must be made larger and heavier (e.g. 50-80 kg) if old fuel is stored next to spent fuel with short cooling time and/or the water is strongly contaminated.

Figure 3 - OVERALL VIEW OF A SPENT FUEL ATTRIBUTE TESTER FOR FUEL WITH LONG COOLING TIME

3.3 Air-filled Pipe

At the lower end of the detector housing an air-filled pipe is attached. The length and diameter of this pipe depends on the age and the design of the spent fuel and of the storage geometry. Pipes with lengths of 1-2 meters and an inner diameter of 20 - 30 mm are used. The tip of this pipe is covered by a polyethylene end piece which can be removed in case it becomes contaminated.

3.4 System for Handling the SFAT

Two different strategies have been demonstrated and tested: 1) using the operator's fuel handling tools; and 2) using a so called "sliding mounting bracket". A sliding mounting bracket is equipped with rollers and can be moved along the bridge rail (see Fig. 6). On its top there is a manual or motor driven winch which to raise and lower the SFAT.

3.5 Gamma Spectrometer

A NaI detector based gamma spectrometer is used. The crystal size is either 2" X 2" or 1" X 1". A Portable Multi Channel Analyzer (PMCA) with an upgraded amplifier /2/ is used. The upgrade is needed to assure a high

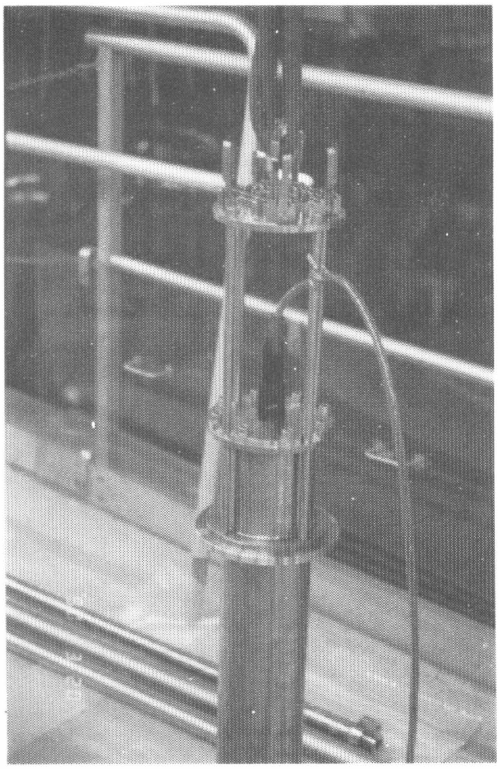

Figure 4 - TOP STRUCTURE OF AN SFAT FOR BWR TYPE ASSEMBLIES

throughput, good resolution and a low pileup rate at very high countrates. Since the gamma peaks have a low intensity and reside on a high background of scattered gamma rays, a good high countrate performance of the gamma spectrometer is essential to reduce the measurement time. A watertight underwater TV cable is used to supply the voltages to preamplifier and photomultiplier and to transmit the signal to the PMCA.

4. Handling and Verification Procedure

Handling and positioning of an SFAT can vary widely between facilities. It can be handled from the bridge with a crane or hoist operated by the facility operator. It can be handled from a steel band and hook attached to a "sliding mounting bracket" fitted over the bridge railing. It can be handled using the facility's fuel handling machine. Facility-specific handling procedures are being worked out in cooperation with operators. Examples of the handling and verification procedures in different facilities were recorded for demonstration on video tape.

The setup and verification procedure /3/ includes the following steps:

- test of the device outside the spent fuel pond using background radiation or a test source;
- transfer of the SFAT into the spent fuel pond (done by the operator);

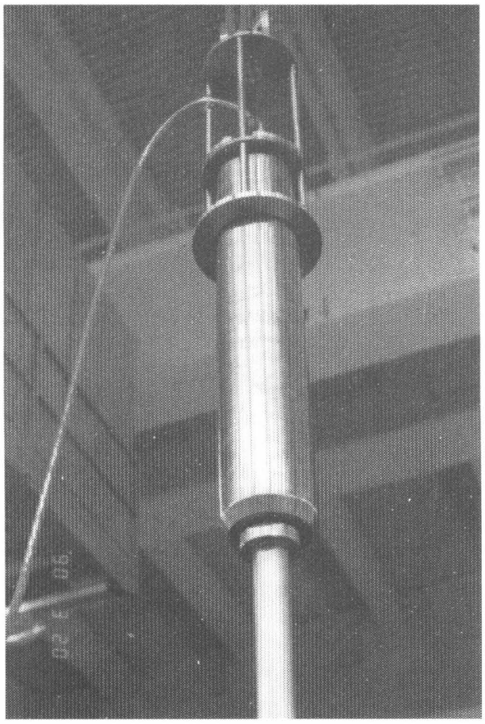

Figure 5 - DETECTOR HOUSING

- measurement of the background gamma spectrum caused by contamination of the water;
- positioning the SFAT above the fuel assembly to be verified (done by the operator);
- measurement of the gamma spectrum; the measurement time amounts from tenths of seconds in favorable cases to several minutes in unfavorable cases; and
- optional measurement of the gamma spectrum above the gap between the fuel assembly to be verified and the adjacent fuel assembly.

The data evaluation and conclusion drawing is done on the spot. If a Cs-137 peak can be recognized visually on the display of the PMCA, the assembly is considered verified. In marginal cases, the Cs-137 peak area must be measured above the assembly and above the gap. The determination of the Cs-137 peak area and its error gives in these cases a quantitative acceptance criterion.

The verification procedure and a typical measurement result for WWER-440 type spent fuel with short cooling time is illustrated on Fig. 7.

5. Results

The basic performance of the method has been studied for various spent fuel and storage geometries and fuel assemblies with different values of the cooling time

Figure 6 - SLIDING MOUNTING BRACKET AND WINCH

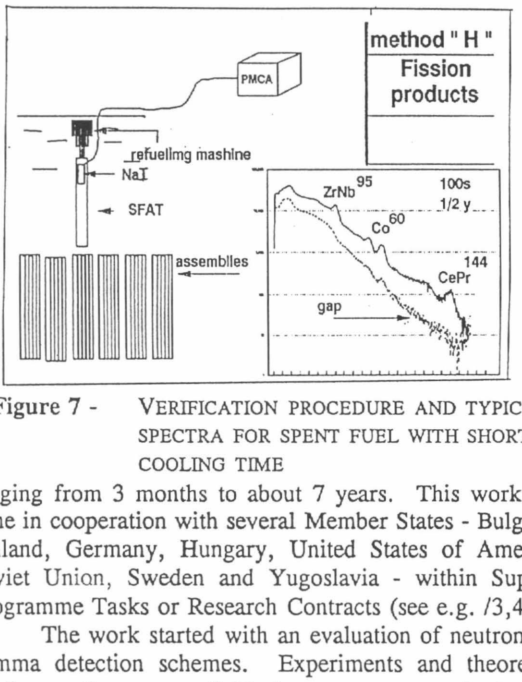

Figure 7 - VERIFICATION PROCEDURE AND TYPICAL SPECTRA FOR SPENT FUEL WITH SHORT COOLING TIME

ranging from 3 months to about 7 years. This work was done in cooperation with several Member States - Bulgaria, Finland, Germany, Hungary, United States of America, Soviet Union, Sweden and Yugoslavia - within Support Programme Tasks or Research Contracts (see e.g. /3,4,5/).

The work started with an evaluation of neutron and gamma detection schemes. Experiments and theoretical studies on the neutron field above a storage rack showed, that this radiation cannot be used because of the strong near neighbor effect /5/. Subsequently, the work concentrated on gamma detectors. NaI detectors, CdTe detectors, ionization chambers and Geiger Mueller tubes were compared. The best performance to cost ratio was obtained for NaI detectors. In the following, the main results obtained for NaI detector based SFAT devices are summarized.

It was found that the performance of the method (defined as sensitivity to detect in a given measurement time spent fuel specific radiation features) depends on several factors:

5.1 Cooling Time and Burnup of the Spent Fuel Assembly to be Verified and its Neighbors

The following cases are considered:

5.1.1 Spent Fuel with Short Cooling Time (≤ 4 Years)

In general, fuel assemblies in this range of cooling time can be verified by CVD. In some cases, when the transparency of the water is low, it might be required to verify also short-cooled fuel assemblies with SFAT.

In this case, the highly penetrating Pr-144 gamma radiation (energy 2186 keV, half life 284 days) can be used for the verification (see Fig. 7). For such an application, the SFAT has to be designed with a heavier weight and longer air-filled pipe (2-2.5 meter). The method performs well for a short cooling time since the Pr-144 radiation is highly penetrating and located in a low background area of the gamma spectrum. Also fast scanning measurements can be considered. With the age of the fuel approaching and exceeding 4 years, it becomes more and more difficult to recognize the Pr-144 signal on the background of pileup of the pulses caused by the strong Co-60 signal. A well designed pulse pileup rejection system of the gamma spectrometer is essential in this case.

5.1.2 Spent Fuel Assemblies with Long Cooling Time (≥ 4 Years)

Cs-137 is used for this verification (gamma energy 662 keV, half life 30.1 years). There are two factors which in this case determine the sensitivity of the method: 1) the absorption of the Cs-137 signal in the material layer above the fuel pellets; and 2) the level of background radiation below the Cs-137 peak. The background radiation below the Cs-137 peak has mainly two origins: 1) compton scattered gamma rays of Co-60 (gamma energy 1172 and 1132 keV, half life 5.3 years); and 2) Pr-144, when fuel assemblies with short cooling times are stored next to long cooled fuel assemblies. The first case is typical for all wet long term storage facilities, the second for some reactor ponds.

It is obvious that in the first case the method performs better the longer the cooling time becomes. The interfering Co-60 radiation decreases with a half life of 5.3 years while the Cs-137 radiation remains much longer. Therefor, it is possible to verify a long-cooled BWR fuel assembly in as short as 10 sec only (see Figs. 8 and 9), while some minutes are required to recognize the Cs-137 signal from a 5-year old fuel assembly.

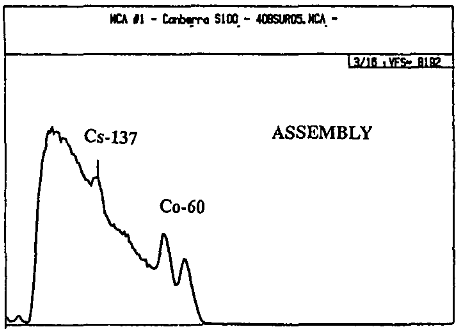

Figure 8 - GAMMA SPECTRUM OF BWR ASSEMBLY WITH LONG COOLING TIME; MEASUREMENT TIME 10 SEC.

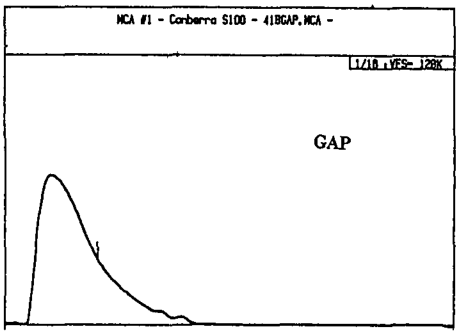

Figure 9 - GAMMA SPECTRUM ABOVE THE GAP

5.2 Design of the Top Structure of the Assembly Determining the Absorption of the Gamma Rays emerging from the Fuel Pellets

On their way from the fuel pellets to the detector, the gamma radiation has to penetrate several material layers - the gas filled plenum above the pellets, the end plugs of the fuel rods, the upper tie plate of the assembly, other structural element which might be present and the water gap needed for safety reasons between the top of the fuel assembly and the lower end of the air filled pipe. Although there are differences in the design of fuel assemblies, we were able to show that the fuel assemblies of the major types (BWR, WWER-440 and PWR) can be verified with this method. However, problems can occur if the absorbing layer is much larger than for the above mentioned predominant fuel assemblies. In this case, it might only be possible to use the Co-60 radiation for the verification.

We also investigated the dependence of the Cs-137 signal on the water gap between the top of the handle of a BWR fuel assembly and the lower end of the SFAT. It is recommended that this gap ideally amounts to about 50 mm but should not to exceed 100 mm.

5.3 Design Features of the SFAT (size, weight, length and diameter of the airfilled pipe, lead collimator/shielding, NaI detector)

The above mentioned design parameters of the SFAT are of great importance for optimizing the measurement method - e.g. to reduce the weight of the device and the measurement time. Although a complete theoretical analysis and optimization of the design features is still under way, we have shown that for long-term storage facilities containing only long-cooled fuel, the weight of the device needs not to exceed about 20 kg. In case of reactor ponds with contaminated water and fuel assemblies with shorter cooling times, the weight of the device might have to be 5 to 10 times larger.

6. Conclusions

A new spent fuel verification method has been developed and demonstrated for the major spent fuel types. From the results obtained, it is concluded that the method can be used for spent fuel assemblies with long cooling times such as those present in wet, long-term storage facilities. The method uses spent fuel specific radiation signatures and does not require fuel movement. It is, however, more intrusive than Cerenkov Viewing Devices, since equipment has to be introduced into the spent fuel pond. More effort is underway to optimize the measurement systems improving the sensitivity and the method, lowering its intrusiveness and extending the method to more difficult application cases like research reactor spent fuel and old fuel surrounded by short-cooled fuel assemblies.

7. Acknowledgement

The authors wish to express their thanks to facility operators in Bulgaria, Finland, Germany, Hungary, and Sweden for assisting in the test of the method.

8. References

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CALCULATION OF Pu 242 FROM ISOTOPIC COMPOSITION MEASUREMENTS BY GAMMA SPECTROSCOPY

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ABSTRACT

Accuracy of measurements for determination of the Plutonium isotopic composition has notably improved in the last few years, due to the progress in hardware and software. In particular the use by the inspectors of Euratom Safeguards Directorate (ESD) of the MGA code (1, 2, 3) has given very satisfactory results for the verification of the isotopic Composition declared by the operators.

The Pu242 is not measured by gamma spectroscopy but calculated with an algorithm. In this paper we present two algorithms used for the calculation of Pu242 in reprocessing plants (one for the Pu reprocessed from PWR elements, one for the Pu reprocessed from MAGNOX reactors). The results of calculations presented in this paper show an improvement in the accuracy of calculated Pu242.

1. INTRODUCTION

La mesure de la composition isotopique du Plutonium est effectuée régulièrement par les Inspecteurs du Contrôle de Sécurité d'Euratom, essentiellement pour l'évaluation des mesures neutroniques ayant pour but de vérifier la masse de Plutonium contenue dans les articles sélectionnés lors des inspections.

La précision de cette mesure a été très notablement améliorée dans les dernières années, par l'utilisation d'unités de détection des rayons gammas de plus en plus performantes (détecteurs, unités d'acquisition et de traitement), et particulièrement par l'utilisation d'un programme d'évaluation (code MGA), qui associé à l'utilisation de systèmes informatiques récents permet des mesures rapides et précises des rapports des isotopes Pu 238, Pu 239, Pu 240 et Pu 241 (références 1, 2, 3).

L'isotope Pu 242 ne peut pas être mesuré par spectroscopie gamma, il est calculé par un algorithme en fonction de la balance des isotopes 239, 240 et 241 - les deux algorithmes actuellement incorporés dans le code MGA (références 4, 5) sont indépendants de la matière mesurée. L'expérience nous a montré que la précision du calcul du Pu 242 peut être améliorée en recherchant des corrélations spécifiques pour des matériels déterminés.

Dans ce papier, nous présentons deux suggestions d'algorithmes tous deux relatifs aux usines de retraitement où, compte tenu du flux de matières et des nécessités du safeguard, l'un des objectifs est la mesure de 100% des articles de PuO2 produits. La première formule s'applique au matériel produit après retraitement d'éléments MAGNOX, la seconde pour le matériel issu du retraitement d'éléments PWR. Les résultats montrent que des corrélations spécifiques apportent une importante amélioration dans le calcul du Pu 242.

II. METHODES UTILISEES POUR LA RECHERCHE DE L'ALGORITHME

La vérification du produit fini (containers de PuO2) est l'un des objectifs majeur dans les inspections effectuées en sortie de procédé avant entrée dans le stockage et/ou lors de leur expédition dans les installations de fabrication.

La vérification quantitative du plutonium est effectuée par mesure neutronique. L'évaluation de la mesure neutronique nécessite la connaissance de la composition isotopique. La DCS utilise des systèmes basés sur la spectrométrie gamma pour mesurer la composition isotopique. Nous avons déjà évoqué l'important progrès dans la précision de la détermination des rapports des isotopes Pu 238 à Pu 241 par l'utilisation du code MGA (réf. 1, 2, 3). Compte tenu des précisions obtenues avec ce code et de la précision de la mesure neutronique, nous avons cherché à améliorer l'évaluation du Pu 242 de telle façon que l'erreur sur la mesure quantitative d'un article basé sur des vérifications indépendantes de l'inspecteur (mesure neutronique, spectrométrie gamma inclus algorithme de calcul Pu 242) soit minimisée.

La table 1 montre l'influence du Pu 242 sur l'évaluation de mesures neutroniques effectuées sur deux articles mesurés avec un compteur UFBR sur des boîtes de PuO2 obtenue à partir de matières d'origine réacteurs PWR.

Table 1

	Lot 1 ******					Lot 2 *****				
	Pu 238	Pu 239	Pu240	Pu 241	Pu 242	Pu 238	Pu 239	Pu240	Pu 241	Pu 242
I.C. (1) declared	0.793	62.706	23.507	9.208	3.786	2.546	50.264	26.984	11.995	8.211
I.C. (2) Pu 242 -30%	0.802	63.446	23.784	9.317	2.650	2.614	51.613	27.708	12.317	5.748
I.C. (3) Pu 242 -20%	0.799	63.199	23.692	9.280	3.029	2.592	51.163	27.467	12.210	6.569
I.C. (4) Pu 242 - 5%	0.795	62.829	23.553	9.226	3.597	2.557	50.489	27.105	12.049	7.800

U.F.B.R. measured PU mass:

with I.C. (1)= 9876 g. Pu
with I.C. (2)= 10107 g. Pu
with I.C. (3)= 10029 g. Pu
with I.C. (4)= 9912 g. Pu

U.F.B.R. measured PU mass:

with I.C. (1)= 10731 g. Pu
with I.C. (2)= 10890 g. Pu
with I.C. (3)= 10836 g. Pu
with I.C. (4)= 10758 g. Pu

On constate que l'erreur additionnelle est pour le lot 1 de 2.3% (lot 2 : 1.6%) pour une estimation erronée de 30% de l'abondance du Pu 242 et de 0.3% (lots 1 et 2) pour une estimation erronée de 5%.

Divers modes de calcul utilisent l'historique des combustibles retraités. Due à la difficulté pour les inspecteurs de vérifier ces informations, et compte tenu de fonctionnement des nouvelles usine en dissolution continue, l'algorithme choisi devait être indépendant de l'irradiation et du temps de refroidissement du combustible.

Enfin, compte tenu du fait que par leurs activités d'inspection, les utilisateurs ont connaissance de la date de séparation des matières à mesurer le principe est d'appliquer les formules de calcul à la date de séparation après mise à jour des balances Pu 238 à Pu 241 mesurées.

Le Pu 242 est forcé à 0 de telle sorte que :

$$(\text{Pu}238)'+(\text{Pu}239)'+(\text{Pu}240)'+(\text{Pu}241)' = 100$$

L'algorithme est appliqué pour le calcul de (Pu242)'.

La composition isotopique finale est calculée pour une normalisation à 100 de la somme (Pu238)' à (Pu242)'.

III. CORRELATIONS PROPOSEES ET PRECISIONS ATTENDUES

L'analyse des données relatives à 5 installations de retraitement où de nombreuses mesures ont été effectuées par les inspecteurs de la DCS a amené au choix de deux corrélations :

- une pour les matières issues de réacteurs MAGNOX
- une pour les matières issues de réacteurs PWR.

Nota : Nous avions proposé originalement une corrélation valable pour les réacteurs PWR et BWR. L'intégration de nouvelles données et une étude statistique plus affinée ont montré que les corrélations sont différentes pour ces deux types de matière.

Dans les deux cas et après avoir intensivement testé des corrélations linéaires, polynominales etc..., il ressort que des corrélations exponentielles donnent les meilleurs résultats.

Pour les matières de réacteurs MAGNOX, la corrélation entre le Pu 242 et les autres isotopes à la date de séparation dans l'usine de retraitement est :

$$\text{Pu } 242' = 3.2646 + (\text{Pu } 239)'^{-1.823} + (\text{Pu } 240)'^{2.075} + (\text{Pu } 241)'^{0.003}$$

Pour les matières issues de réacteurs PWR :

$$\text{Pu } 242' = 1.594694*(\text{Pu } 239)'-1.481 * (\text{Pu } 240)'+1.471*(\text{Pu } 241)'1.010$$

Les tables 2 (réacteurs MAGNOX) et 3 (réacteurs PWR) montrent quelques exemples de calculs sur des lots typiques extraits des banques de données de résultats de mesure.

Les figures 1 (réacteurs MAGNOX) et 2 (réacteurs PWR) montrent les écarts entre les valeurs calculées et déclarées en fonction de l'abondance du Pu 242 dans les matières mesurées.

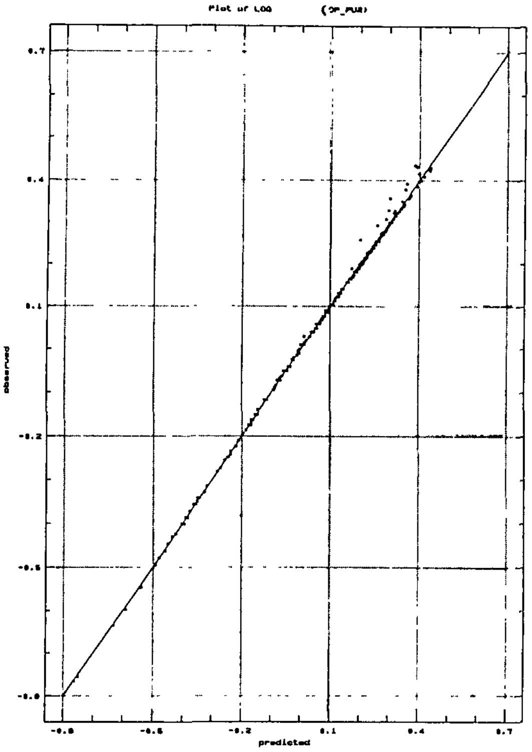

Figure 1

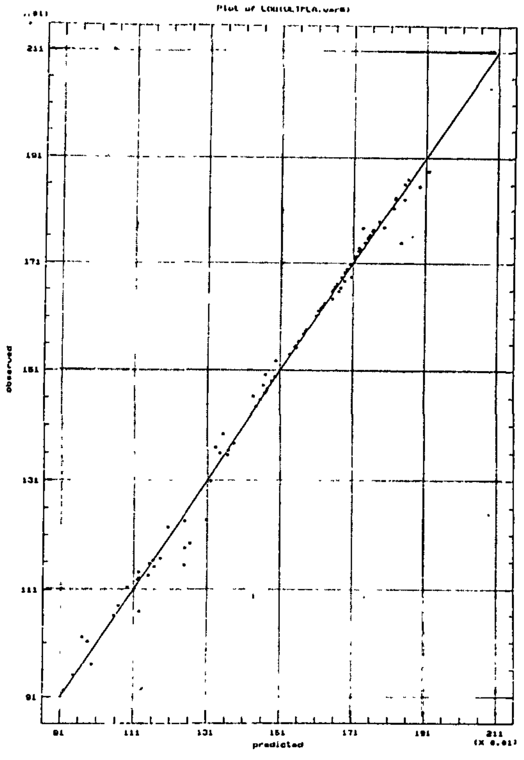

Figure 2

TABLE 2

PU238	PU239	PU240	PU241	PU242	PU242(1)	diff.%	PU242(2)	diff.%	PU242(3)	diff.%
0.295	65.52	27.285	5.375	1.525	1.536	-0.71	1.811	-18.73	1.226	19.58
0.245	68.515	25.255	4.785	1.2	1.204	-0.35	1.364	-13.70	0.985	17.88
0.17	73.585	21.705	3.765	0.775	0.771	0.57	0.800	-3.21	0.736	5.03
0.23	71.255	23.41	4.145	0.96	0.957	0.33	1.013	-5.51	0.808	15.79
0.26	67.565	25.945	4.915	1.315	1.307	0.62	1.481	-12.59	1.041	20.86
0.15	75.085	20.605	3.49	0.67	0.666	0.54	0.676	-0.90	0.640	4.43
0.12	77.42	18.935	2.995	0.53	0.528	0.31	0.501	5.39	0.506	4.45
0.13	77.055	19.245	3.02	0.55	0.551	-0.23	0.519	5.67	0.527	4.22
0.31	64.14	28.09	5.755	1.705	1.697	0.46	2.083	-22.15	1.391	18.44
0.28	66.3	26.75	5.23	1.44	1.442	-0.14	1.687	-17.14	1.159	19.52
0.26	67.64	25.89	4.91	1.3	1.298	0.12	1.473	-13.28	1.037	20.20
0.28	66.38	26.755	5.145	1.44	1.439	0.04	1.656	-14.98	1.135	21.20
					average	0.13		-9.26		14.30
					std.dev.	0.40		8.92		7.04

```
REMARKS : (1) : PU242 = 3.2646*(PU239'^-1.823)*(PU240'^2.075)*(PU241'^0.003) ,
                       new formula , cf. this paper.
          (2) : PU242 = 53*(PU240)*(PU241)/(PU239)^2    ( old MGA formula).
          (3) : PU242 = 150 -2.680647*(PU239') -0.3865709*(PU240') -0.6504233*(PU241')+
                  +0.0120555*(PU239')^2 -0.01155217*(PU240')^2 -0.01151055*(PU241')^2 ,
                      ( new MGA formula ).
```

TABLE 3

PU238	PU239	PU240	PU241	PU242	PU242(1)	diff.%	PU242(2)	diff.%	PU242(3)	diff.%
0.444	67.869	21.033	8.218	2.437	2.568	-5.36	1.989	18.39	1.864	23.50
0.498	68.014	21.118	7.853	2.518	2.458	2.37	1.900	24.54	1.873	25.61
2.456	49.916	26.923	11.842	8.864	9.114	-2.82	6.782	23.486	7.448	15.97
1.815	54.748	24.877	11.965	6.595	6.976	-5.78	5.263	20.194	5.584	15.32
1.780	57.061	25.124	10.434	5.601	5.713	-2.00	4.267	23.812	4.946	11.69
1.763	56.908	24.694	10.889	5.747	5.854	-1.86	4.400	23.438	4.989	13.20
1.511	55.497	25.471	11.085	6.436	6.528	-1.42	4.859	24.504	5.006	22.22
1.417	56.879	25.298	10.598	5.809	5.906	-1.67	4.392	24.392	4.595	20.90
1.133	61.084	25.553	8.268	3.963	4.085	-3.09	3.001	24.27	3.354	15.37
1.749	55.917	24.553	11.525	6.257	6.356	-1.58	4.797	23.338	5.210	16.73
1.152	59.500	23.991	10.461	4.895	4.993	-2.01	3.757	23.240	3.828	21.80
					average	-2.29		23.05		18.39
					std.dev.	2.05		1.87		4.37

```
REMARKS : (1) : PU242 = 1.5947*(PU239'^-1.475)*(PU240'^1.471)*(PU241'^1.0397) ,
                       new formula , cf. this paper.
          (2) : PU242 = 53*(PU240)*(PU241)/(PU239)^2    ( old MGA formula).
          (3) : PU242 = 150 -2.680647*(PU239') -0.3865709*(PU240') -0.6504233*(PU241')+
                  +0.0120555*(PU239')^2 -0.01155217*(PU240')^2 -0.01151055*(PU241')^2 ,
                      ( new MGA formula ).
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Les résultats sur les matières mesurées montrent :

- qu'il n'y a pas d'erreur systématique sur le Pu 242 calculé quelque soit le burn-up, le temps de refroidissement ou la composition initiale du combustible,
- que la déviation standard des écarts entre la valeur calculée et déclarée est inférieure à 5%.

IV. CONCLUSIONS

Les discussions constructives avec MM Gunnink et Ruhter, Wagner et Goerten (voir références) nous ont amené à étudier des corrélations spécifiques pour améliorer la précision du calcul de l'abondance en Pu 242. Les résultats de mesures de composition isotopique effectuées sur l'oxyde de Plutonium produit dans différentes usines de retraitement de la Communauté Européenne ont permis une étude statistique montrant que des algorithmes dépendant uniquement du type de réacteur origine de la matière, permettent d'obtenir des précisions inférieures à 5% dans le cas des réacteurs MAGNOX et PWR. D'autres corrélations seront développées dans le futur pour différents types de réacteur.

Ces résultats permettent d'affiner sensiblement l'évaluation indépendante des résultats de mesures quantitatives de plutonium effectuées par les inspecteurs du Contrôle de Sécurité d'Euratom.

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EFFECTIVE GO/NO GO ENRICHMENT MEASUREMENTS

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1. Abstract

A simple plumbing bypass modification would eliminate excessive systematic error and improve precision for uranium hexafluoride gas enrichment measurements at centrifuge enrichment plants having small-diameter cascade pipes. Present gas enrichment measurements on typical small-diameter product pipes are indeterminate, because overwhelming systematic errors and large statistical errors result from the high deposit-to-gas ratios. The bypass would essentially eliminate the deposit from measurements. The intended purpose of go/no go measurements is to confirm that the enrichment of randomly selected product streams is ≤20%. The Hexapartite Safeguards Project participants agreed by consensus in 1983 on the importance of go/no go measurements.

2. Introduction

Large-scale commercial enrichment of uranium by gas centrifuge technology, which is commercially highly sensitive, began and dramatically expanded in several European countries during the 1970's. For proprietary reasons, technology holders had strong objections to permitting access of IAEA (International Atomic Energy Agency) inspectors into production areas, called "cascade halls". This resistance constituted a serious problem in safeguarding large-scale gas centrifuge enrichment plants; indeed, for large-capacity plants measurement uncertainties implied that materials accountancy procedures alone were not adequate to provide assurance that significant quantities of highly enriched uranium (HEU) were not being produced.

Consequently, international safeguards inspectorates (the IAEA and EURATOM) and technology holders (Australia, Japan, the U.S.A., and Troika - comprising the F.R.G., the Netherlands, and the U.K.) engaged in the Hexapartite Safeguards Project from 1980-83, which consisted of joint discussions for the purpose of reaching consensus on effective and efficient means for safeguarding commercial gas centrifuge enrichment plants. Principal conclusions of the Hexapartite Project and follow-up discussions, reached by consensus, were that: (1) In cascade areas there should be limited frequency unannounced inspections (LFUA inspections), of short duration, and (2) Inspectors should have the right to perform so-called "go/no go" measurements - defined as fairly quick non-destructive assay measurements on cascade-to-header product connection pipes, capable of discriminating between low enriched and highly enriched uranium hexafluoride gas in the pipes.

There was Hexapartite Project consensus that in order to verify absence of production of HEU it could be necessary to perform go/no go measurements. Hexapartite Project participants recognized various possible means of producing HEU that would probably not be detected through LFUA visual inspections. For example, cascade flows could be adjusted to yield higher enrichment than declared, batches could be recycled through a unit cascade to yield progressively higher enrichment, or a cascade that includes increased separative capacity added after the initial verification inspections could be dedicated to HEU production. Since the inspectors must allow the operator up to two hours before gaining access to a cascade hall, visual evidence such as portable feed and withdrawal stations could be confidently removed from a cascade area before entry of inspectors. Consideration of such weaknesses led to Hexapartite Project consensus on the importance of go/no go pipe measurements, intended to confirm that randomly selected streams of product gas are enriched to ≤20% in U-235 /1/.

Unfortunately, in practice, for plants having small-diameter cascade pipes, including the URENCO plants in Almelo (the Netherlands) and Gronau (Germany), go/no go measurements have been repeatedly indeterminate. The principal reason for the indeterminacy is that the high U-235 deposit-to-gas ratios that are common in low-pressure small-diameter process pipes create insurmountable calibration errors as well as very large statistical errors /2&3/. Excessively large, unpredictable calibration errors have not been sufficiently reduced despite years of research.

It is vital that the problem be resolved. Indeed, use of gas centrifuge technology is expanding. It would be a terrible precedent if the present safeguards failure were permitted to persist.

3. Present Measurements

The 186-keV Method

For low-deposit cascade-to-header product pipes of all sizes, simple (1-geometry) 185.7 keV gamma ray detection for U-235, when combined with X-ray fluorescence for measuring the total uranium in the gas, is a reliable and viable method for discriminating between ≤5% and >20% gas enrichments. Use of the 13PO collimator, illustrated below in Figure 1 as the "first geometry", has given particularly satisfactory results /4/. Because the deposit contribution is not subtracted out, the gas assay is systematically roughly 70% high, but statistical errors are small enough for short measurement times that ≈70% bias allows reliable and practical discrimination between 5% and 20% gas enrichment. The method's viability for low-deposit pipes at Almelo is described by the researcher K. van der Meer as follows: "The gas enrichment in pipes with low deposit (1:1) is easily verified with the 186-keV method. In 99.6% of the cases a total measurement time of 1 1/4 hour will be sufficient, if the gas enrichment is 5% enr." /5/

The Deposit Correction Technique

The deposit correction technique, not discussed in this paper, is one of two techniques that has been investigated for go/no go measurements on medium-deposit and high-deposit pipes. With this technique the enrichment of the deposit is assumed known. Experimental results reported by German researchers Lauppe, Richter, & Stein show that, especially for high deposits, the calculated gas enrichment "reacts very sensitively to slight deviations of the deposit enrichment value from the true value."/6/. Since in reality the deposit enrichment is not known and could vary over time with changes in gas enrichment levels, this method would not reliably discriminate between 5% and 20% gas enrichment.

The Two-Geometry Technique

The two-geometry technique was devised to determine the fraction of total measured U-235 that is in the gas, with no assumption on enrichment of the deposit. The diagram below (Figure 1) illustrates the measurement set-up for this technique, which utilizes two different collimators for obtaining two distinct counts of 185.7 keV gamma rays from U-235 in the pipes. The two-geometry technique is used successfully when the U-235 deposit-to-gas ratio is low or moderately low, but (contrary to initial hopes) it fails when the U-235 deposit-to-gas ratio is high, because of excessive measurement errors.

The X-Ray Fluorescence Technique for Total Uranium in the Gas

An X-ray fluorescence measurement determines the amount of total uranium gas in the pipe, which allows conversion of the U-235 gas measurement to percent U-235 enrichment. The X-ray fluorescence measurement utilizes a Co-57 source to excite the uranium K-shell with 122 keV photons; with careful positioning and collimation of both the Co-57 source and the germanium detector, the uranium K X-rays from only the gas are "directly" detected, (although detected K X-ray emissions from the uranium deposit induced by Compton

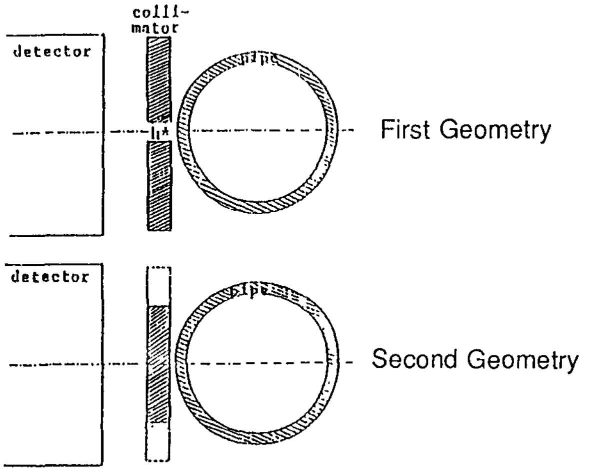

Figure 1. In the two-geometry technique, two collimators having different relative efficiencies for detecting U-235 in the gas vs in the deposit are used separately for obtaining two distinct measurements on cascade-to-header product pipes. (Source: reference 2.)

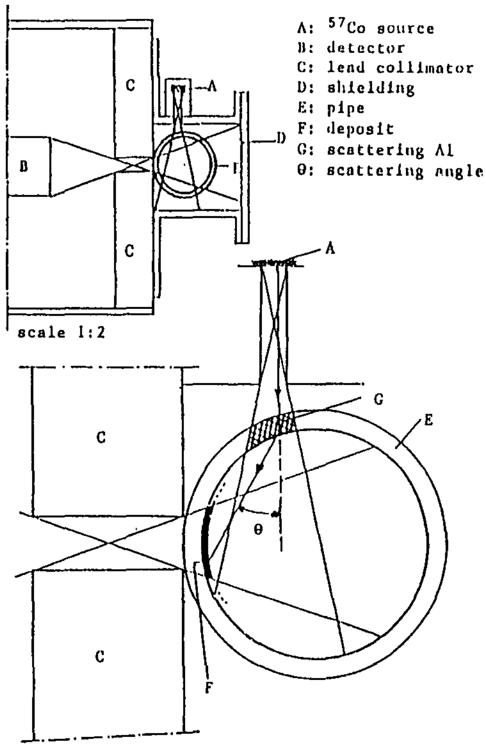

Figure 2: The set up for X-ray fluorescence measurements to determine the total uranium in the gas. The bottom diagram illustrates the possibility of detection of uranium K X-rays emitted from the deposit following stimulation by Compton scattered photons of high enough energy. (Source: reference 7.)

scattered Co-57 photons contribute to the count). In principle the measurement is proportional to the total uranium in the gas. The X-ray fluorescence measurement set-up is illustrated in Figure 2.

The Impact of Deposits on Measurement Errors

High U-235 deposit-to-gas ratios magnify the distortion effect on calculated gas enrichment of any error in the ratio of collimator deposit efficiencies. Indeed, the error magnification is proportional to the deposit, as shown in equation (A3) in the appendix. In practice, at the URENCO facility in the Netherlands, with actual gas enrichment of about 4% U-235, even careful selection and positioning of collimators for application of the two-geometry method has resulted in relative errors of ±200-300% in calculated gas enrichment, for pipes having high uranium deposit-to-gas ratio /8/. This corresponds to an error in calculated gas enrichment of about ±8-12% U-235. With the same deposit, if the uranium gas were highly enriched rather low enriched, the systematic error in calculated gas enrichment would likewise be at least ± 8-12% U-235, (as indicated in the appendix). The presence of systematic errors that are typically of roughly this magnitude obviously renders discrimination between ≤5% and 20% enriched uranium hexafluoride gas extremely unreliable.

For situations where the expected magnitude of relative systematic error is less than about 100%, further analysis is required. The calculated gas enrichment can be expressed in the form

$$\text{Calc Gas Enr} = c_1 R_1 - c_2 R_2 \qquad (1)$$

where R_1 and R_2 are the measured 185.7 keV count rates for the two geometries and the coefficients c_1 and c_2 are constants obtained by dividing formula (A2) in the appendix by an overall coefficient for total uranium determination with the X-ray fluorescence measurement. As shown in formula (A2), the coefficients are derived from estimated collimator efficiencies and efficiency ratios. Estimated efficiencies rather than true efficiencies are utilized for go/no go measurements, because true efficiencies are not independently known by the IAEA, and because (for small-diameter pipes) values of true efficiencies for an actual measurement are sensitive to even slight imprecisions in placement of the measurement apparatus with respect to the pipe /9/.

Let us now summarize the effects of three principal sources of error in two-geometry measurements for small-diameter pipes:

(A) Error in the ratio of collimator deposit efficiencies causes a calibration error (which is reflected in the coefficients c_1 and c_2 in equation 1). It results primarily in a systematic shift of the calculated enrichment from the true enrichment. The magnitude of this shift is proportional to the U-235 deposit, and for moderate or high deposits is relatively many times as great as the magnitude of the input error.

(B) Error in the X-ray fluorescence measurement for total uranium causes a calibration error which affects both count rates equally. If "f" denotes the relative fluorescence error then the relative error in each of the coefficients c_1 and c_2 is $-f/(1+f)$. This error results both in a systematic shift by the factor $-f/(1+f)$ of the calculated enrichment from the true enrichment, and in a multiplicative effect on the statistical error in calculated gas enrichment by the factor $1/(1+f)$. Uncertainties of up to 30% for small-diameter high-deposit pipes are indicated in research results /10/. The practical effect on go/no go measurements is an increase in the necessary measurement time.

(C) Statistical errors in count rates are transformed by equation (1) into statistical errors in calculated gas enrichment. For small-diameter Almelo pipes having a deposit-to-gas ratio of 10, and gas enrichment of 5%, the statistical standard deviation is about 6% enrichment for a 6-hour measurement /11/.
For low-deposit pipes statistical error is much lower, especially if only one measurement (one collimator) can be used. The statistical effect of high deposits is to provide a high background and, through equation 1, to make it necessary to subtract out more than half the gas signal obtained with the first collimator. Consequently, for high-deposit pipes the measurement time must be many times as long to achieve a particular statistical standard deviation as for low-deposit pipes.

It is recognized (and can be simply demonstrated) that statistical standard deviations of ≥6% enrichment do not permit reliable discrimination between ≤5% and >20% gas /12/. Moreover, it is recognized that measurement times much longer than 6-8 hours might not even be effective at detecting HEU gas if HEU production ceases when inspectors arrive. Indeed, as K. van der Meer states, "Due to short equilibrium time of a centrifuge cascade, an enrichment verification measurement which intends to detect a previous production of HEU must be started within a few hours after the change to normal conditions. A second verification (e.g. a follow-up measurement after the first verification indicated HEU) must also be performed within the equilibrium time." /13/.

Consequently, go/no go measurements for very high-deposit pipes could be "reliable" only if systematic errors are essentially non-existent and measurements times are at least 8 hours. This requirement on systematic errors is unrealistic. Accordingly, an alternative approach is needed.

4. Proposed Solution

The problem could be solved by a plumbing bypass modification comprising, at measurement points, the following two features:

(1) Emplacement of a short removable section of pipe (for measurements) in parallel with the existing pipe, and

(2) A system of double valves to securely direct the flow of gas through one pipe or the other.

With this bypass modification (Figure 3), measurements would be made on pipe sections having no or very little uranium deposit. Between measurements, the gas would flow through the normal pipe. If necessary, bypass pipes could be ocassionally removed to be stripped of deposit, to assure that they remain low-deposit pipes.

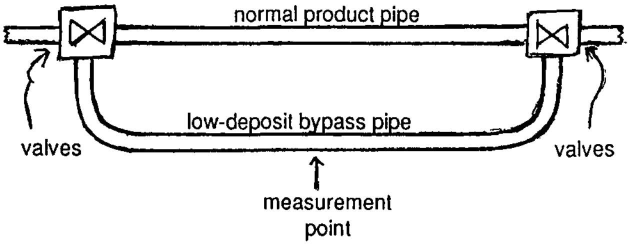

Figure 3. Schematic view of proposed bypass plumbing modification for cascade-to-header product pipes; this modification would greatly reduce calibration and statistical errors, because the bypass pipe would include very little uranium deposit.

It is logical to consider whether a bypass modification for one cascade-to-header product pipe could be used to obtain accurate calibrations for all identical cascade-to-header pipes. This problem is described in the appendix. An essential issue is that the literature indicates substantial uncertainty on the question of whether correct calibration for actual cascade-to-header product pipes is substantially independent of possible nonuniformities in actual uranium deposit. Very limited experiments have produced conflicting indications /14/. Many measurements on actual cascade pipes would be needed to investigate the issue of whether initially "identical" pipes are really identical for calibration purposes. Further, it would be necessary to demonstrate that the declared collimator gas efficiencies ratio is quite accurate--to within 3%. This might pose a problem, because in the past discrepancies of 7% and 14% were found between gas efficiencies at Almelo and at the IAEA Seibersdorf Test Loop/15/In addition, in order to calibrate normal pipes "identical" to the bypass pipe, it would be necessary for the length of the measurements to be very long, in order to reduce statistical errors to very low levels. A relevant question is this: To what extent would the IAEA observe these verifications and calibrations?

As outlined earlier, without bypass constructions, even if systematic errors in the U-235 measurements were practically eliminated, a large proportion of measurements would take so long (more than six or eight hours) that the capability of such measurements to detect rapidly disappearing evidence of HEU production would be dubious. There is now no assurance that even diligent larger-scale research would result in near elimination of systematic U-235 measurement errors, even with the use of several bypass constructions for calibrations. Moreover, the requisite research (having uncertain results) would take a long time, perhaps years.

Effective go/no go enrichment measurements are rendered especially vital by the protection pipe deposits would afford in concealing production of HEU. Indeed, if a plant operator starts HEU production in a cascade with pipes that already have a moderately heavy deposit of low enriched uranium, the rate of increase of U-235 in the deposit is slow, because the rate of new deposition is very slow (roughly a few per cent per year) /16/. Eventually when the deposit contains enough U-235 that inspection measurements might reveal the enrichment of the deposit to be suspiciously high, the operator can move his HEU production from that cascade to another cascade having lower deposit enrichment, and he can clean out the pipes of the previous HEU cascade. Thus, in the absence of definitive gas enrichment measurements on randomly selected cascade-to header product pipes, pipe deposits would serve to prevent detection of HEU production.

5. Conclusion

This paper identifies a practical and highly effective solution to the lingering important problem of excessive errors and consequent indeterminate results in enrichment measurements on high-deposit small-diameter cascade-to-header product pipes in gas centrifuge enrichment plants. The proposed solution involves a simple bypass plumbing construction for every cascade-to-header product pipe. This method would promote the most effective and efficient international inspections, because it would minimize the magnitude of errors and the number of necessary measurements. Moreover, because the technology is well developed, this approach could be implemented quickly and immediately, with no need for time-consuming and uncertain research. Thus, the Hexapartite Safeguards Agreement could be fully implemented quickly in a manner to provide high credibility of safeguards effectiveness.

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/13/ ref. 4, p. 470
/14/ ref. 2, p. 185, and ref. 3, p. 49-50.
/15/ ref. 2, p. 186.
/16/ ref. 11.
/17/ ref. 2, Tables 2 and 3.

7. Appendix: Some Aspects of Two-Geometry Measurements

With the two-geometry technique, for each pipe subjected to measurements, two counts of 185.7 keV gamma rays are obtained, one for each of the two geometries. Specifically, one count utilizes one collimator alone, while the second count utilizes the other collimator alone. (See Figure 1.) If there were no errors from counting statistics, then the following equations would apply:

$$\begin{aligned} R_1 &= e_{1d}D + e_{1g}G \\ R_2 &= e_{2d}D + e_{2g}G \end{aligned} \qquad \text{(A1)}$$

R_1 , R_2 are the counting rates in the two separate geometries, assuming no counting statistics errors

e_{id}, e_{ig} for i=1,2, are the true efficiencies for the two collimators for detection of 185.7 keV gamma rays from U-235 in the deposit and in the gas

G is the actual U-235 gas activity

D is the actual U-235 deposit activity

The actual U-235 gas activity is then represented by the following formula:

$$G = \frac{R_1 - (e_{1d}/e_{2d})R_2}{e_{1g} - (e_{1d}/e_{2d})e_{2g}} \qquad \text{(A2)}$$

For go/no go measurements, this formula is used to calculate the U-235 gas activity from <u>measured count rates</u> and <u>estimated efficiencies</u>.

The following equation expresses the systematic error in calculated U-235 gas activity as a function of the relative error in the ratio of collimator deposit efficiencies (assuming no other errors):

$$G_\alpha - G = k\,D\,\frac{a}{1+ca} \qquad \text{(A3)}$$

G_a = calculated U-235 gas activity assuming error in ratio of collimator deposit efficiencies but no other errors in calibration or measurement

G = actual U-235 gas activity

D = actual U-235 deposit activity

a = relative error in ratio of collimator deposit efficiencies

k, c are constants for a particular cascade pipe (or category of identical pipes) and pair of collimators. These constants depend on actual collimator efficiencies, or efficiency ratios. For example, for some pipes at the URENCO plant in the Netherlands, $k \approx -1.6$ and $c \approx 1.3$, as calculated from reference 2, tables 2 & 3.

When error in the estimated collimator deposit efficiencies is large, it would typically cause most of the systematic error in calculated gas enrichment. In such cases equation (A3) implies that the systematic error in calculated gas enrichment, as percent U-235, is approximately directly proportional to the amount of U-235 deposit. Equivalently, the relative error in calculated gas enrichment is directly proportional to the deposit-to-gas ratio. (In reality, the uranium deposit builds up very slowly.) As a realistic example, suppose that the U-235 deposit-to-gas ratio D/G were 10 if the actual gas enrichment were 4%, that a= ±15% and represents the only measurement error, that k=-1.6 and c=1.3; then for a fixed U-235 deposit, the error in calculated gas enrichment would be ±8-12% U-235, independent of the actual gas enrichment.

If it were demonstrated that in practice inaccuracies in estimated collimator gas efficiencies are indeed nearly always small enough so as not to cause substantial errors in calculated gas enrichment, then whenever the gas enrichment is known for a pipe of high or moderately high U-235 deposit-to-gas ratio, equations (A1) could be used for calibration. Specifically, if the estimated gas efficiencies e_{1g} and e_{2g} are known to be reasonably accurate, then knowledge of G would yield a reasonably accurate value for the calibration coefficient e_{1d}/e_{2d}, the ratio of deposit efficiencies. Indeed,

$$\frac{e_{1d}}{e_{2d}} \approx \frac{R_1 - e_{1g}G}{R_2 - e_{2g}G} \qquad \text{(A4)}$$

where we assume that, by means of very long measurement times, the count rates R_1 and R_2 do not include significant statistical errors.

Let us estimate the magnitude of the relative error in (A4), assuming first that there are no errors in either R_1 or R_2. Let Δ_e be the relative error in e_{1g}/e_{2g} and let Δ_G be the relative error in G. Assume both errors are fairly small, so that we can omit their product in estimating the error in (A4). Then the magnitude of the relative error in (A4) lies approximately between the magnitudes of the two values of $(\Delta_e + \Delta_G)(e_{ig}G)/(R_i - e_{ig}G)$ for i=1,2. We have $e_{1g}/e_{1d} \approx 1.5$ and $e_{2g}/e_{2d} \approx .82$ for some high-deposit pipes at Almelo /17/. In this case, substituting $R_i \approx e_{id}D + e_{ig}G$, we find that the magnitude of the relative error in (A4) is very roughly the absolute value of $1.1(\Delta_e+\Delta_G)(G/D)$. For example, if $\Delta_e = \Delta_G = .03$, errors in (A4) are small: for a U-235 deposit-to-gas ratio D/G of 10, the relative error magnitude is only about .007, while for D/G = 5 the relative error magnitude is still only about .013.

But in reality, especially for small-diameter pipes, there could be significant systematic errors in the count rates if the measurement equipment is positioned just slightly differently for the calibration determination than for actual go/no go measurements. The resulting relative error in (A4) would be greater than the sum of the magnitudes of the relative errors in R_1 and R_2 if the two count rate errors have opposite signs. For example, if the relative errors in R_1 and R_2 are .01 and -.01, the corresponding error in (A4) would have magnitude more than .02. When combined with a relative error magnitude of .01 attributable to deviations in the calibration inputs e_{1g}, e_{2g}, and in G, the magnitude of the resulting relative error in (A4) could be more than .03.

CONTINUOUS MONITORING OF VARIATIONS IN THE ^{235}U ENRICHMENT OF URANIUM IN THE HEADER PIPEWORK OF A CENTRIFUGE ENRICHMENT PLANT

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Abstract

Non-destructive assay equipment, based on gamma-ray spectrometry and X-ray fluorescence analysis has previously been developed for confirming the presence of low enriched uranium in the header pipework of UF_6 gas centrifuge enrichment plants. However inspections can only be carried out occasionally on a limited number of pipes.

With the development of centrifuge enrichment technology it has been suggested that more frequent, or ideally, continuous measurements should be made in order to improve safeguards assurance between inspections.

For this purpose we have developed non-destructive assay equipment based on continuous gamma-ray spectrometry and X-ray transmission measurements. This equipment is suitable for detecting significant changes in the ^{235}U enrichment of uranium in the header pipework of new centrifuge enrichment plants.

Results are given in this paper of continuous measurements made in the laboratory and also on header pipework of a centrifuge enrichment plant at Capenhurst.

1. Introduction

Since the establishment of the Hexapartite Safeguards Project in November 1980, research has been conducted in several countries, including the UK, to develop techniques that could be incorporated into a non-destructive assay instrument that was capable of confirming the presence of low enriched uranium (LEU) in cascade header pipework of a UF_6 gas centrifuge enrichment plant on a rapid 'Go/No-Go', basis. The techniques reported are based on gamma-ray spectroscopy and X-ray fluorescence analysis /1/2/. The development has been complicated by the presence of comparatively large masses of uranium deposited on the pipework, especially on some of the earlier centrifuges.

With the development of centrifuge enrichment technology it has been suggested that on new plant the main header pipework should be continuously monitored to improve safeguards assurance between inspections. Although this type of equipment would be more expensive to install than the transportable system already developed, it would require less effort by both Operators and Inspectors to carry out the necessary inspections.

2. Comparison of off-line and on-line techniques

Where possible the techniques developed for the off-line NDA ^{235}U enrichment monitor, which is used for safeguarding centrifuge enrichment plants at Capenhurst, have been incorporated in the on-line instrument. Both instruments check that the enrichment of the UF_6 gas in the pipe being safeguarded is consistent with being LEU (less than 20%), by measuring the number of 185.72 keV gamma-rays emitted from the UF_6 gas, which is proportional to its enrichment and pressure. As 185.72 keV gamma-rays are also emitted from any uranium that may be deposited on the pipework, it is necessary when inspecting some pipes, especially those on older enrichment plants which may have comparatively large masses of deposited uranium on them, to establish the number of gamma-rays emitted only by the UF_6 gas/2/. A method, known as the "two geometry technique", developed for separating gamma-rays emitted from the UF_6 gas from those emitted from any deposited uranium, has been shown to be suitable for use in both off-line and on-line instruments/1/3/. It was combined with an X-ray fluorescence UF_6 gas pressure measurement in the off-line gauge to confirm that the enrichment of the UF_6 gas in product pipes in the centrifuge enrichment plant at Capenhurst was consistent with being LEU/1/2/. As the on-line instrument incorporates low resolution scintillation counters, rather than the more expensive high resolution liquid nitrogen cooled germanium detector used in the off-line instrument, it is not possible to use the X-ray fluorescence technique to measure the UF_6 gas pressure. Therefore an X-ray transmission technique has been developed, which is capable of detecting changes in UF_6 gas pressure of less than 1 torr in the header pipework in centrifuges at Capenhurst/3/. This is combined with the gamma-ray measurement to check continuously for any changes in the ^{235}U content, and hence the enrichment of the UF_6 gas in product, feed, waste and dump header pipes at Capenhurst.

3. The electronic detection system

The electronic system, which was originally developed at Harwell for use in the mineral industry, uses standard ECB-bus microprocessors and peripherals in conjunction with a specially designed nuclear pulse ADC (256 channels per detector)/4/. Gain stabilisation of the NaI(Tl) scintillation counters, based on the measurement of the 88 keV gamma-rays which are emitted by a ^{109}Cd source, is maintained by the microprocessor in the slave units, each of which may control up to six detectors. The master computer controls the acquisition of data and the setting up of appropriate regions of interest for each of the four detectors. It also calculates the results and compares the measurements with previous values which are kept in an appropriate constants file.

Normally two hourly measurements are made and daily averages calculated. The results are printed out and can also be displayed on the monitor of the computer. A photograph of the four detector assemblies, special electronic system, master computer and printer being used in the service corridor at Capenhurst is shown in Fig 1.

4. Choice of detectors

It was considered both expensive and impractical to install permanently several high resolution liquid nitrogen cooled semiconductor detectors on even all header product pipes. As it is suggested that for some centrifuge plants it may even be necessary to monitor also, feed, waste and dump pipes, it was agreed that only low resolution scintillation detectors would be acceptable for continuous monitoring.

Laboratory measurements showed that scintillation detectors fitted with NaI(Tl) crystals 75mm in diameter and 25mm thick were sufficiently sensitive and could be adequately shielded and permanently installed without causing undue interference to the Operators.

Initially three detectors were mounted on a product header pipe. One detector measured transmitted X and gamma-rays from a ^{109}Cd source, the other two measured 185.72 keV gamma-rays. One detector was uncollimated and the other was collimated, in order to investigate the possibility of using the "two geometry technique" to separate gamma-rays emitted from ^{235}U in the UF_6 gas from those emitted from any deposited uranium/1/.

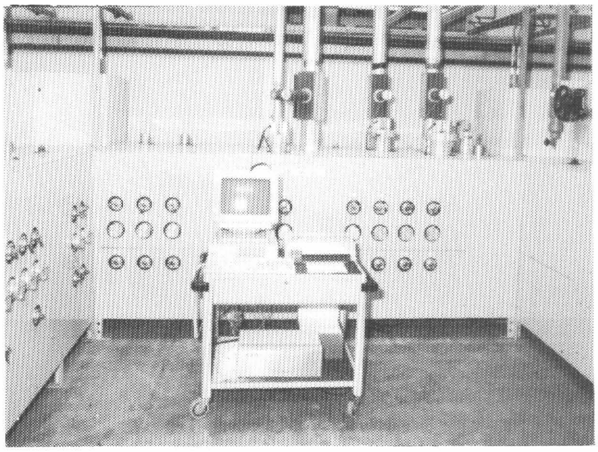


Fig 1 The monitor installed on an enrichment plant at Capenhurst

These trials showed that there was relatively little deposit on the header pipes in the latest design of centrifuge for which this development is aimed. Interest has therefore been concentrated on investigating ways of reducing the overall cost of the equipment while maintaining acceptable performance.

Therefore later measurements have used a single detector fitted with a 50mm diameter crystal to measure both 185.72 keV gamma-rays for determining the mass of ^{235}U in the pipe and the transmitted X and gamma-rays to detect changes in UF_6 gas pressure.

5. Choice of source for the transmission gauge

Header pipes in the Capenhurst centrifuge enrichment plants are made of aluminium, the product, waste and dump pipes have internal diameters of 110mm with 5mm thick walls, while the feed pipe has a 4mm thick wall but is only 72mm in diameter. Therefore any X or gamma-ray used for the transmission gauge must be able to penetrate 10mm of aluminium while being relatively highly absorbed by a few mg/cm^2 of uranium. (Path of UF_6 gas across 110mm pipe at pressure of 10 torr = 1.1 mg/cm^2). Table 1 gives the energy of gamma-rays emitted by radio-isotope sources that

Table 1 Choice of source for transmission gauge

Source	Half Life yrs	Gamma-ray Energy keV	Abundance	Mass Absorption Coefficient cm^2/g		Fraction Transmitted(%) by Al Pipe with wall		Absorbed(%) by 1 torr UF_6 Gas pipe diameter(mm)		
				U	Al	4mm	5mm	55	75	110
^{57}Co	0.74	122	0.855	3.5	0.15	72	67	0.02	0.03	0.04
^{241}Am	433	59.6	0.852	6.4	0.27	56	48	0.04	0.05	0.08
^{109}Cd	1.26	88	0.038	3.3	0.20	65	58	0.02	0.03	0.04
^{109}Cd	1.26	25	0.17	74.0	2.0	1.5	.50	0.40	0.55	0.80
^{109}Cd	1.26	22	1.00	96.0	2.8	.24	.05	0.50	0.70	1.00

were considered for this application and their relative abundance and the mass absorption coefficients for aluminium and uranium for the X and gamma-rays emitted. It also gives the relative intensities of radiation transmitted through aluminium pipes with 4 and 5mm thick walls and also the percentage absorbed by UF_6 gas at a pressure of 1 torr. It is seen that ^{109}Cd is the optimum source as it emits AgK X-rays whose energies 22 and 25 keV are just above the Ul absorption edges. It is seen that a change in UF_6 gas pressure of 1 torr in the largest diameter pipe (110mm i.d.) will reduce the transmitted intensity of AgK X-rays by approximately 1%, in addition approximately 0.1% of them will be transmitted through an aluminium pipe with walls 5mm thick. Table 2 gives the calculated and measured intensities of X and gamma-rays emitted by ^{109}Cd that are transmitted through pipes of different size, where it is seen that there is relatively good agreement between the two values. The advantage of using a thinner walled pipe should be noted, although at Capenhurst this advantage is accompanied by a lower sensitivity due to the smaller UF_6 path length across the pipe. A graph of the channel countrates obtained from X and gamma-rays emitted from ^{109}Cd that have been transmitted through an aluminium pipe with 5mm thick walls is given in Fig 2. It is seen that although the intensity of transmitted 88 keV gamma-rays is much higher than the AgK X-rays, it is still possible to separate them using low resolution detectors.

6. Results

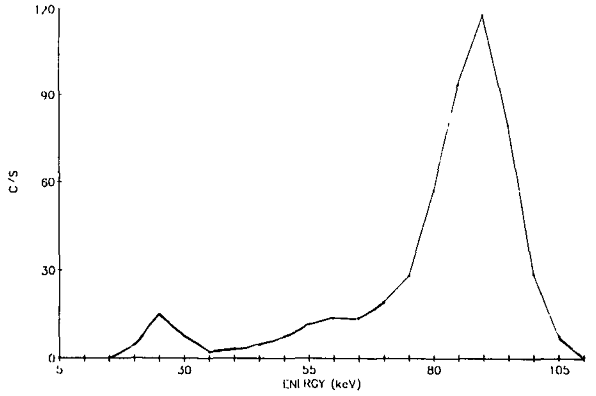
6.1 With a multi-detector system

These measurements were made with three scintillation detectors, two fitted with 75mm diameter NaI(Tl) crystals detecting 185.72 keV gamma-rays, one being uncollimated the other fitted with a 30mm wide collimator. This allowed for the investigation of the "two geometry technique", which separates 185·72 keV gamma-rays emitted from ^{235}U in the UF_6 gas from those emitted from any deposited uranium. The third detector was fitted with a 50mm diameter NaI(Tl) crystal and detected transmitted AgK X-rays and 88 keV gamma-rays which were emitted from a ^{109}Cd source with an activity of 22MBq.

As there were no suitable sources of UF_6 gas available at Harwell, the gamma-ray spectrometers were approximately calibrated using samples of uranium that had been deposited onto filter papers. The transmission gauge was calibrated using aluminium filters.

Table 2 Comparison of the relative transmitted intensities of radiation emitted from ^{109}Cd.

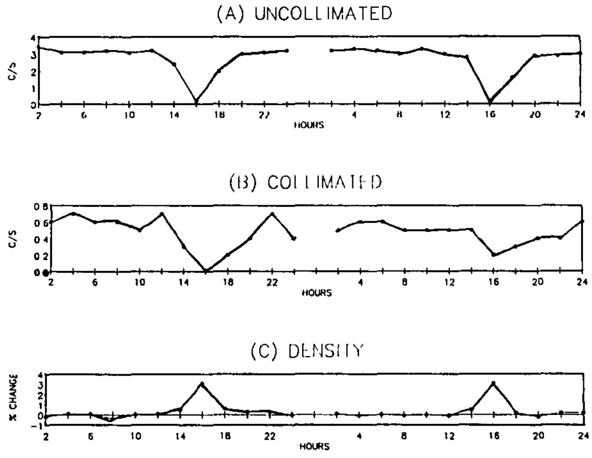
Transmitted Radiation	Relative Transmitted Intensity Calculated Aluminium Wall Thickness (mm) 4	 5	 Measured 4	 5
22 keV X-rays	.08	.02	.11	.05
25 keV-X-rays	.08	.03		
88 keV-gamma-rays	1.1	1.0	1.1	1.0
88/22+25	7.0	20	10	20



AgK X-rays 22,25 keV, 88 keV gamma-rays

Fig 2 Transmission spectrum obtained when measuring aluminium pipes with 5mm thick walls

The assembly was mounted on a header product pipe at Capenhurst and two hourly measurements made between September 1989 and January 1990/3/. The results obtained during two periods when the UF_6 gas was temporarily removed from the pipe are shown in Fig 3. It is seen that when the UF_6 gas was present, the net countrates obtained with the uncollimated and collimated detectors in the 185.72 keV gamma-ray channel were approximately 3.0 and 0.6c/s respectively. The errors due to counting statistics (95%c.l.) were approximately 0.1c/s for both detectors when using a 7200 second measurement time. Both countrates reduced to approximately zero when


(A) UNCOLLIMATED

(B) COLLIMATED

(C) DENSITY

Fig 3 Results of plant measurements made with the multi-detector system during two periods when the UF_6 gas was temporarily removed the UF_6 gas was temporarily removed from the pipe, showing that there was only a relatively small mass of uranium deposited on the pipes. At the same time that the UF_6 gas was removed from the pipe the density gauge reading increased by approximately 3% relative. However if the increase in the density reading, reduction in UF_6 gas pressure, had been accompanied by the gamma-ray countrate either being unaltered, or even increased, then an increase in the enrichment of the UF_6 gas would have been suspected and further investigation may have been needed to resolve the anomaly.

During this period of continuous measurement only three significant increases in the density readings occurred, indicating a reduction in UF_6 gas pressure. They were all accompanied by a large reduction(over 95%) in the gamma-ray counts confirming the loss of UF_6 gas. There were no significant increases in the gamma-ray measurements during the same period so that it was possible to confirm that this centrifuge was only producing LEU gas during this period.

6.2. With a single detector system

The mass of uranium deposits on the pipes of the latest design of centrifuge have been shown to be comparatively small/3/. It was therefore decided that there was no necessity to make gamma-ray measurements with a collimated detector in order to separate gamma-rays that were emitted from the UF_6 gas from those emitted from the deposited uranium. Furthermore, if it was found to be possible to make the uncollimated gamma-ray and transmission measurement with the same detector, it would result in a considerable reduction in the capital cost of the system.

The original reason for not using a single detector was the possibility of 'double peaks' produced from the relatively high number of transmitted 88 keV gamma-rays from the ^{109}Cd source not being resolved from the 185.72 keV gamma-rays emitted from ^{235}U. The magnitude of this interference is shown in Fig 4, where the spectra obtained from a pipe,110 mm in diameter, containing 0.5mg/cm^2 of 3% enriched uranium, equivalent to a UF_6 gas pressure of approximately 13 torr, is shown when measured without a ^{109}Cd source. Also shown are the spectra obtained with two ^{109}Cd sources of different activity measured without uranium. As the pulse doubling effect is a function of the square of the 88 keV countrate the interference is much greater for the higher activity source (30MBq). The error introduced using the smaller source (10MBq), an acceptable activity for this application, is comparatively small (equivalent to the counts obtained from UF_6 gas at a pressure of approximately 2 torr of 3% enriched UF_6 gas). This error can be allowed for by subtracting a fraction of the number of detected 88 keV gamma-ray counts from the 185.72 keV reading. Sources of ^{109}Cd with activities of approximately 5MBq were installed in the two detector systems that were to be used on the product and waste pipes and sources with activities of 10MBq in the feed and dump pipe systems.

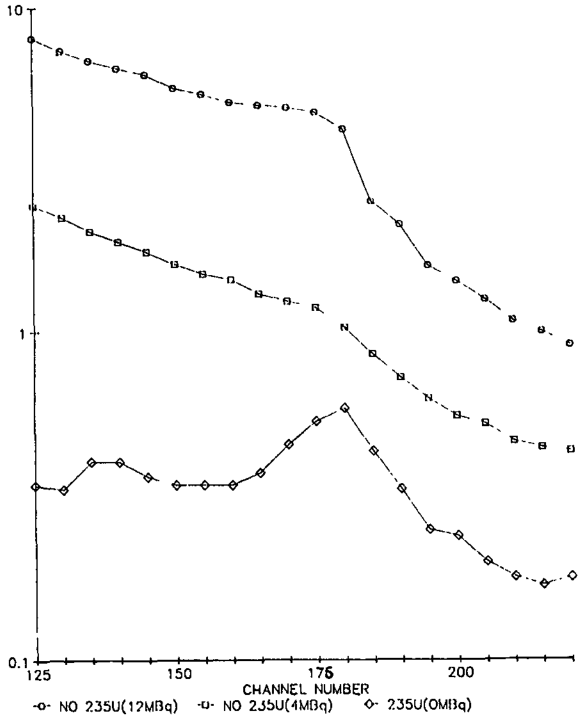


Fig 4 Interference in the ^{235}U 185.72 keV gamma-ray channels(163-192) due to 88 keV gamma-ray "double peaks" from ^{109}Cd sources with different activities

Measurements were made on empty pipes at Harwell when the net countrates obtained in the 185.72 keV gamma-ray peaks were corrected by subtracting approximately 6 x 10^{-8} times the square of the 88 keV gamma-ray countrates. The results of two hourly measurements of corrected 185.72 keV gamma and relative AgK X-ray counts made in the laboratory over a period of 5 days for the 4 detectors are shown in Fig 5. The errors due to counting statistics (95% c.l.) were approximately 0.08 c/s for the 185.72 keV gamma-ray measurements

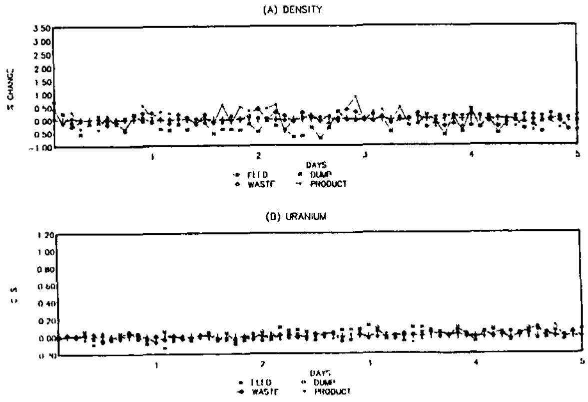


Fig 5 Results obtained in the laboratory when measuring empty pipes

and between 0.3 and 0.12 % relative for the AgK X-ray measurements depending on the activity of the ^{109}Cd source that was incorporated. It is seen that the percentage variations in the density readings are generally within ± 0.5%.

The equipment was then installed on header pipework at Capenhurst, two hourly measurements made and daily averages calculated. Corrections for dead time losses, the decay of the ^{109}Cd source and for unresolved 88 keV 'double peaks' on the gamma-ray measurement were made. A plot of variations in the transmission density and uranium gamma-ray measurements during two periods when the UF_6 gas was temporarily removed from the pipes is shown in Fig 6. It is seen that when the UF_6 gas was removed the transmission density gauge readings on the product, waste and dump pipes increased by approximately 3% relative. The corresponding increase in the smaller diameter, thinner walled, feed pipe was slightly smaller. There was a corresponding reduction in the gamma-ray reading on the product pipe of approximately 1c/s, one third of that obtained on the original gauge. This is due to the smaller diameter detectors used, 50mm in diameter instead of 75mm, in order to reduce the cost and also the weight of the lead shielding required. There were corresponding smaller reductions in the gamma-ray counts obtained with the other detectors, being consistent with that expected according to their pipe diameters, pressures and enrichments. The daily average readings of both density and uranium over the period 6/12/90 to 10/3/91 for the product pipe are given in Fig 7. The periods when the UF_6 gas was temporarily removed from the pipe are clearly identified by increases in the transmitted countrates, indicating a reduction in UF_6 gas pressure and a reduction in the number of detected uranium gamma-rays. The times when the enrichment of the UF_6 gas in the product pipe was temporarily increased, and then decreased, by

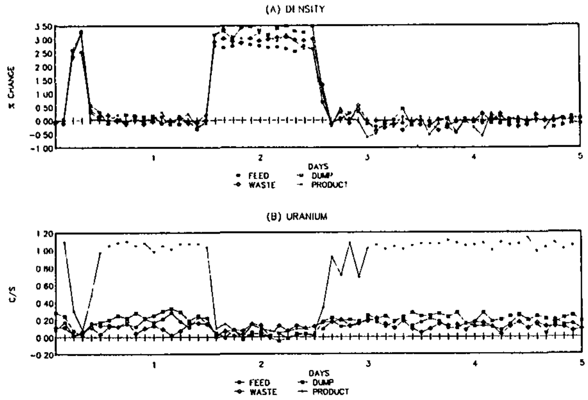


Fig 6 On-line two hourly results obtained during the time that the UF_6 gas was temporarily removed from the pipes. Periods identified by high transmitted counts (low density) and low ^{235}U counts

Table 3 Print of time when UF_6 gas was temporarily removed from the pipes.

Run started at 9.40 6/12/90 Run time = 7200s

FEED			DUMP			WASTE			PRODUCT		
S	D	U	S	D	U	S	D	U	S	D	U
0.6	-0.08	-0.08	0.9	-0.33	0.06	0.8	-0.06	0.00	0.8	-0.08	-0.02
0.4	0.02	0.01	-0.4	-0.21	0.02	-1.6	-0.12	-0.00	1.5	-0.04	0.03
2.2	2.33	-0.15	1.5	2.34	-0.15	4.3	2.36	-0.09	1.5	2.30	-0.76
3.6	2.59	-0.12	1.4	2.99	-0.20	4.3	3.39	-0.05	1.2	3.14	-0.98
3.2	0.35	-0.00	1.3	0.32	-0.07	-0.8	0.40	0.00	1.3	-0.00	-0.58
1.0	0.10	-0.03	0.3	0.07	-0.05	-1.1	0.14	-0.08	0.2	-0.01	-0.09
2.7	0.03	-0.02	-0.9	-0.37	-0.02	-1.6	0.20	0.01	2.0	-0.04	-0.01
1.6	0.08	-0.04	1.2	-0.26	0.03	-2.2	-0.01	0.01	1.2	0.16	0.02
2.1	0.01	-0.01	1.2	-0.03	0.00	-1.3	-0.13	0.07	1.5	-0.24	0.04
2.4	0.07	-0.04	-0.1	-0.06	0.06	-1.9	0.16	-0.07	0.4	-0.08	-0.01
1.3	0.01	0.05	-0.5	-0.11	0.00	-2.2	-0.14	-0.01	2.0	0.00	0.02
1.8	0.13	0.01	0.1	-0.07	0.03	-2.4	-0.05	0.02	1.3	0.01	-0.08
Averages:											
1.9	0.47	-0.04	0.5	0.36	-0.02	-0.5	0.50	-0.02	1.2	0.43	-0.20

S, D and U are the differences between the measured and constant file values of standard, relative density (%) and ^{235}U respectively.

N.B. All daily average density readings are positive and all uranium readings are negative with respect to the constant file values observed during normal operation. This confirms that UF_6 gas had been removed during these measurements.
(The times that the gas was removed can be obtained from the individual two hourly values.)

20% relative without significant change in the UF_6 gas pressures, are also clearly visible.

There were small systematic variations in the density readings (up to 0.8%) during the 3 months of measurements, possibly due to instrumental drift or to small errors in the decay correction used for the ^{109}Cd source due to small impurities in the sources. Therefore an alternative method of correcting the density gauge measurements was used, namely by taking the ratio of detected AgK X-ray to 88 keV gamma-ray intensities. This automatically corrects for the decay of the ^{109}Cd source as well as for dead time losses. This method was found to give small systematic errors as can be seen in Fig 7. As there are no significant systematic errors on the ^{235}U determination, it appears that the correction used for correcting the 185.72 keV gamma-ray readings for 88 keV gamma-ray interference is adequate for this application.

6.3 Presentation of results

The results obtained are compared with previous values kept in an appropriate constants file. Variations from these values are at present saved as files on a two hourly and daily average basis and printed out as shown in Table 3. They can also be displayed across the screen of the PC. The chosen one of the four density or uranium readings is selected from the keyboard. Provided that an Inspector was confident that the equipment was working satisfactorily, (by checking the relative counts in the standard channels), he need only be concerned if the pressure of the UF_6 gas in the pipes decreased significantly, (by at least a factor of 2), while at the same time the uranium counts remained the same or increased. These rare occasions are probably the only time that the results need to be printed out from the data files. This approach will be more relevant if one PC and printer is used to measure continuously the four pipes on several cascades. Alternatively the files could be printed out on demand during routine inspections.

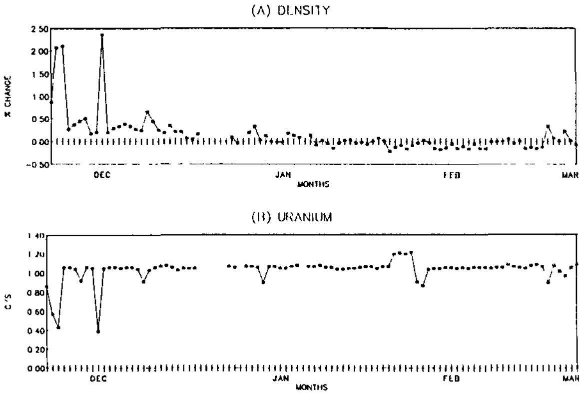


Fig 7 Daily average on-line results obtained from product pipe. Periods when UF_6 gas was removed for less than a day identified by high transmitted counts (low density) and low ^{235}U counts. Periods of 20% higher and 20% lower ^{235}U enrichments identified by corresponding changes in ^{235}U counts without significant change in the transmission density measurement

7. Conclusions

Continuous monitoring of at least one header pipe of a cascade at Capenhurst has been carried out in two periods between September 1989 and January 1990 and between December 1990 and March 1991. During these periods the UF_6 gas pressure measurement has been significantly reduced on less than 10 occasions. On each occasion the uranium countrate also reduced, almost to zero, confirming the Operators' records that the UF_6 gas had been temporarily removed.

If an inspector had access to these results, it would have been possible for him to have monitored this centrifuge, 24 hours a day, for the period of six months for which these measurements were made. Although the initial capital cost of this type of automated monitoring equipment is comparatively high, it is suggested that the cost would soon be recovered by the saving of Operator and Inspector time.

8. Acknowledgements

The author would like to record his thanks to M R Wormald of AEA Technology for providing and programming the detection equipment. Thanks are also given to K Connor and M Eastell of British Nuclear Fuels and G Preece of AEA Technology for their help in making the measurements and presenting the results.

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MONITORING OF NEUTRON AND GAMMA FIELDS INSIDE POWER REACTOR SPENT FUEL ASSEMBLIES BY SMALL-SIZE DETECTORS

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Abstract

A set of small-size solid state and activation detectors was used for monitoring transuranium and fission product contents in spent fuel assemblies of a WWER-440 type PWR in Paks, Hungary. Neutrons and gamma-radiation were detected inside and on the top of the fuel assemblies by means of track etch and bubble damage detectors for fast neutrons, indium activation detector for thermal neutrons and hard gamma-rays as well as semiconductor Si diodes as gross gamma-detectors.

1. Introduction

At the previous ESARDA Symposium, 1989, a report was given on preliminary studies concerning the in-field application of novel solid state and activation detectors as possible attribute test methods for NDA of spent fuel assemblies. Indium activation (neutron and gamma), track etch and bubble damage neutron detectors as well as semiconductor silicon diodes as gross gamma detectors were applied on the top of and inside spent fuel assemblies of a WWER-440 type PWR in Paks, Hungary/1/. The small size and ease of application make the use of these detectors attractive, allowing free access to inner part of assemblies. In addition, using fast neutron sensitive track etch and bubble detectors the cross-talk from neighbouring assemblies can be minimized, facilitating verification without fuel movement.

2. Experiments at NPP Paks

Assemblies of three different burnup ranges (about 10, 20 and 30 MWd/kgU), three initial enrichments (1.6, 2.4 and 3.6 %) and cooling times in the range of 1.5 month to 3 y were assayed. The assemblies were arranged vertically either stacked in storage rack or separated from neighbours on sidewall of the pond. P" (proton sensing) CR-39 track etch detectors type MA-ND made of organic polymer by MOM, Budapest, and/or metallic indium foils in stainless steel capsules were inserted for irradiation into the 8.9 mm diam. central dosimetric channel of the assemblies with the help of a 13 m long special guide tube used also earlier/1/. Boron loaded and polyethylene radiators were applied on 3 mm wide stripes of the CR-39 material.Exposure time varied from 10 mint to 2 h. Indium samples of 2 cm diam. by 0.5 mm thick (folded up in the capsule) were exposed for 1 to 10 h. After irradiation the In foils were unfolded and placed in contact with a 35 cm^3 Ge detector for 10000 s measurement of 336 keV and 417 keV peaks of ^{115m}In and ^{116m}In, respectively.

It was recognized that the high gamma-background may cause heavy damage and a drastic change in the bulk etch rate and in the detection sensitivity of the CR-39 material/2/. In order to avoid overexposure, irradiation times were established with the help of a Si diode inserted precedingly into the central channel of assemblies for measuring the gamma-dose-rate inside. The sensitivity of the diode was 22 nA/kGy/h. In this way the track etch detectors were prevented from receiving doses over 2 kGy.

The diode was at the same time used for scanning gross gamma-profile of the assemblies, as well as bigger (more sensitive) diodes were also used lowering down to the assembly heads. The diode response was confirmed by Al_2O_3 thermoluminescence dosemeter pills of 6.4 mm diam. The pills were cut half to go in the capsules when axial scanning was performed. Exposures were not allowed to exceed 1 kGy for TLD. Bubble damage detectors and In foils surrounded by a Be photoneutron converter were also irradiated on the top of assemblies in steel or plastic containers.
The Be converter was the same as described earlier/1/.

The influence of the gamma-background is much less in the case of bubble detectors/3/. The effect of radiation damage can be neglected for exposure times relevant to irradiations by spent fuel, but it seemed necessary to use some kind of thermal insulator because of the strong temperature dependence of the response. Zero energy threshold type BD-100R detectors of Bubble Technology Industries, Canada/4/ were used, the performance of which was tested both under laboratory and in-field conditions in a power and a research reactor/1,5/.

3. Results and interpretation

Axial neutron and gamma-profiles of two assemblies of about 30 MWd/kgU burnup, taken by In activation, are given in Figs.1 and 2, respectively. The neutron profiles follow thermal neutron distribution inside assemblies. Neutrons are mainly due to ^{242}Cm (half-life: 163 d) content. Thermal neutron fluxes deduced from saturated count rates of 417 keV gamma-peaks were estimated in the order of 10^4 cm^{-2}s^{-1}. The gamma-profiles follow mainly ^{140}Ba content, i.e.

reactor power distribution before shutdown/6,7/. In Fig.3 the results of bulk etch rate measurements on CR-39 material are also plotted. The ratio of bulk etch rates before to after irradiations can be used as a measure of the gamma-dose received/2/.

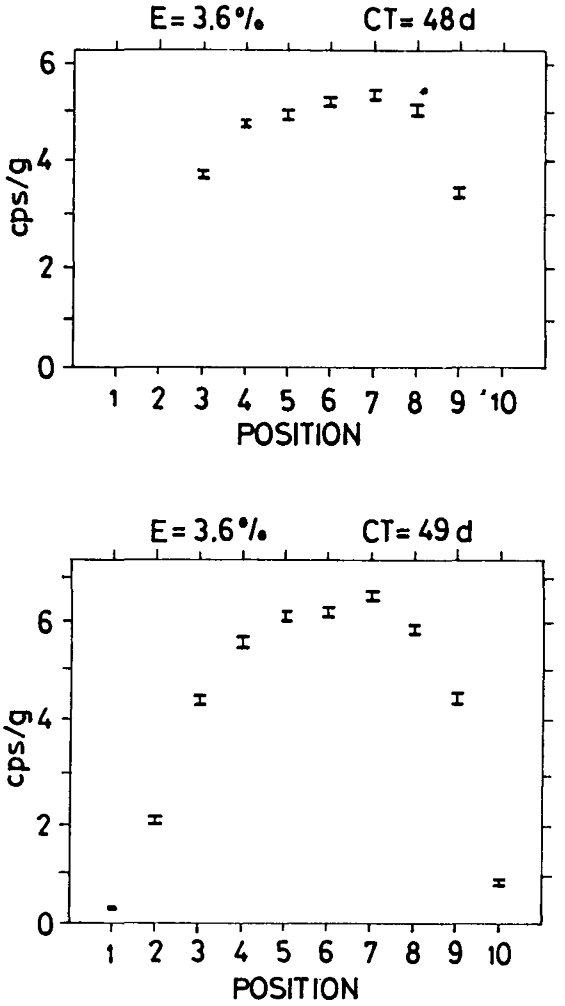

Fig.1. Axial distribution of thermal neutrons inside assemblies of 30 MWd/kgU burnup taken by 417 keV (^{116m}In) gamma-ray counting. The profile follows ^{242}Cm distribution

Fig.3 shows the axial distribution of thermal and fast neutron inside two fuel assemblies measured by CR-39 track etch detectors/2/. Although there are minor discrepancies, the possibility of neutron flux measurements in readily demonstrated.

Correlation between track density and burnup was studied for eight assemblies with various burnup, initial enrichment and cooling time. Mean track densities were assigned to mean burnup values of each assemblies according to the empirical formula

$$N \sim a(BU)^b \qquad (1)$$

giving neutron response N as a function of burnup BU. Burnups values were calculated for each assemblies by the staff of Reactor Physics Department of NPP Paks using the In-Core Fuel Management Code "C-Porca"/8/. The evaluation of the results is in progress.

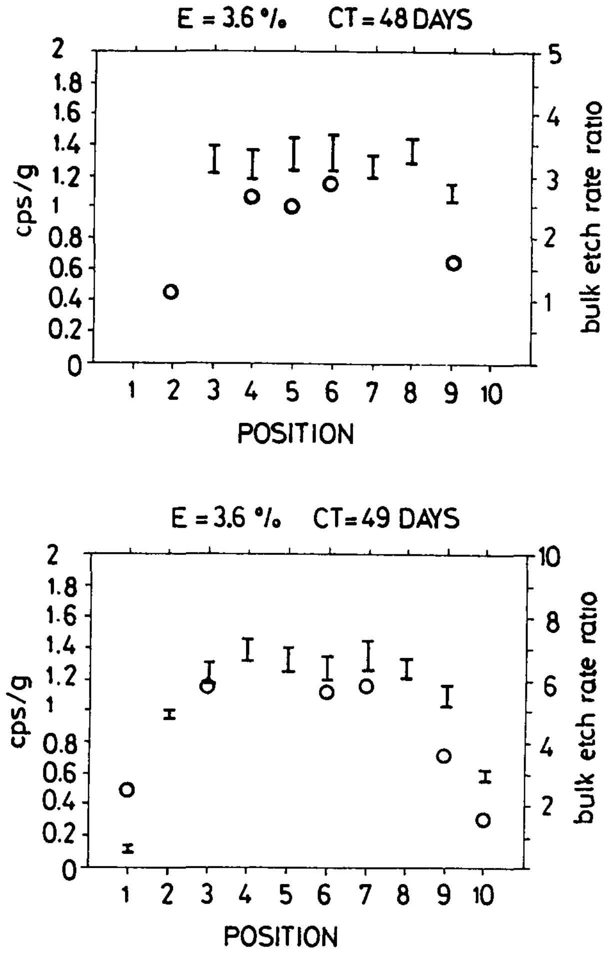

Fig.2. Axial distribution of gamma-radiation inside assemblies of 30 MWd/kgU burnup. Values given by error bars (90 % confidence level) were obtained by 336 keV gamma-ray intensity (^{115m}In) measurements. This profile follows the distribution of ^{140}Ba content, i.e. power distribution before shutdown. Circles represent CR-39 bulk etch rate ratio measurements (ratio of rates before to after irradiation)

The results of the bubble detector irradiations performed on the top of the assemblies showed similar results as earlier/1/. Although the detectors were carefully prevented from getting warm, thermal insulation was not quite perfect and results of considerable spread were obtained. Typical exposure time was 10 min. The production rate of bubbles was 4-5 min^{-1} at about one year cooling time. We are going to apply a new insulation upon the next run.

Studies were performed with measured data provided by the Si diodes. Fig.4 shows axial gross gamma-profiles taken by a diode inside three assemblies, stored among neighbours and separately. The profiles exhibit a near-neighbour effect of about 15 % or less, depending on the assemblies' environment. A comparison with results of TLD measurements shows a reasonable agreement.

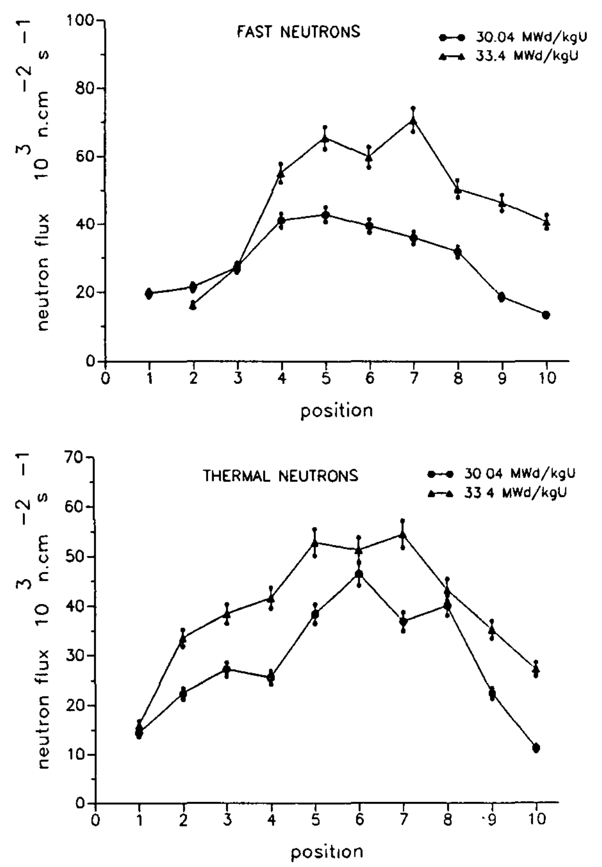

Fig.3. Axial distribution of thermal and fast neutrons inside two fuel assemblies (cooling time: 475 d) measured by CR-39 track etch detectors

The correlation between the diode-yielded dose rate data and calculated burnup values provides a verification possibility for the cooling time on the basis of the empirical formula

$$G = 75BU/T, \qquad (2)$$

where the gamma response G is converted to Gy/h and cooling time T is given in years. Results of the axial scans of individual assemblies with and without neighbours around are also taken into consideration if Fig.5, where GT values are plotted versus burnup. A straight line can be fitted to the data with a standard deviation of 15 % including also near-neighbour effect.

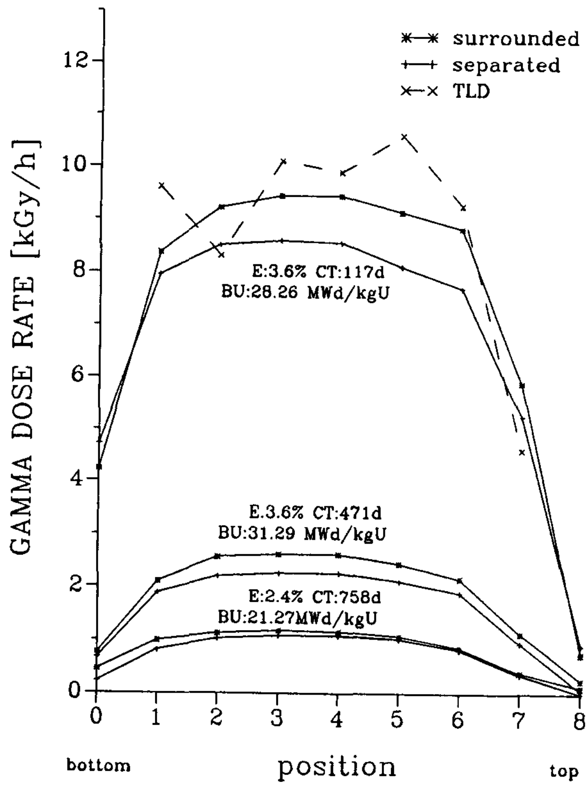

Fig.4. Gross gamma-profiles taken by Si diode inside three assemblies stored among neighbours and separately. Comparison is given with results of Al_2O_3 TLD measurements

4. Conclusions

Indium activation is a rather reliable method, allowing instant reading of the results. Near-neighbour effect is, however, large because of its sensitivity to long-range thermal neutrons and hard gammas in water.

Bubble detectors seem to be promising because of promptly readable results with short exposure time, but some improvement (thermal insulation) is needed for a routine field application.

Track etch detectors are almost insensitive to high gamma background. They give reliable results in a reasonable exposure time, but the results are not immediately available on the site.

Si diodes provide fast, reliable results with a prompt reading. Three of the methods (use of indium activation, track etch and bubble detectors) are spent-fuel specific.

The study of near-neighbour effect by Si diodes yielded favourable results; the method seem to be suitable for performing attribute test in verification without fuel movement.

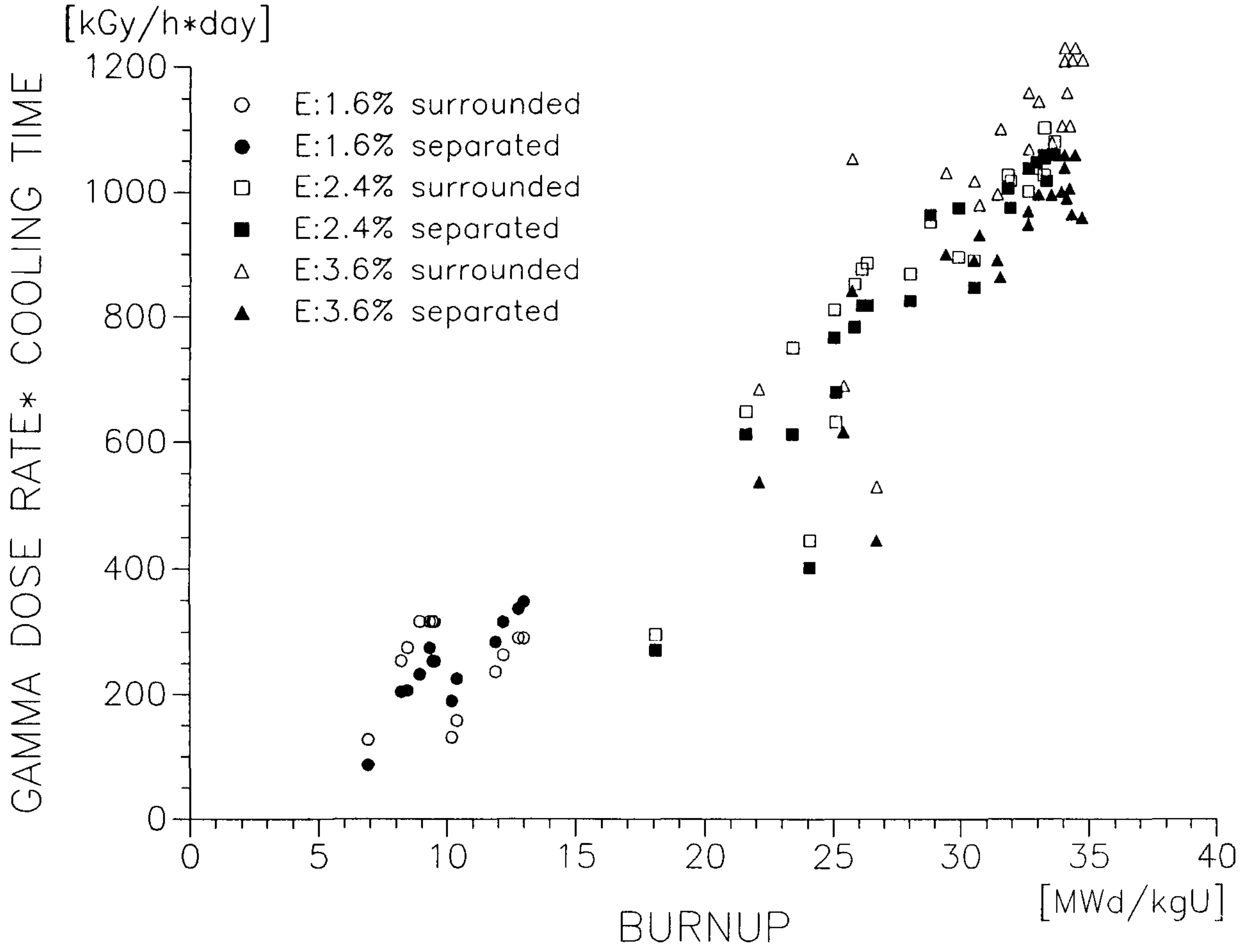

Fig.5. Results of axial scans with a Si diode showing the product of the gamma response and cooling time as a function of burnup

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CROSS CALIBRATION OF A SPENT BWR REFERENCE FUEL ASSEMBLY

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Abstract

A spent BWR reference fuel assembly of the TVO KPA-STORE in Finland was cross calibrated for burnup using spent fuel of a similar design in an other country, namely the CLAB in Sweden. The reference assembly is used by the IAEA for inspection purposes. The verification was performed by passive neutron detection using GRAND I and a fork detector. High resolution gamma spectrometry was used to give complementary information. Normalization between different measurement geometries was performed using sealed neutron and gamma calibration sources.

Based on the results of passive neutron detection, consistency between the declared burnup data of the reference assembly and the selected assemblies used for the calibration curve is better than 1 %.

1. Introduction

Non-destructive assay (NDA) of spent fuel is an essential part of the verification methods used by the IAEA in the international safeguards. Due to the limited application of reprocessing, more and more spent LWR fuel will accumulate in separate storage facilities. It is known that the strategic value of spent fuel increases with the increasing of cooling time. For verification of ageing spent LWR fuel, a quantitative method called GBUV /1/ has been developed for use by the IAEA. This method is based on high resolution gamma spectrometry (HRGS) and it allows consistency checking of the declared burnup data. The linear dependance of the measured average Cs-137 activity (662 keV) on the burnup is used for verification. If combined with passive neutron assay (PNA), partial defect verification of LWR fuel can be performed according to the present safeguards criteria of the IAEA.

GBUV is directly applicable at the AFR facilities of Sweden (CLAB) and Finland (TVO KPA-STORE) where a similar type of BWR fuel will be stored for several decades.

It is a general practice in analytical work to use certified standards for quantitative measurements. Following this analogy, a spent fuel reference assembly has been selected and separately sealed by the IAEA at the TVO KPA-STORE in Finland /2/. Using the reference assembly, measurements performed at different times and by different equipment can be normalized with each other. Measurement of the reference assembly is also a part of the authentication measures of the GBUV method.

The verification level of the GBUV method can be raised by getting more information of the reference assembly. Because separate fuel assemblies cannot be moved in practice from country to country, cross calibration with spent fuel of a similar design at an other country has to be arranged by transporting the instruments and the calibration sources. It was felt that two independent methods would give a much higher degree of confidence than just one method. The methods selected, the PNA and HRGS, can be easily used at the facilities where these measurements were performed.

This report gives an overview of the calibration activities.

2. Measurement Techniques

The two independent methods used give information of completely different parameters. In the PNA, neutron emission of transuraniums is detected. These isotopes are produced mainly from the fertile U-238 under neutron irradiation. The main method used, GRAND I and a fork detector, is well known and widely used e.g. by the EURATOM for the verification of spent LWR fuel /3/. In the other method used, the gamma emission of fission products is detected. These isotopes are produced mainly from fissile isotopes, e.g. U-235 and Pu-239. The HRGS techniques were used according to the principles of the GBUV method /1,4/.

Calibration was performed by first measuring the spent fuel assemblies and calibration sources at the CLAB. After this the sources and the measurement instruments were sent to the TVO, where the reference assembly and the sources were measured. Use of the calibration sources was an essential part of the calibration procedure.

2.1 PNA Equipment and Method

The basic principles of the GRAND I and the fork detector are the same as originally designed at the LANL for the IAEA /5/. The new BWR fork detector of the IAEA is, however, optimized for the dimensions of the BWR fuel measured. It makes use of an AMPTEK preamplifier/discriminator among other changes.

For transport, the detection head can be bent 90 degrees and it fits into a flat transport case. The detection head was attached to a 10 m long steel pipe assembled from 2.4 m long standard pipe sections with connecting flanges /5/. The detection system was assembled on the floor of the storage hall and lifted into the storage pool by a crane. At the CLAB the fork detector system was attached to the pool

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railing with a mounting bracket. At the KPA-STORE an instrument stand used by the operator was used to hold the pipe system. The GRAND I electronics package was used to control the measurements.

A Cf-252 neutron source (about 1.8 x 10^6 n/s), designed for spent fuel verification measurements, was used for normalization. The source, constructed inside a piece of a real BWR fuel channel, was easy to manipulate manually under water. It was repositioned inside the fork detector and remeasured every morning and every evening to find out the precision of the normalization measurement and also to monitor the proper functioning of the whole measurement chain. 200 sec measurement time yielded about 10 000 counts in each of the two neutron channels of the fork detector. Before the measurements at the CLAB, the source was sealed by an IAEA metal seal. It was kept in place until all the measurements at the CLAB and at the KPA-STORE had been finalized.

2.2 PNA Measurements

The fork detector is designed for point wise measurements. Vertical coordinates of the fuel were fixed using a dummy assembly. The assemblies were hanging from the fuel handling machine during measurement. All assemblies were measured in the middle of six odd numbered nodes 11 - 21 (the active fuel length, 3680 cm, is divided into 25 nodes). After rotation of 90 degrees, the measurements were repeated. The selection of the measurement mode was based on the results of the preliminary measurements of the burnup profile.

The measurement time per point varied between 15 and 150 seconds depending on the burnup and the cooling time. At least 10 000 neutron counts were collected.

The assemblies measured were selected to have an enrichment close to 2.7 % and a cooling time more than three years (see Table I). Due to long cooling time and high burnup, the dominating neutron activity is Cm-244. This is why the measured count rates of the cadmium wrapped fission chambers used were corrected only for decay of Cm-244. The count rate of the reference assembly was also normalized with the Cf-252 source. The average activity of the 12 points measured of each assembly was used. A Symphony spreadsheet was used for data handling.

The fitting of the calibration curve was performed by the Deming curve fitting code /6/. The statistical uncertainty of the neutron count rate and the 2.5 % uncertainty of the declared mean burnup were used for weighting the data points. Weighting factor is the reciprocal of the square of the uncertainty.

2.3 HRGS Equipment and Method

Both facilities have a gamma scanning arrangement consisting of a horizontal collimator installed in the wall of one of the fuel handling pools and a specially designed elevator, "gamma wagon", which holds the assemblies during scanning /2,4/.

The measurements were performed with a Ge spectrometer /7/, viewing the corners of the assemblies through a collimator slit. The measurement geometry of the detector was kept constant during each campaign.

Each corner was scanned over the full fuel length and the arithmetic mean of all the four scans was used as a measure of the burnup. By averaging over the four corners, effects of burnup gradients as well as possible bending of the assemblies will be canceled to first order.

For normalization between the two facilities, a Cs-137 reference source was used /4,8/. The source consists of a pellet of about 1 Ci of Cs-137 which can be mounted in the same position as the fuel assemblies. It can be oscillated vertically with a constant speed and a maximum amplitude of about 10 cm. As seen by the detector, the reference source thus represents a vertically extended line source.

The time averaged rate, R(ref), of the cesium peak in the detector when measuring the reference source may be written:

$$R(\mathrm{ref}) = \eta T \Omega \epsilon \qquad (1)$$

where η is the number of quanta per second and unit solid angle from the surface of the source towards the detector. T is the transmission of the radiation through the absorbing media (mainly water) between the fuel assembly and the collimator. Ω is the solid angle covered by the part of the detector that can be seen through the collimator by the source. ϵ is the intrinsic efficiency of the detector for the 662 keV radiation.

A corresponding expression can be written for the count rate R(fuel), when measuring a fuel assembly. In this case, however, R and η are average values for the whole fuel assembly. Furthermore, the simple product of eq. 1 is replaced by an integral over the projected area of the fuel assembly that contributes to the radiation hitting the detector at a given moment of time. In principle, the three quantities, η, T and Ω, then vary over the radiation area. However, with an essentially one dimensional slit, it can be shown that the double integral that should replace eq. 1 can be approximated with an expression similar to eq. 1 with sufficient accuracy /9/.

The factors η, T and Ω then stand for values averaged over the radiating area. In comparing fuel assemblies of identical mechanical dimensions and composition, it is assumed that η is proportional to the burnup of the assembly. An analysis /9/ of the double ratio:

$$K = \frac{R_1(\mathrm{fuel})}{R_1(\mathrm{ref})} \Big/ \frac{R_2(\mathrm{fuel})}{R_2(\mathrm{ref})} \qquad (2)$$

where the indices denote measurements made at different facilities, shows that this can be simplified to :

$$K = \frac{\eta_1(\mathrm{fuel})}{\eta_2(\mathrm{fuel})} \qquad (3)$$

Necessary conditions for this are:

(1) The positioning of the reference source relative to the fuel assembly must be identical in the two measurements. Since the reference source is confined in a standard ABB-Atom fuel channel and is mounted in a holder in the gamma wagon, this is true within an uncertainty of ± 1 mm at both facilities. This uncertainty corresponds to an error of less than 1 % in the gamma intensities.

(2) The acceptance solid angle Ω of the collimator-detector system for a point source can be different for the two measurements (different slit heights) but must not vary with the position of the point source along a line perpendicular to the collimator axis in the horizontal symmetry plane of the collimator.

2.4 HRGS Measurements

At the CLAB, the gamma spectrometer was set up in a standard way /1,4,7/. A weak Zn-65 source emitting 1115 kev radiation was attached to the detector in order to provide a check on the stability of the measuring conditions.

The collimator slit height was 5 mm. The speed of the gamma wagon was adjusted and checked to be constant within 1 %. The angular position of the assembly was adjusted with an precision of about 3 degrees.

The total scanning time per corner was about 200 sec. During the scan, 70 separate spectra were recorded, each representing a fuel length of about 6 cm.

The reference source was measured several times at CLAB. Each time the source was oscillated 20 times with an amplitude of ± 30 mm. The positioning of the source was measured to contribute less than 1 % to the error.

The measurements at the TVO were performed in a similar way as at the CLAB. The same spectrometer was used with identical settings of electronics. The main difference was the slit height that was 3 mm high at TVO.

Each partial spectrum at the TVO was measured for 5.5 seconds while it was 3 seconds at the CLAB. The longer measurement time was motivated by the difference in the slit height. In this way approximately the same statistical precision was obtained during both campaigns.

The calibration source was measured also at the TVO in the same way as described above. No cesium background was observed during either of the two campaigns.

2.5 Other Measures

The seals used were verified later to guarantee the quality of the calibration information. Supporting data for neutron measurements was achieved by analyzing the water samples taken from the pools where measurements were performed. A small difference in the amount of neutron absorbers (boron) was detected in these samples analyzed in the IAEA. It is understood that the neutron calibration source takes these changes properly into account.

The gauge of the fuel handling machine of each of the two facilities was used to measure the consistency of the gross weight of the assemblies measured. This served as an extra check against errors caused e.g. by the measuring of assemblies with or without the fuel channel.

3. Measured Assemblies

Table I lists those assemblies selected for measurement. All assemblies, except the 1405, had a fuel channel.

The BWR reference assembly is made by ABB-Atom, Sweden, and it is of the 8x8-1 design. It has an initial U-235 enrichment of 2.748 %. This assembly has been irradiated during four successive cycles between June 1980 and June 1984. It is stored with a standard fuel channel. Table I lists also the average neutron and gamma count rates corrected for cooling time. The values for the TVO assemblies have been normalized to the CLAB data.

Table I. Characteristics of the BWR assemblies measured. Assemblies nos 1-15 are from CLAB and nos 16-18 from TVO. R = reference assembly.

No.	ID	IE (%/ U-235)	BU (MWd/ kgU)	CT (days)	Corrected Cs-137 CR (cps)	Corrected Cm-244 CR (cps)
1	5711	2.745	34.89	1879	1914 ± 38	777 ± 16
2	5769	2.756	32.62	1176	1672 33	553 11
3	1196	2.644	34.17	1880	-	745 15
4	4922	2.683	32.24	1880	1672 33	539 11
5	4921	2.685	32.24	1880	1653 33	539 11
6	1179	2.642	33.68	1880	-	661 13
7	5808	2.754	35.99	1177	1903 38	907 18
8	5762	2.746	33.19	1881	1774 35	643 13
9	5717	2.747	34.59	1881	-	758 15
10	4855	2.788	23.89	2766	1274 25	158 3
11	4854	2.789	24.82	2766	1294 26	155 3
12	4857	2.788	24.16	2262	-	120 2
13	5721	2.750	34.75	1883	-	764 15
14	4858	2.787	25.44	2767	-	208 4
15	1405	2.210	19.83	3020	1035 21	-
16 R	6782	2.748	31.28	2340	1602 30	485 10
17	4538	1.941	18.03	2340	901 18	-
18	7501	2.817	33.06	1980	1662 33	-

4. Results

4.1 PNA calibration

Figure 1 shows the results of the PNA measurements. The corrected fast neutron rates, detected by Cd-wrapped detectors, are shown versus declared burnup. Similar curves were established also for the thermal flux detected by bare fission chambers. The calibration curve

$$CR = a * BU^{b} \qquad (4)$$

has been fitted to the CLAB data using the Deming code. The fitting parameters have the following values, a = (7.1 ± 3.5) * 10^{-5} and b = (4.57 ± 0.14). The 95 % Miller confidence intervals are also shown /6/. As can be seen, the measured data seem to follow the exponential law very well. The difference between the declared burnup of the reference assembly 6782 of the TVO, (31.280 ± 0.78) MWd/kgU and that interpolated from the calibration curve using the Deming code, (31.40 ± 0.63) MWd/kgU, is - 0.4 %. The difference obtained with the bare fission chambers is even smaller.

4.2 HRGS calibration

Figure 2 shows the results of the gamma calibration. Corrected and normalized average Cs-137 intensities with the total error for all measured assemblies (Table I) are plotted versus declared burnup. The calibration line

$$CR = a * BU \qquad (5)$$

was fitted to the data set of the CLAB. The fitting parameter was calculated using the Deming code to be a = (52.5 ± 0.4). From this figure it is obvious that the Cs-137 activity has a linear dependence, within the error bars, on the burnup. The difference between the declared burnup of the reference assembly 6782, (31.28 ± 0.78) MWd/kgU, and the burnup interpolated from the calibration line using the Deming code, (30.49 ± 1.34) MWd/kgU, is 2.6 %.

The Zn-65 activity was stable within the statistical errors during the measurements.

Figure 2 also shows the three assemblies measured at the TVO. For the reference assembly 6782, the measured value of the burnup agrees within the errors with the declared value. For the other two a slightly larger deviation is observed. A separate study after the completion of the measurements showed that this deviation is due to small irregularities in the transmission function of the TVO KPA-STORE collimator.

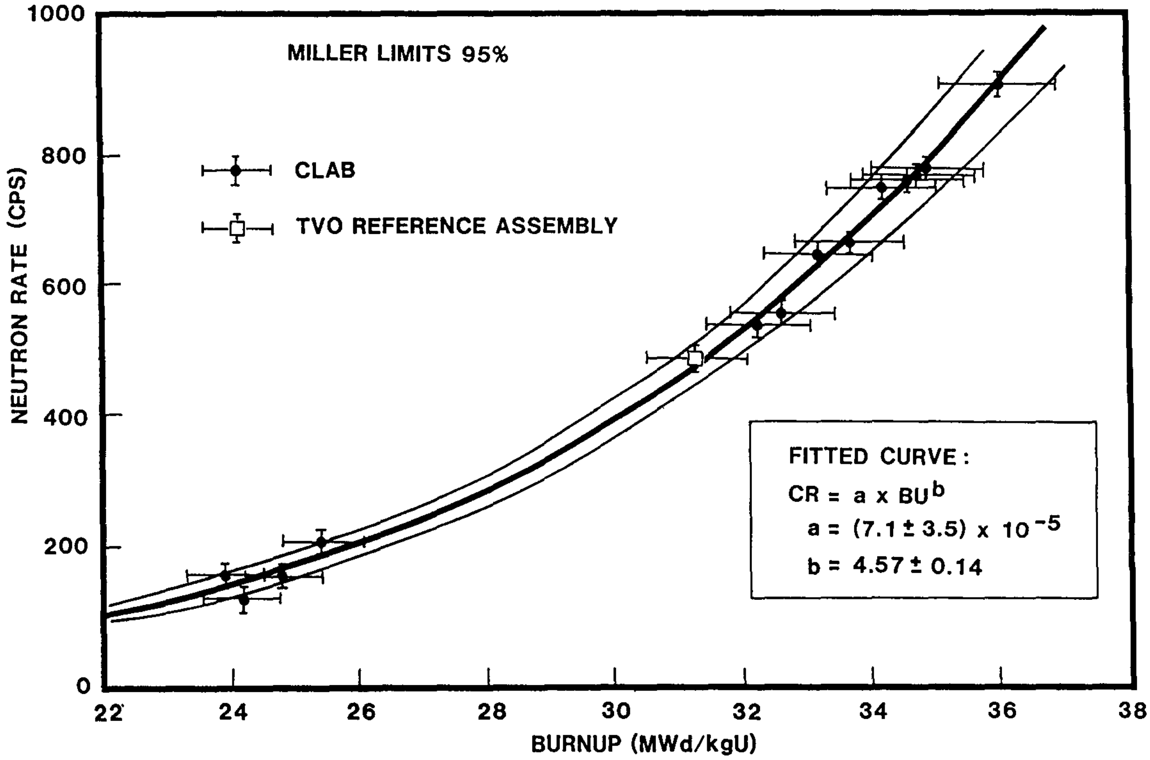

Figure 1. Corrected neutron rate of the 14 CLAB measured assemblies used for the calibration curve. The count rate of the reference assembly is plotted versus its declared burnup.

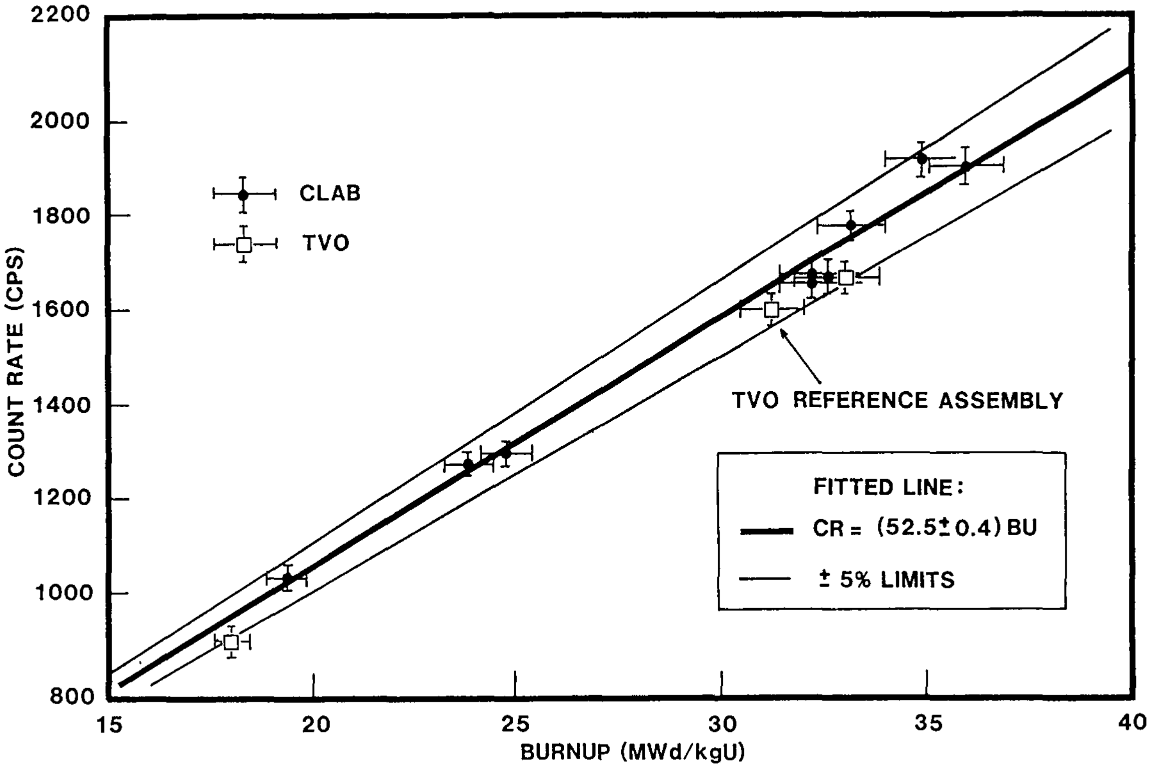

Figure 2. Cs-137 count rates of the 9 CLAB measured assemblies used for the calibration line. Normalized count rates of the TVO measured assemblies are also plotted versus declared burnup.

5. Conclusions

The PNA, used as the main method of calibration, made use of the 21 repeated calibration source measurements at the CLAB, and 14 at the TVO, respectively. After decay correction of the Cf-252, the reproducibility of the Cf-252 measurements at the two facilities using the same detection system is better than 1%.

Taking into account all the complementary measures, the neutron measurements are considered to be very reliable. The high degree of consistency between the PNA data of the reference assembly at the TVO and those measured at the CLAB, is understood to be based partly on the way the measurements were performed. First, an average of 12 points of each assembly in the plateau burnup region was used. These measurements were also used to check that the burnup profiles were normal. Measurements at six different vertical positions smooth out the effect of local burnup changes which have a strong influence to the detected neutron rate.

Because the initial enrichments were selected to match with each other, cooling times were selected long enough (for Cm-242 to decay) and the burnups were high enough, the influence of the irradiation history to the measured neutron rates was minimized.

Based on the PNA measurements it is concluded that the consistency between the burnup of the TVO reference assembly 6782 and the burnups of the CLAB measured assemblies is better than 2.5 %, which is the accuracy of the declared mean burnup.

It is known from earlier HRGS measurements with this type of fuel that the operator declared burnup data are of high quality. Internal consistency of the burnup of the order of 1 % has been detected /2/. These measurements show, however, that a consistency of the order of the accuracy of the declared mean burnup exists also between two countries.

The HRGS measurements support the results obtained with the PNA within the combined errors of the measurements and the declared burnup values.

6. Acknowledgements

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A MULTIFONCTION RADIOACTIVE WASTE MONITORING SYSTEM

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Abstract

The monitoring of unknown radioactive transuranic wastes mixed with fission products (FP) needs several measuring technics : passive and active neutron methods, gamma rays spectrometry and, sometimes, emission tomography to localize the "hot points" in the waste packages.

The goal is to achieve a whole system from the most updated electronics sub-assemblies to provide these characterizaton measurement at the lowest cost and in the simplest manner.

The control of the different measurements is made by only one computer and an unusual way of using the gamma spectrometry A.D.C. and multichannal analyser makes possible to control the neutron analogic electronics : neutron counter high-voltage supplies, amplifiers and discriminators; many of the gamma spectrometry sub-assemblies are also used for the gamma emission tomography. The different measurements are automated and different programms offer the possibility to choice the proper measurement methods for each item.

The waste package handling apparatus is not included in the system but the control of such handling might be perfored by the micro-computer. We describe the main parts and features of the system.

1. Introduction

The multifunction station described below consists of high performance equipment and combines different non-destructive methods of monitoring radioactive wastes (fig.1).

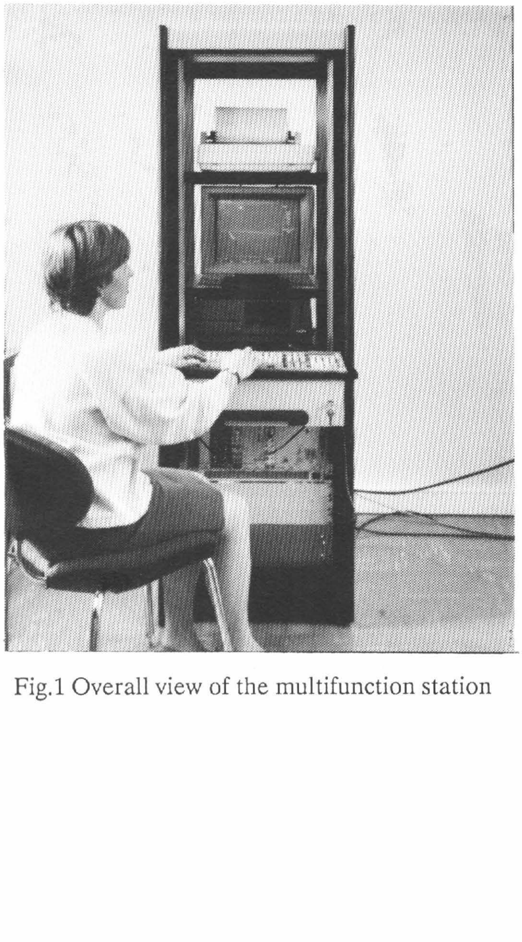

Fig.1 Overall view of the multifunction station

1.1 Measurement and Identification

- of the mass of plutonium by counting the number of neutrons emitted with spontaneous fission of the even isotopes of plutonium,
- of the mass of fissile isotopes (^{235}U and ^{239}Pu) by the active neutronic method, using a 14 MeV neutron generator,
- the isotopic composition of plutonium by gamma spectrometry,
- the fission products by gamma spectrometry.

With the additional capabilities, by studying multiplicity, of :

- reducing cosmic ray backgound noise,
- detecting the possible presence of curium and californium.

1.2 Why a multifunction station?

The combining on a single platform of a maximum number of electronic subsystems provides the user with the necessary hardware to run different processing programs. It also makes it possible to:

- considerably reduce the cost of the electronics,
- centralize control of the different functions,
- simplify checking and adjustment of the analog processing channels,
- more easily automate the handling of the containers and execution of the measurement cycle.

2. Description of the Measuring Station

2.1 Measuring device

This consists of :

- a chamber enabling thermalization and detection of neutrons by an array of six sets of three proportional helium-3 counters,
- preamplifiers and discriminators (one module per detector set), with integral gamma neutron threshold adjustment (SADN),
- a power amplifier (SADP) which transmits, at low impedance, pulses to the processing modules installed in the two NIM racks.

To simplify and limit the number of leads, thus considerably reducing the cost of installation, the SADN components are interconnected for the transmission of neutrons pulses and high and low voltages. These modules were designed by CEA-LETI/DEIN and are marketed by the company Novelec (Figure 2).

2.2 Passive neutronic measurement

When the neutronic activity in the container is sufficient, the mass of plutonium can be determined by measuring the number of spontaneous fissions of the even isotopes of plutonium (^{238}Pu, ^{240}Pu and ^{242}Pu). The equipment with which this measurement is made consists of a counting and signal processing system which establishes temporal correlations between the neutrons detected amid background noise due to the presence of (α, n) neutrons

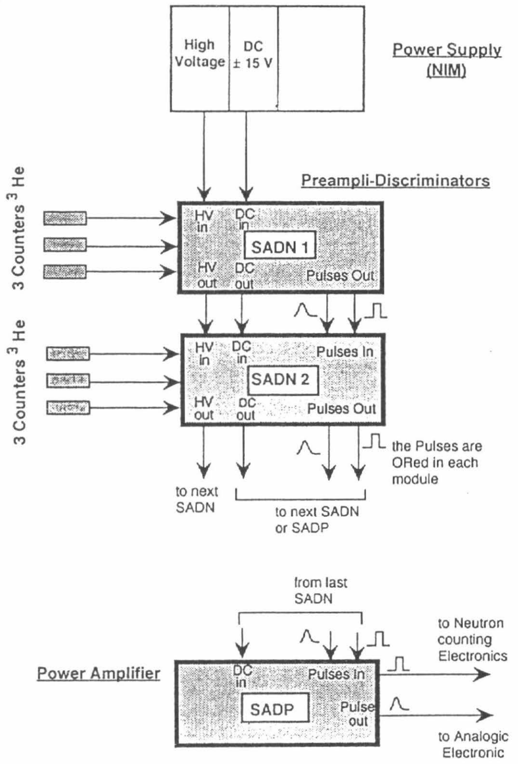

Fig. 2 Passive and Active Neutron Counting Head Electronics (Simplified scheme)

and allowing to correct the multiplication effects of the nuclear material examined. Two measurement techniques are used depending on the count rate :

- a shift register coincidence type counting module (SCRD) is used for count rates exceeding 5000 per second, corresponding to a quantity of plutonium between a few grams and a few kilogrammes. The operating principle consists in accumulating in two separate counters the neutrons presence in two shift registers, one of which is triggered on detection of a neutron (presence of coincidences) and the other after a long delay relative to the neutrons originating from the same fission). The difference in the counts between the two channels, which is equal to the number of true coincidences, is proportional to the number of spontaneous fissions, and hence to the quantity of plutonium present /1/. The coincidence windows, which have a maximum size of 128 μs, are shift registers activated by a clock operating at 4 MHz. The pre-delay, which is adjustable up to 8 μs, ensures proper operation of the system with regard to the dead time of the measuring analogics.

- a counting module of the STMV type for small quantities of plutonium, between around 0.1 and 20 grammes, using the dead time correction relationship, whose properties, in the case where the distribution of the pulses is not Poissonian, make it possible to deduce the mass of plutonium present in the container measured /2/. The duration of the dead time window can vary up to 300 μs.

- a module of the SDMN type, which decodes the neutronic multiplicity in the temporal windows to the ninth order /3/, makes it possible to :

  - correct for the influence of multiplication effects on the calculation of the mass of plutonium,
  - reduce the background noise due to cosmic rays,
  - detect the possible presence of any curium and californium.

The modules intercommunicate via a parallel bus specific to the data exchanged and also linked to the central computer by a serial interface module (ISCN2). Acquisition of the data is controlled automatically by the central computer running a measurement program (CEA-LETI/DEIN), or by the operator from the keyboard. Similarly, a number of data-processing algorithms can be chosen and implemented on the computer. An analog multiplexer makes it possible to benefit from an analog-digital encoder (necessary for gamma spectrometry), to check the proper operation of the neutronic acquisition analog channel and to accurately adjust the neutron/gamma discrimination threshold.

2.3 Active neutronic measurement

The "fast neutron" method, using a 14 MeV neutron generator to bombard the object examined, is used when the activity of the container is low. This can be used to measure the masses of fissile isotopes under effect of thermal neutrons, i.e. mainly ^{235}U and ^{239}Pu.

Fast and thermal neutron flux decay is recorded with a many inputs time-based encoder connected to the computer via a digital multiplexer, which make it possible to plot curves for these neutron fluxes (monitor signal and signal representing induced fissions) as a function of the time elapsed since the last burst of neutrons from the generator.

As in the case of passive neutronic measurement :

- acquisition and processing of the data is controlled automatically by a special program in the computer memory or by the operator from the keyboard,
- there are means of checking the proper operation of the analog neutron data acquisition channels and adjusting the neutron/gamma discrimination thresholds.

2.4 Gamma Energy Spectrometry

Either one or two complete analog channels can be connected to the computer via the analog selector. Two spectrum collation programs managed automatically or by the operator make it possible to :

- measure the quantities of fission products contained in the waste using special software marketed by Canberra,
- determine the isotopic composition of the plutonium with a program developed by CEA-DTA/DAMRI.

2.5 Checking a proper operation

The credibility of the readings obtained depends on rigourous checking of the detection channels in service. The power supplies from the NIM racks comprising gamma and neutron instrumentation modules are checked systematically at the start and end of each measurement cycle. The absence of any of the voltages cancels the measurement cycle and a message is output on the printer to warn the operator.

The following operations are also carried out :

- automatic calibration of the measurement channel using a reference gamma source,
- verification of the channel energy resolution,
- automatic transfer of the calibration parameters to the fission products identification program. This can then re-calibrate itself automatically in the event of slow drift in analog channel. The possibility offered by the Canberra software of concatenating control procedures makes it

possible to minimize operator intervention in gamma measurement. This program which checks proper operation was developed by CEA-LETI/DEIN.

3. The programs

3.1 Options

The multifunction station has no built-in neutronic measurement channel processing software, software is selected and stored by the user. Different combinations and sequences of measurement and processing programs can be obtained and the final configuration will depend on what is chosen. For example, the following are possible :

1) gamma spectrometry
2) gamma spectrometry and passive measurement
3) gamma spectrometry and active measurement
4) gamma spectrometry and active and passive measurement
5) passive measurement
6) passive and active measurement
7) active measurement

8) passive neutronic measurement channel adjustment
9) active neutronic measurement channel adjustment
10) gamma spectrometry channel adjustment

3.2 Automation of package handling and measurement process

After the methods of measurement have been selected by the operator or by instructions resident in the central computer memory, the latter manages the measurement process via a SMC 600-slave PLC essentially concerned with automation of handling and surveillance of the safety devices.

The first seven programs can be invoked by an operator or automatically; they require the actual presence of a package and only truly begin after authorization by the SMC after it has checked proper operation of the installation and that the initial conditions of functioning are fulfilled: installation in safety position, presence of a waste container etc. These programs are terminated after the installation has been made safe to the exterior by an "end of program run" signal.

Programs 8, 9 and 10 are controlled by the operator, they can only be run if the installation is in the initial safety position and if the neutron generator is shut down. There is no provision for automatic container handling in these last programs, but all control functions are available to the operator.

The acquisition parameters can only be modified if it is the operator who invokes a program.

At the present time, the settings of the neutron generator (maximum output, duration of bursts, frequency of repetition etc.) cannot be changed by the central computer. It is only start up and shut down which are controlled by the slave SMC, to ensure maximum safety. Management of drum movement is not presently provided, and may be introduced using the SMC PLC connected to the computer, depending on user requirements.

4. General information on interconnection of the different multifunction station modules

The diagram in Figure 3 shows the general arrangement of the different parts of the measurement station. The task of the central computer is to manage the acquisition of data arriving from the three measurement channels and to process the results; it also handles dialogue with :

- the operator, by means of the keyboard, screen and printer,

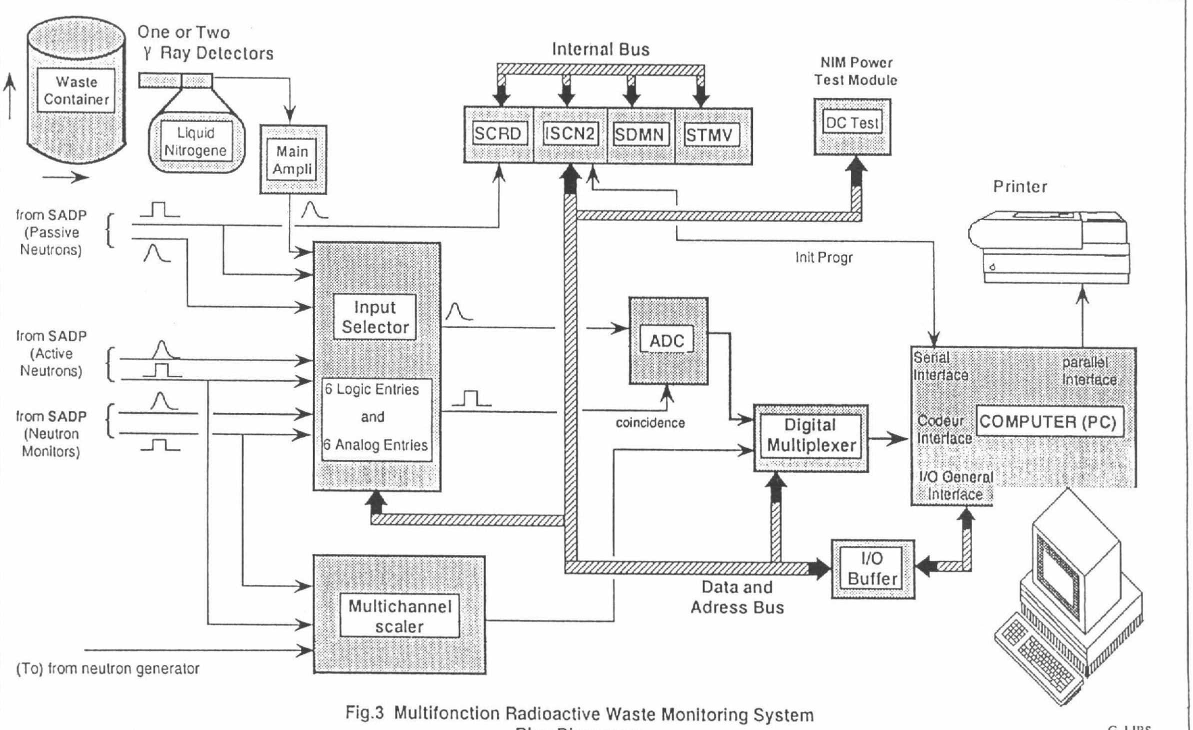

Fig.3 Multifonction Radioactive Waste Monitoring System
Bloc Diagramm

G. LIBS
DEIN/SAI

- an SMC 600 slave PLC, which handles control and monitoring of the safety devices, the handling mechanisms and the neutron generator. The link with the computer is via a serial interface in the computer,

- possibly a second computer from which it receives commands and to which it supplies reports on execution of its tasks.

The three measurement channels (passive and active neutron measurement and gamma spectrometry) form independent units with all their parts grouped in two NIM racks. The SCRD, SDMN and STMV modules process the pulses originating from the passive channel and are interconnected by a special data bus. All these parts are controlled from the central computer via the ISCN2 interface serial port.

The active neutron channel includes a 8000 channel multi-scale module, data from which is fed by a digital multiplexer to the computer encoder interface card.

The gamma spectrometry channel consists of a high resolution preamplifier, an amplifier with conventional shaping and an amplitude encoder (ADC). The data from the latter are fed by the digital multiplexer to the encoder interface card installed in the computer.

One of the main advantages of this arrangement is that it makes it possible to benefit from the gamma acquisition chain for displaying the energy spectrum of the neutrons from the neutron channels to accurately adjust the preamplifier neutron/gamma discrimination threshold. This arrangement is obtained by the multi-input analog channel selector, development work by CEA-LETI/DEIN on which mainly related to preserving high linearity in the measurement channel.

5. Conclusion

Checking of unknown radioactive waste containing transuranic elements mixed with fission products necessitates different measurement techniques: active and passive neutronic methods, gamma spectrometry and, on occasions, emission tomography to localize "hot spots".

Our intention is to propose a complete system using the latest electronic techniques to obtain a cheaper and simpler measurement station.

The different measurements are checked by a single computer and, as is not normally the case, the components of the gamma channel (ADC and multi-channel analyzer) are used to check the neutron measurement analog electronics: adjustment of the neutron counter high-voltage supply and the amplifier/discriminator thresholds. Some of the modules of the spectrometry chain will also be used for gamma emission tomography. The different measurements are automatically concatenated and many programs offer the possibility of selecting the methods of measurement appropriate for a particular application from a range of options.

The container handling device is not included in the system but handling can be controlled from the microcomputer.

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A REVIEW OF PASSIVE NEUTRON COINCIDENCE MEASUREMENTS IN THE EURATOM SAFEGUARDS DIRECTORATE

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Abstract

The different passive neutron coincidence measurement techniques used by the Euratom Safeguards Directorate are reviewed. The inventory of non-destructive assay (NDA) instrumentation is presented, as are their different material and instrument specific calibrations. The principle of unique material specific calibrations for a given instrument type is described. A series of assay results using one specific High Level Neutron Coincidence Counter (HLNC-II) is shown and discussed in detail. A systematic analysis of the different sources of error in performing passive neutron coincidence measurements is presented.

1. Introduction

Passive neutron coincidence counters are appropriate for the determination of plutonium in many types of samples. They are relatively simple devices which have been developed into reliable portable instruments. As a result, a large fraction of NDA measurements made by the Euratom Safeguards Directorate (DCS) used examples of these counters. Over a period of several years, the use of DCS instruments has produced a large body of passive neutron data. This paper describes the DCS collection of passive neutron NDA systems and their calibration for different types of material and the performance which is achieved. It then goes on to discuss the sources of uncertainty on the measurements and summarises the implications of moving to unattended monitoring systems.

2. NDA Equipment Inventory

A list of passive neutron measuring equipment currently operated by DCS is shown in table 1. All the equipment is fitted with Amptek amplifier/discriminators and is used with Jomar JSR-11 shift register electronics.

These detectors are well-known and no attempt is made here to describe them. References are given in table 1. All this equipment is transportable and can be used at numerous different installations. Despite this, good reliability of the systems has been achieved.

Detector	Number in use	Measurement Campaigns per year	Typical Items	Reference
HLNC	11	131	PuO2 powder Pu metal MOX powder MOX solids	1
NCC	7	35	MOX LWR fuel assembly Boxes of fuel pins MOX FBR fuel assembly	2
UFBR	4	52	MOX FBR fuel assembly PuO2 powder	3
INVS	4	46	PuO2 powder MOX powder MOX solids	4
Cerceuil	4	46	MOX fuel rods	5
Ion-fork (mod)	1	-	MOX assemblies (under water)	6

Table 1 Euratom passive neutron measuring equipment

3. Calibration

The calibration of each detector type for a certain material is based on a single calibration curve for one particular detector. The curve is established by measurements of known amounts of material. The difference between the individual models of a particular type of detector is determined by measuring a particular neutron source in each one. The neutron source measurements allow the establishment of a cross-calibration constant for each detector which is used to correct the acquired data to make the single calibration curve applicable in each case. This method is described in reference 7. The method is used both because it provides a logical structure for calibration data which is easy to maintain and also because the effort involved in calibrating a family of detectors is greatly reduced. The calibration of each detector is made using non-multiplication corrected data and multiplication corrected (ref 8) data. A quadratic equation is used to fit the non-multiplication corrected calibration curve whereas a linear fit is used for the multiplication corrected curve. The values of the calibration constants currently used are shown in table 2.

	rho zero	multiplication corrected		not corrected		
		a1*x + a2		b1*x*x +b2*x +b3		
UFBR: x=240Pueff:						
UFBR-II powder	.04992	3.8683	-511.9	1.39e-3	-.198	9.88e3
UFBR mod powder	.0591	4.7272	-32.32	5.78e-4	5.33	-5.77e2
UFBR FBR assembly	.05009	3.886	0	9.55e-4	5.963	0
NCC: x=240Pueff/cm:						
NCC(No Cd)LWR assy	.0814	647.94	0	64.6	219.6	0
NCC(No Cd)Pin boxes	.0814	673.81	-44.67	51.7	590.4	0
NCC(No Cd)FBR assy	.0814	627	0	9.95	853	0
NCC(Cd) FBR assy	.0814	572	0	9.65	574	0
HLNCC: x=240Pueff:						
HLNCC PuO2	.103	18.665	-0.17	-2.27e-1	23.71	1.87e-2
HLNCC Pu metal	.103	18.354	-0.17	-2.31e-2	24.08	1.45e-1
HLNCC MOX solid	.103	18.443	-0.17	-2.55e-1	26.47	3.81e-2
HLNCC MOX plates	.103	18.18	-0.17	-2.77e-1	28.76	5.50e-2
HLNCC MOX powders	.103	19.198	-0.18	1.68e-2	27.87	0.
INVS: x=240Pueff:						
INVS PuO2/MOX		-	-	0.983	58.24	0.
INVS Nitrate		-	-	4.193	59.27	0.

Table 2. Currently used passive neutron calibration constants

4. Performance

Figure 1 shows the operator-inspector differences for a range of several hundred measurements made with an HLNC2. The average operator-inspector difference is 2.2% compared to an average uncertainty on each measurement of 1.1%.

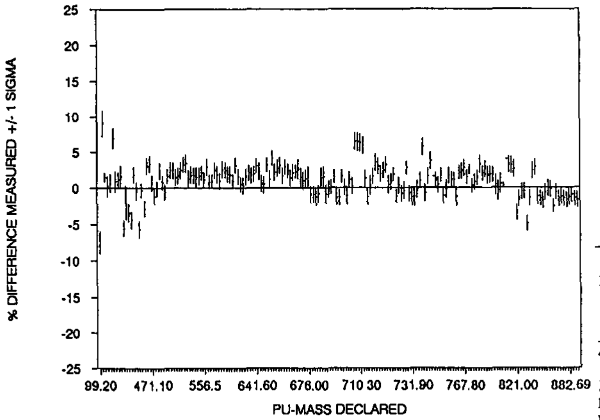

Figure 1 Operator Inspector Differences vs. mass

5. Sources of Uncertainty

Table 3 shows the sources of systematic error associated with passive neutron coincidence counter measurements. The values are estimates of the relative standard deviation in each case. The uncertainty due to counting statistics varies with the time of data acquisition, but can usually be made much less than 1%. The table has been compiled for the case of the plutonium mass obtained from the multiplication corrected calibration curve, for which the alpha value (the ratio of (alpha,n) neutrons to spontaneous fission neutrons) is required as an input parameter.

The sources of error have been divided into two types: those related to the use of the detector and those related to the properties of the sample. In the former category the errors are under the direct control of the user. Frequent normalisation and background checks, together with the use of the correct analysis parameters can reduce the error from all of these sources to very small values. There are three further possible sources of error in the second category which are also under the user's control: use of the appropriate calibration curve, correct isotopic values and correct reference date of the isotopic analysis. Errors from these sources should also be very small. The most important sources of error are related to the properties of the sample itself. One property which causes very large effects on the non-multiplication corrected results is the sample density. In the case of powders this can vary by considerable amounts (1.5 - 2.5 g/cm3) depending on how the sample has been treated. Fortunately the multiplication correction procedure reduces this effect to insignificant levels. The remaining sources of error determine, to a large extent, the performance of the measurement technique: (i) the presence of other materials (including impurities) (ii) the container and (iii) sample positioning. The magnitude of these errors depends on the extent to which the material can be classified into sets suitable for a specific calibration. Such sets would be, for example, MOX fuel assemblies

EFFECT	SIZE %	EFFECT ON Pu MASS %	IMPROVEMENT IN AUTO MODE?	COMMENTS
A. Detector Related				
Background	10%	1.5 - 7%	Yes	
Normalisation	5%	1%	Yes	
Malfunction	100%	100%	No	time delay
rho-zero	5%	6%	Yes	*
deadtime	10%	0.1%for 500g	Yes	*
gatelength	factor 2	10%	Yes	*
pre-delay	20%	5%	Yes	*
B. Sample Related				
Density	25%	small	Batch	
Wrong Calibration		10%	Yes	*
Wrong Isotopics			No	*
Wrong date	1 year	2 - 4 %	No	*
Other materials (including impurities)				
- other fissile		small	Batch	
- neutron poisons			Batch	
- (a,n) material			Batch	
- moderator		small	Batch	
Container			Batch	
Positioning			Yes	

* = user may be supplied with, or enter, incorrect analysis parameters

Batch = greater uniformity for samples within one batch

Table 3 Sources of uncertainty in coincidence counter measurements of a particular design, or well characterized powder samples. In these circumstances measurements show systematic uncertainties of 1 - 2%. In order to achieve this performance it is essential that the samples used for calibration are representative of the overall population of samples to be measured, including the effects of containers. These values are similar to those given by other authors (reference 9).

6. Example MOX Powder Samples

Some MOX sample measurements were re-analyzed in detail to investigate the sensitivity of the results to various parameters. In field measurements, the background, normalisation and isotopic composition (including date) are potential sources of error. These are considered in turn.

6.1 Background

Figure 2 shows the percentage change in the multiplication corrected reals as a function of the percentage change in background for three MOX samples with masses of 250, 540 and 900g. The change in the background rate (between 2000 and 20000 cps) was chosen because this range has been observed in measurements at a fixed location in a particular installation. The figure shows that, in this case, a relatively small error (10%) in background determination can lead to errors of 6%, 2%, and 1.3% for masses 250, 540, and 900g respectively.

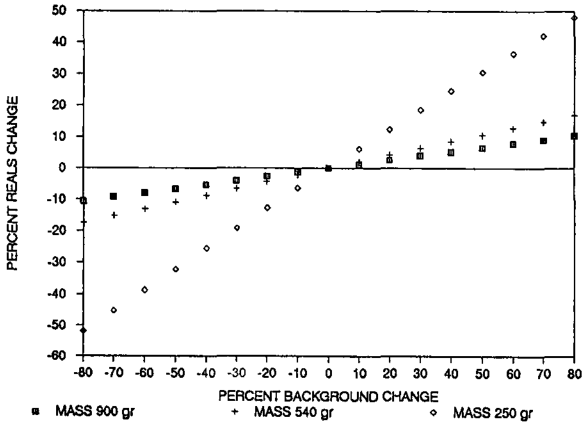

Figure 2 Rmc vs. Background

6.2 Normalisation

In general, experience shows that normalisation measurements are performed well. However it is possible in installations where several detectors are used that the reference source from the wrong detector is used. Very often this type of error is detected immediately because of the large change in the normalisation constant, but for small changes the new value may be accepted. Figure 3 shows the effect of a change in the normalisation constant on both the non-multiplication corrected and the multiplication corrected reals. The non-multiplication corrected reals change in direct proportion to the normalisation constant. The multiplication corrected reals are less sensitive, and change by about one third of the percentage change in the normalisation constant.

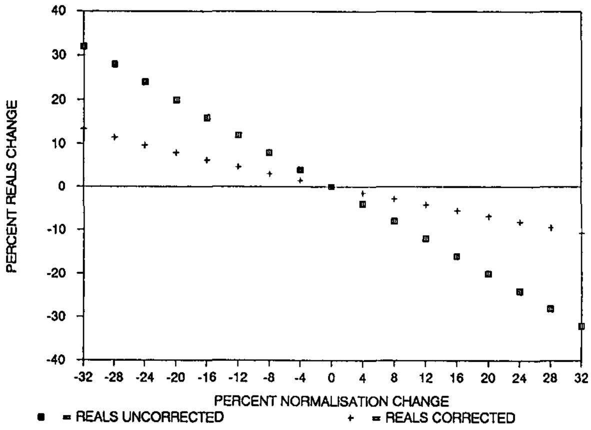

Figure 3 Reals vs Normalisation

6.3 Date of Isotopics

An incorrect reference date of isotopic analysis affects the multiplication corrected reals by affecting the updating of the isotopics before the calculation of the alpha value. In practice incorrect dates are an unusual occurrence, but the result of a variation of the date by up to five years is shown in figure 4. This gives an indication of the accuracy required in entering the information. An error of one year causes the multiplication corrected reals to change by about 2%.

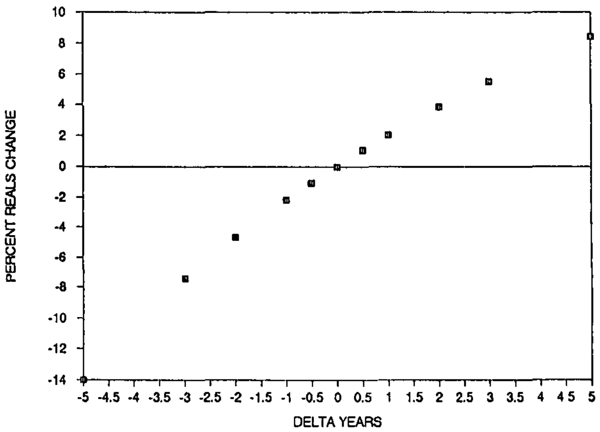

Figure 4 Rmc vs isotopic date

7. Automated Measurements

A new phase of safeguards instrumentation has recently begun with the installation into large facilities of fixed measuring equipment which can operate continuously in an unattended mode. This has lead to the need to develop specialised software in order to realise the full potential of these devices. In general, it is to be expected that the error associated with measurements carried out by such automated equipment would be smaller than that from manual equipment for a number of reasons. Firstly, with a fixed detector system the opportunity for damage during transport between installations is removed. Secondly, automated systems remove many of the user errors which can occur in normal systems, for example the choice of a wrong calibration curve. Thirdly, automated systems are usually applied to places in a plant where the type of material and the container used is very well determined. This means that the measurement system can be 'tuned' to give optimum performance for a particular sample type. In table 3 which shows the sources of error in passive neutron measurements, the column headed 'Improvement in Auto Mode' denotes this latter point as 'batch', which indicates that certain properties of the samples will be common to all the material measured with a particular detector and should also be common to the calibration. Thus the error contribution is removed. A good example would be that the container of all the samples is likely to be the same. In addition, product which is being produced by a certain plant is likely to have a very much smaller variation in, for example, impurity or moisture levels than material of different origin which has been stored for different periods of time. For the latter, any calibration will be much less representative of the complete ensemble of material.

However automated systems, unless they are carefully designed, have also the potential to make much worse measurements than manually operated ones, mainly because no user is present to correct faults as they arise. If a manually operated system gives an unexpected result, then it is possible to check the equipment and make a re-measurement while the sample is still available. In automated systems this option may not exist, therefore a number of features should be included in order to minimise errors due to malfunctions. Firstly, the use of duplicate electronic chains can give extra security of data acquisition. Tests on the acquired data such as the comparison of the calculated and measured accidental coincidence rate allow the state of the detector and electronics to be determined. Secondly, a permanently installed small neutron source allows the efficiency of the detector to be checked continually between measurements of samples. This not only enables a warning to be given of efficiency changes, but should also allow analysis of the data collected during the period of altered efficiency, excluding of course the case of complete failure.

Another test which can be useful in automated mode, is to compare the value of the Reals to Totals ratio with a reference value. In general, the R/T value for a multiplying sample is given by:

$$\frac{R}{T} = \frac{eF\,\overline{v(v-1)}}{2\,M\,(1+a)\,vs}$$

where

e = detection efficiency

F = fraction of coincidence in the shift register gate

$\overline{v(v-1)}$ = 2nd factorial moment including the effect of multiplication

M = multiplication

a = alpha (ratio of (alpha,n) neutrons to spontaneous fission neutrons

vs= mean number of neutrons per spontaneous fission.

This ratio contains all of the information on the effect of multiplication in the sample and is the basis of the well-known Ensslin multiplication correction technique (ref 8). This ratio depends on the mass, shape, density and alpha value of the sample. It does not contain any more information than is normally used in the complete analysis of the measurement, but the interest in the case of automated measurements arises because of two conditions: firstly because of the uniformity of material type referred to above, the R/T value depends in a simple way on the mass of the sample, and secondly the ratio is very simple and quick to calculate. Figure 5 shows a plot of the R/T value for some particular sets of data from MOX and PuO2 samples as a function of sample mass. It can be seen that the R/T value is relatively independent of mass over quite a wide range, but that differences exist between the two sets. This presents a potential check on the quality of the measurement (or a change in sample type) which could

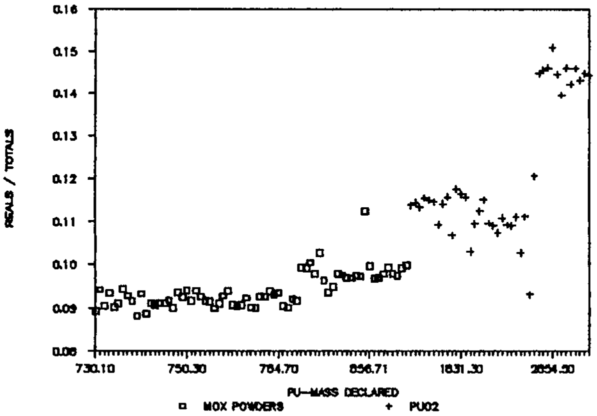

Figure 5 R/T vs mass for different materials

be carried out as data is collected by the automated system. The usefulness of this test will be very installation dependent and will require the compilation of suitable representative data.

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ASSAY OF PLUTONIUM BY THE USE OF PASSIVE ASSAY TECHNIQUES

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Abstract

The isotopic composition of Plutonium 242 can not be measured by gamma spectroscopy. For this reason the gamma and passive neutron assay technique has been historically combined with the isotopic correlation to obtain the Plutonium 242 weight percentage. In this report experimental data from high resolution gamma spectroscopy, passive neutron assay and calorimetry were used to determine the weight of Plutonium, the respective weight percentages and the Americium 241 content without resorting to correlations.

Data are presented for the plutonium oxide PERLA standards in the weight range of 100 g - 2500 g. The data were collected in the JRC-PERLA facility and the results are compared to nominal values. The accuracy is always better than 0.5% on the total Plutonium mass for the 14 samples in this evaluation.

1.Introduction

Historically the assay of a Plutonium sample has been done with the use of a High Level Neutron Coincidence Counter (HLNCC) together with a gamma ray isotopic determination. This technique may introduce errors depending on the method used to estimate the Plutonium 242 content.

The use of another measurement such as the thermal power of the Plutonium sample measured with a calorimeter gives another relation which links the unknown weights of the isotopes of plutonium and of the americium 241 to a measured quantity.

This report describes a method to evaluate the isotopic composition of Plutonium, in particular the isotope Plutonium 242, which gives a better estimation of the total plutonium mass, the isotopic ratios, and the Americium 241 fraction. The method uses the measurement results from the MGA gamma technique, the HLNCC and the calorimeter.

The programme for the evaluation of these quantities has been implemented with Mathematica of Wolfram Research. This programme is designed for general use. The instrument availability, time available for this activity, plant constraints and reliability of the sample data all affect the user needs. The user may chose one of the following possibilities:

I) If the isotopic composition of Plutonium and Americium content is well known a measurement with the HLNCC or calorimeter is enough to measure the mass of Plutonium;

II) If an isotopic correlation is used for Plutonium 242 determination, the gamma spectroscopy can be combined with either HLNCC or calorimetry;

III) If only the chemical form of the material is known the three techniques together permit the evaluation of the isotopic ratios, the Americium 241 fraction, and the mass of Plutonium.

2.Method

The gamma ray technique is used to measure the ratio between the isotopes of plutonium : Pu238/Pu239, Pu240/Pu239, Am241/Pu239 /5,6/.

The neutron technique (HLNCC) is used to measure a quantity "Real coincidence counts" corrected for background and for dead time, which is proportional to the Pu_{240} eff, which is the sum of the weights of the plutonium 238, 240, 242 each one multiplied by a known constant /1,2/.

The assay of a plutonium sample is impossible without another relation which links together the weights of the 242 isotope of plutonium and to the other quantities.

The calorimeter measures the thermal power of a plutonium sample. This quantity is proportional to the sum of the weights of the plutonium isotopes plus the weight of americium 241, each one multiplied by a known constant /3,4/. This measure can be used as another relation for the assay of the total weight of a plutonium sample.

The four linear equations of the gamma ray technique together with the two equations respectively of the neutron and heat power measurements gives six linear equations in the six unknown isotopes. These six linear equations can be solved easily in the six unknowns Pu_{238}, ..., Pu_{242} and Am_{241}. The variance matrix of the result is computed exactly as **A.VI.AT** where **VI** is the variance matrix of the input variables and A is the transformation matrix defining outputs as function of inputs.

3.Instrumentation

The measurements were carried out using :

- High Level Neutron Coincidence Counter (HLNCC) model 2 as designed by Los Alamos National Laboratory with the Amptek A-111 electronics chain and with a Jomar JSR-11 Shift Register.
- Gamma spectroscopy which uses a High Resolution Gamma Spectrometer System suitable for high counting rates, the MGA evaluation code, developed at LLNL (Lawrence Livermore National Laboratory).
- Water twin bridge calorimeter developed at Monsanto Research Corporation suitable for measuring heat power up to 45 W.

4.Results

The technique has been applied to 14 plutonium oxide samples with plutonium weight in the range 100-2500 grams. Three Burn-ups were represented; 71.0%, 61.0%, 58.1% of Pu_{239}. The percentage of Pu_{242} is 0.98%, 4.83%, and 5.64%, respectively.

The Fig.1 shows the percent error on the total Plutonium mass assayed with the technique of HLNCC+MGA, as used up to now, relative to the real mass. The average error of the absolute values is 1.75% with a standard deviation of 1.18%.

The Fig.2 shows the percent error on the total Plutonium mass assayed with the technique of CALORIMETER+MGA, relative to the real mass. The average error of the absolute values is 1.62% with a standard deviation of 1.12%.

The Fig.3 shows the percent error on the total Plutonium mass assayed with the technique of HLNCC+MGA+CALORIMETER, relative to the real mass. The average error of the absolute values is 0.206% with standard deviation of 0.139%.

The Fig.4,5 show the error on the Plutonium 242 abundance assayed with the techniques of HLNCC+MGA+CALORIMETER and HLNCC+MGA or CALORIMETER+MGA, relative to the real Plutonium mass, respectively. The average error of the absolute values is 0.23% with a standard deviation of 0.19% for the proposed technique and 1.47% with S.D. of 0.98% for the old techniques.

The techniques of HLNCC+MGA and CALORIMETER+MGA give good accuracy only when they measure Plutonium samples of low burn-up. This is due to the fact that the Plutonium 242 content is evaluated with an isotopic composition correlation which gives high errors for high and medium burn-up.

Our proposed technique gives an alternate estimation of the Plutonium 242 content. It has less bias between the different burnups.

5.Conclusions

The proposed technique gives an accurate determination of the Plutonium weight without prior knowledge of the Plutonium 242 content. The samples assayed give an average absolute error on the plutonium mass of 0.216% and a standard deviation of 0.139%. This is much better than what can be achieved with HLNCC+MGA (Mean = 1.75%, S.D. = 1.18%) or with CALORIMETER+MGA (Mean = 1.62%, S.D. = 1.12%).

Unfortunately the calorimeter needs a very long time to make measurements. When the available time is too short to perform the precise measurement, some time can be used to check each batch for the abundance of the Plutonium 242.

We will evaluate in the future the use of this method to get an estimation of the alpha factor (α-n reaction rate) or of the multiplication factor in the case of samples with known abundance of Plutonium 242.

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"MGA: A Gamma-ray Spectrum Analysis Code for Determining Plutonium Isotopic Abundances"
URCL-LR-103220 April 1990

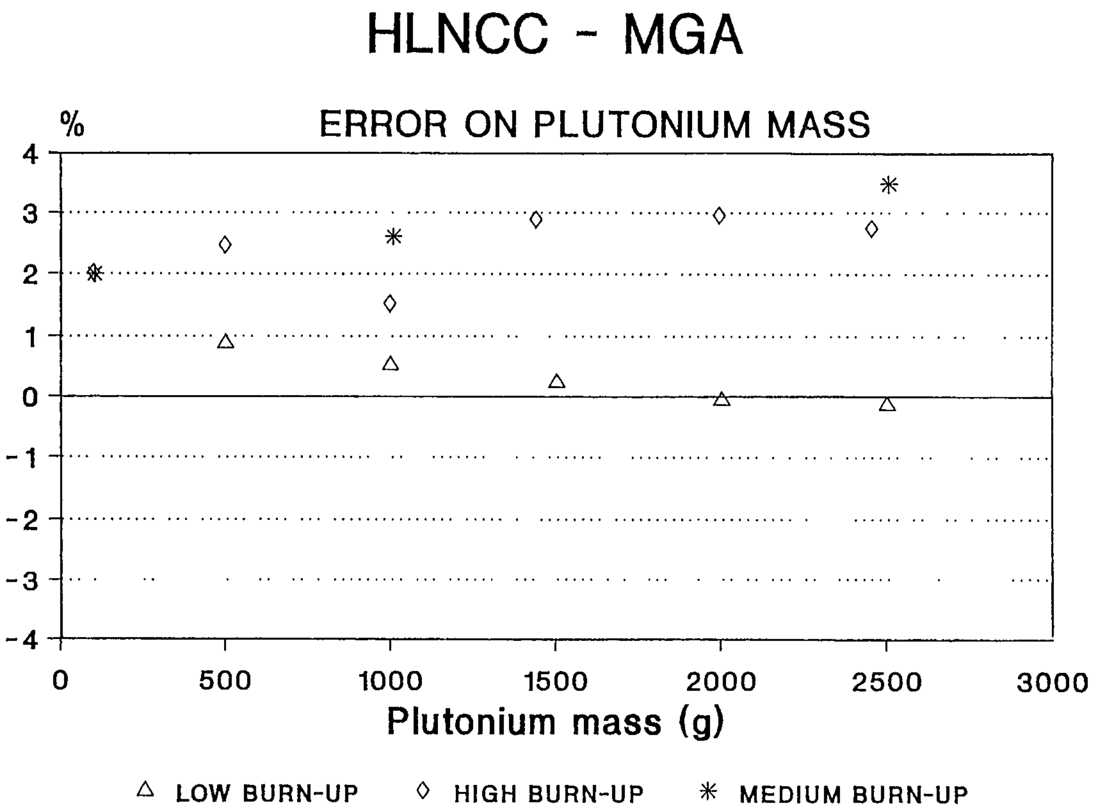

Figure 1 - Error on Plutonium mass : HLNCC-MGA

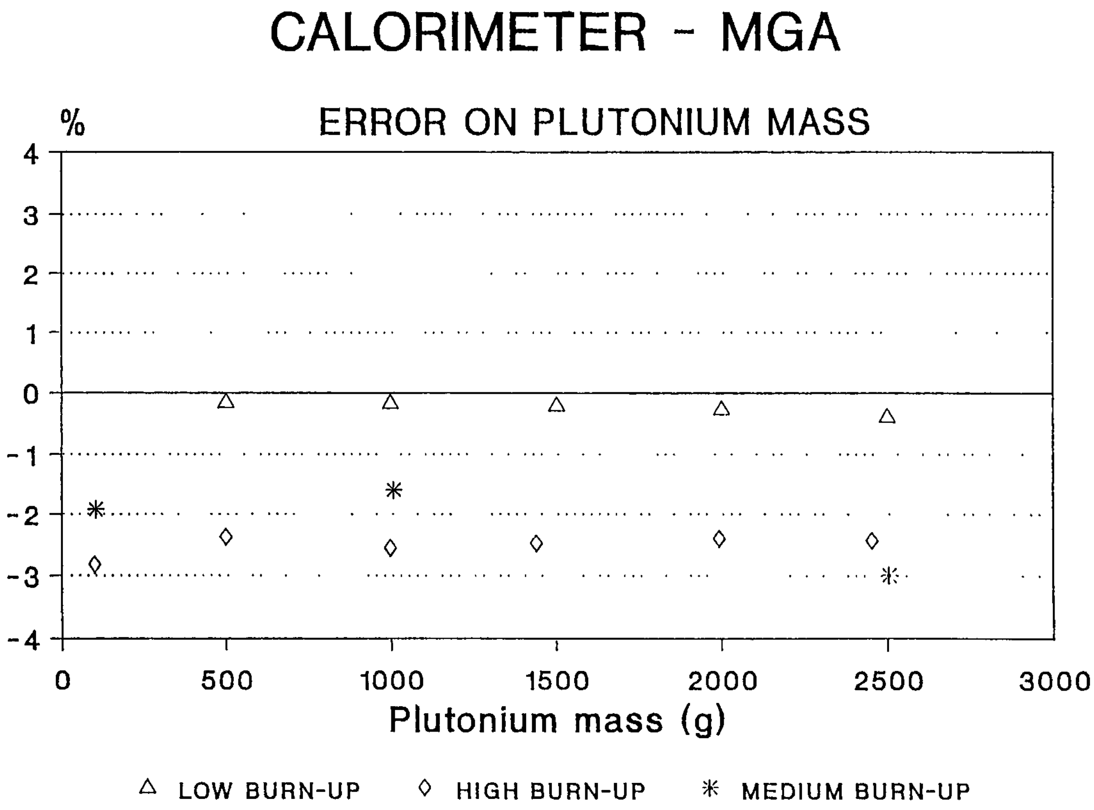

Figure 2 - Error on Plutonium mass : CALORIMETER-MGA

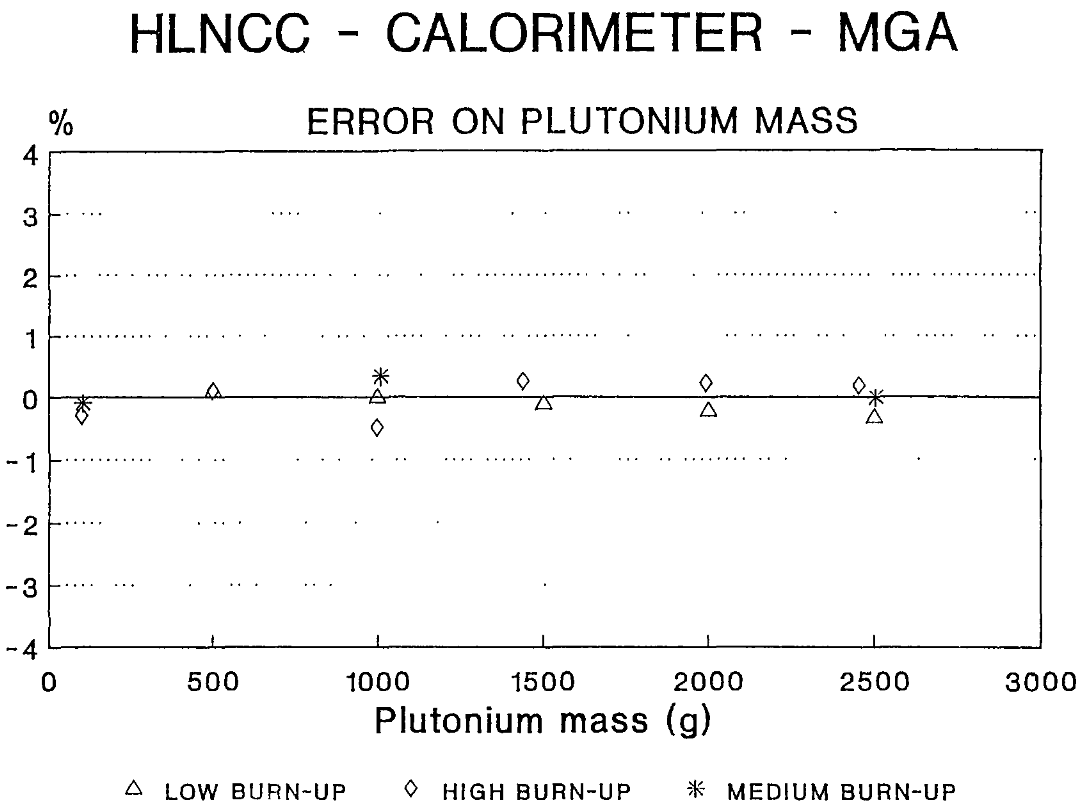

Figure 3 - Error on Plutonium mass : HLNCC-CALORIMETER-MGA

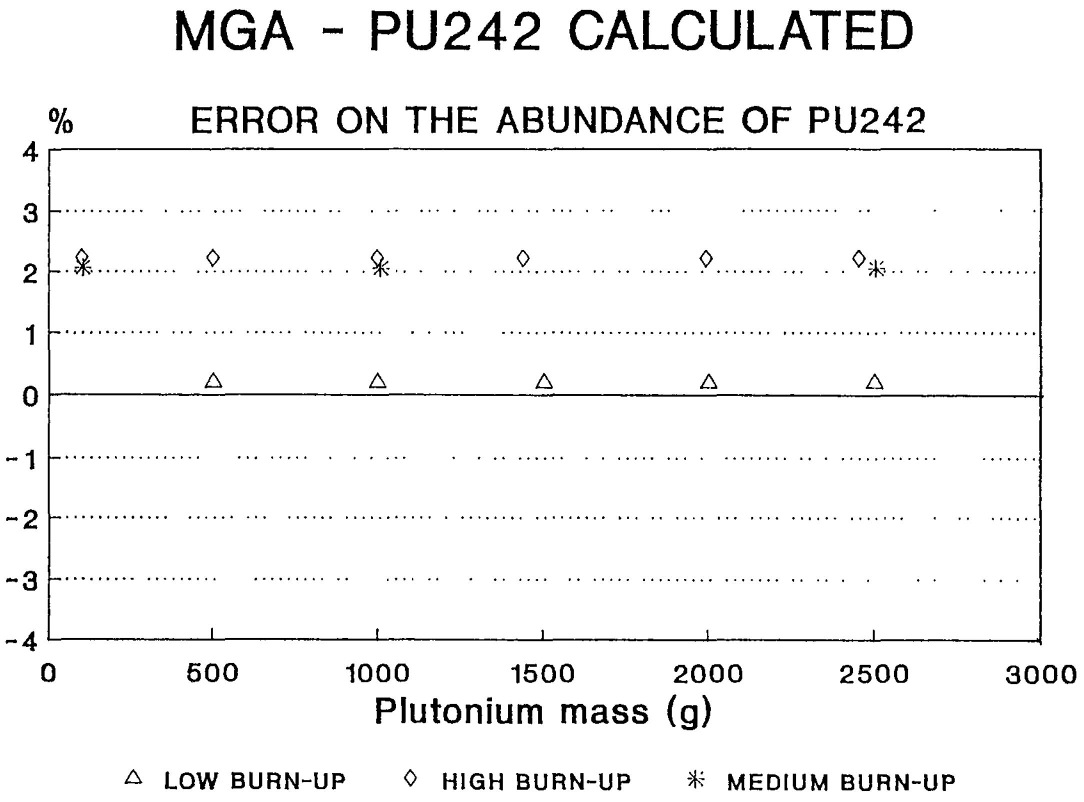

Figure 4 - Error on Plutonium 242 abundance : MGA-Pu_{242} calcul.

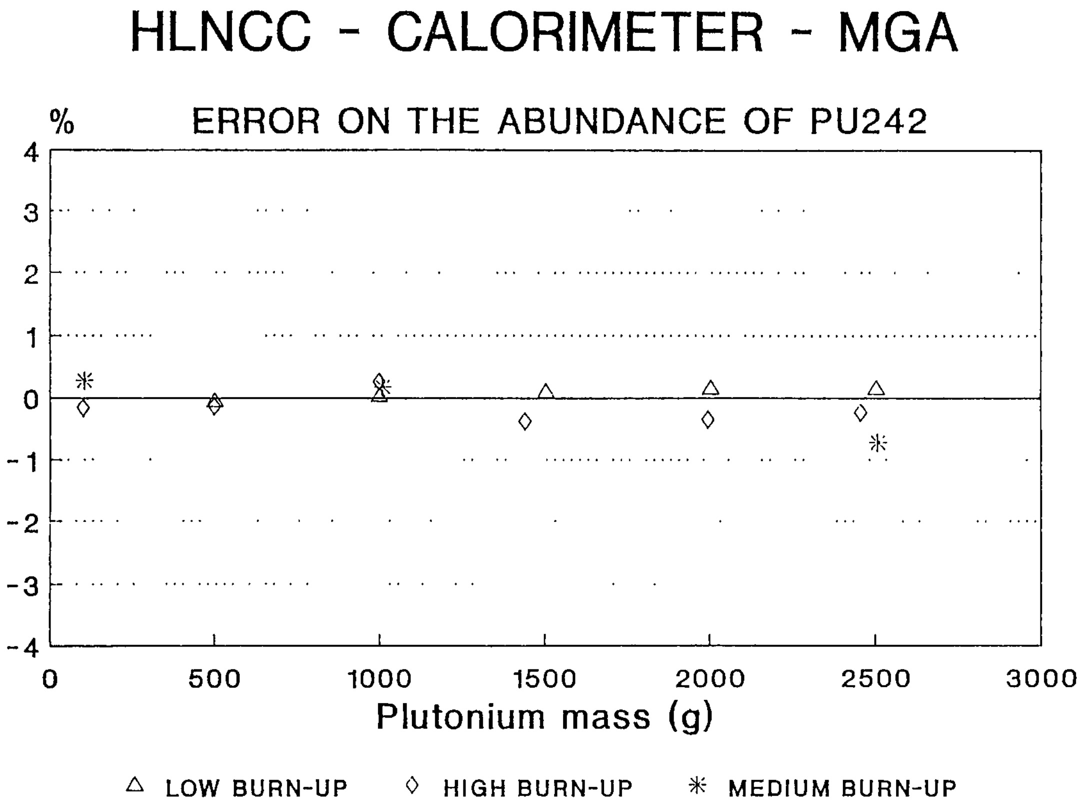

Figure 5 - Error on Plutonium 242 abundance : HLNCC-CALORIMETER-MGA

USE OF AN IMPROVED SIGNAL PROCESSING TO IDENTIFY A NEUTRON SOURCE FROM THE MEASURES OBTAINED WITH A SHIFT REGISTER SYSTEM

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Abstract

Once known the composition of a Pu, a way for verifying the content of a sample is the detection of multiplicities on neutronic coincidences obtained with a shift register system (SCRD).

Such a system provides two laws of coincidences probability for neutrons detected within two gates triggered independently and by neutron themselves.

After a brief presentation of the mathematical model of the detection system, the paper describes some experiences in which the SCRD system produced an assessment for fission neutrons and for (α,n) neutrons emitted by a source, without any other information except countings. For instance, this model shows how to separate a Pu_{240} source from a Cf_{252} source whereas a classical calculation, based on total counting and real coincidences, does not allow discrimination.

The calculation shows that usual statistics (10^8 cps) are sufficient to take the multiplicative effects into account.

La reconnaissance des sources neutroniques: une utilisation spécifique du S.C.R.D.

Introduction

Le système de coïncidences à registre à décalage (SCRD) associé à un ensemble de comptage neutronique donne accès d'une part aux comptages obtenus dans des fenêtres déclenchées aléatoirement et par les neutrons détectés eux-même (résultats A_B et $(R+A)_B$ fournis par l'appareil original de BOEHNEL[1]), et d'autre part aux comptages $A(I)$ et $(R+A)(I)$ dont la normalisation fournit les histogrammes de fréquence représentatifs des probabilités $a(I)$ et $(r+a)(I)$ des coïncidences d'ordre I dans chaque mode, pour I variant de 0 à 7. Chaque histogramme a donc 9 positions, la 9° étant occupée par le comptage cumulé de toutes les coïncidences d'ordre supérieur à 7. Après un bref rappel des types de mesures pour lesquels cet appareil est ou pourrait être utilisé on détaille sur des expériences une utilisation inaccessible au dépouillement classique.

1. Hypothèses utilisées dans la modélisation

1.1 Les effets de temps mort sont négligés

1.2 Réponse impulsionnelle de l'ensemble de comptage

La réponse impulsionnelle pour les neutrons de fission (probabilité $P(t)dt$ de détecter à l'instant t un neutron émis à l'instant 0) se termine par une décroissance exponentielle. Plus précisément on a:

$P(t)=P_{tr}(t)$ pour $t<\theta$

$P(t)=\epsilon\alpha e^{-\alpha t}$ pour $t>\theta$

Le terme transitoire P_{tr} n'est pris en compte que par son intégrale. La modélisation fait donc intervenir 4 paramètres:

$\epsilon_f=\int_0^\infty P(t)dt$: efficacité totale de détection pour les neutrons de fission

θ , ϵ et α déjà définis.

1.3 Conséquence pour les neutrons détectés dans des fenêtres déclenchées avec un retard suffisant.

Le SCRD, perfectionnement de l'appareil original décrit par BOEHNEL, examine la répartition des neutrons détectés dans des fenêtres ouvertes avec un certain retard après détection d'un neutron.

Considérons les neutrons détectés dans une telle fenêtre **et corrélés avec l'ouverture de cette fenêtre**. Les renseignements connus sur ces neutrons sont les suivants:

-ils appartiennent à une cascade de fission émise antérieurement à la détection du neutron qui a ouvert la fenêtre.

-la probabilité de détecter à l'instant t un neutron émis à l'instant 0 est $P(t)dt$

par conséquent, si t est supérieur à θ , ce qui est toujours réalisé lorsque le retard est supérieur à θ , ces neutrons sont détectés avec la probabilité $\epsilon\alpha e^{-\alpha t}dt$ et rien ne distingue ces détections de celles qui se seraient produites si la réponse $P(t)$ était purement exponentielle. Or dans ce cas nous savons calculer la répartition de ces neutrons[2,3]

Dans toute la suite θ désigne le retard avec lequel les fenêtres corrélées sont ouvertes après détection d'un neutron. Il est au moins égal à la durée du transitoire.

1.4 Récapitulation

Le SCRD compte les coïncidences d'ordre I suivant 2 modes:

- accidentelles : $A(I)$ coïncidences d'ordre I
- réelles + accidentelles: $(R+A)(I)$ coïncidences d'ordre I

Ces comptages permettent d'estimer les probabilités correspondantes $a(I)$ et $(r+a)(I)$

Les probabilités de coïncidences réelles $r(I)$ sont liées aux probabilités évaluées par la mesure par:

$$(r+a)(I)=\sum_{i=0}^{I} a(i).r(I-i) \qquad (1)$$

La modélisation nous permet de passer des probabilités $p(\nu)$ d'émission de ν neutrons à la source aux probabilités $r(I)$, et inversement.

La modélisation ne nous permet pas de passer des caractéristiques de la source (les $p(\nu)$ et l'intensité S cascades/s) aux quantités directement mesurées. (Sauf dans le cas d'une source poissonienne: $p(1)=1$, où on a $r(0)=1$ et où a et $(r+a)$ suivent la loi de POISSON de paramètre $\epsilon_f S$ quelle que soit la forme de $P(t)$)

2. Utilisations

Méthode générale

On utilise systématiquement une reconnaissance de forme pour identifier la nature de la source: en effet lorsqu'une source a été reconnue le comptage total est suffisant pour déterminer la quantité de matériau émetteur.

La reconnaissance de forme utilise la distance de 2 histogrammes H_1 et H_2 à 9 positions définie par $d(H_1,H_2)=2\sin(\phi/2)$, où ϕ est l'angle associé à la norme euclidienne: si $H_1(i)$ et $H_2(i)$ sont les nombres d'observations égales à i dans les 2 histogrammes,

$$\cos\phi=\frac{\sum_{i=0}^{8} H_1(i)H_2(i)}{\sqrt{\sum_{i=0}^{8} H_1(i)^2 \sum_{i=0}^{8} H_2(i)^2}}$$

Une distance sur les histogrammes de dimension 9 ayant ainsi été définie, on peut définir sur l'ensemble des histogrammes H_i résultats d'une série de n mesures, et donc de nature aléatoire, des quantités justiciables d'un traitement analogue à celui effectué pour les variables aléatoires à valeurs réelles. Parmi ces notions nous utilisons seulement:

-étendue d'une série de mesures: maximum de la distance $d(H_i,H_j)$ de 2 histogrammes appartenant à la série

-histogramme moyen: défini par

$$\bar{H}(I)=\sum_{i=1}^{n} H_i(I)$$

Remarque: Il est tentant de prendre comme définition de l'histogramme moyen l'histogramme des valeurs moyennes, c'est à dire de diviser par n le second membre de la formule précédente. Mais cela ferait perdre de vue le fait que les histogrammes expérimentaux sont constitués de valeurs numériques entières et que le nombre total N d'événements contenus dans un tel histogramme joue un rôle très important. La formule "intuitive" est vraie pour les histogrammes de fréquence associés aux 2 membres.

-dispersion expérimentale:

$$\bar{d} = \frac{1}{n}\sum_{i=1}^{n} d\left(\bar{H}, H_i\right)$$

La notion de convergence d'une suite infinie d'histogrammes à k valeurs associée à la distance d:

"La suite H_n tend vers H si $d(H_n, H)$ tend vers 0"

est liée à la convergence vers la loi de probabilité h des histogrammes expérimentaux H_N comportant N événements indépendants tirés sur cette loi par la formule[4]:

$$d(H_N, h) \approx \sqrt{\frac{k\left(1 - \frac{(d_0)^2}{2}\right)^2 - 1}{N}} \qquad (2)$$

avec $d_0 = d(h, h_0)$, où h_0 est la répartition plate $h_0(I) = 1$. Pour préciser quantitativement le "peu différent de" dans la formule (2) nous avons calculé la moyenne et la variance de la variable aléatoire $(d(H_N, h))^2$ associée à cette suite. On obtient, pour la loi dont les probabilités sont p_i:

$$E(d^2) = \frac{\sum p_i^2 - \sum p_i^3}{N(\sum p_i^2)^2}$$

$$Var(d^2) = \frac{2(\sum p_i^3)^2 - 4\sum p_i^4 \sum p_i^2 + 2(\sum p_i^2)^3}{N^2(\sum p_i^2)^4}$$

La formule (2) avait été obtenue dans la référence 4 comme extrapolation à partir de certains cas particuliers. On constate qu'elle est équivalente à l'approximation toujours dangereuse en l'absence de calcul: $X = \sqrt{E(X^2)}$ On constate que, l'écart type de d^2 étant du même ordre (1/N) que sa moyenne, cette approximation est correcte mais ne permet pas un calcul précis. Elle est suffisante pour évaluer à un facteur 4 près la durée nécessaire à une mesure, et nous n'en faisons pas d'autre usage.

L'important est de noter que, comme l'indique la formule (2), les valeurs de d correspondant à un risque de première espèce donné dépendent de la forme de la fonction h . Il est donc nécessaire pour en avoir une évaluation d'étalonner l'appareil dans une zone voisine des utilisations prévues. Nous en donnerons un exemple complet.

Cette reconnaissance de forme, pour être interprétable, doit donc être effectuée sur les résultats expérimentaux eux-même, mais la manière de procéder dépend de ce que l'on sait ou croit savoir sur la source avant la mesure, comme dans tout problème de probabilités. Pratiquement on peut distinguer 4 grandes classes de problèmes:

2.1 Contrôle de qualité en fin de fabrication (Exemple: assemblages neufs de réacteurs)

Les dispositions géométriques, compositions isotopiques, et répartitions volontaires ou non des impuretés sont a priori identiques. On doit donc s'attendre à des distributions identiques et il faut vérifier les corrélations au niveau de l'un des histogrammes A ou $(R+A)$. (Chaque histogramme contient en effet l'information complète, ce qui est à l'origine d'approches différentes de celle de BOEHNEL. Un accord obtenu sur l'une des répartitions sans être obtenu sur l'autre est l'indice d'un dysfonctionnement de l'appareil de mesure, comme le serait l'obtention de répartitions A et $(R+A)$ identiques sans être poissoniennes). Une telle exploration systématique n'a pas été mise en oeuvre à notre connaissance avec le SCRD. Il est certain que l'examen détaillé des histogrammes expérimentaux permettrait de retrouver des variations très fines (reconnaissance, dans des éléments MOX, de ceux issus d'un même lot de fabrication,...). Une telle reconnaissance de forme n'implique nullement une modélisation mathématique capable d'expliquer cette forme.

2.2 Suivi d'une masse de plutonium fissile lors de transformations physicochimiques. (assemblages non multiplicatifs)

Lors de ces transformations la composition isotopique du plutonium ne varie pas. L'émission de neutrons par fission spontanée reste donc constante. Par contre l'émission de neutrons de réaction (α ,n) varie suivant la présence et la répartition des autres éléments, d'une façon importante et peu reproductible, même lorsque le Pu est le seul émetteur α présent. Par conséquent dans la répartition $p(\nu)$ de l'émission des neutrons à la source, seul $p(1)$ varie, les autres probabilités restant proportionnelles à des nombres fixes. Dans ces conditions il suffit de mesurer un paramètre pour caractériser complètement la source et remonter à la masse de Pu. C'est ce que permet l'appareil initial. Par contre ce mode opératoire ne permet pas de détecter une substitution accidentelle d'une partie de matière fissile par une autre.

L'utilisation spécifique du SCRD que nous présentons se situe dans ce cadre et tient compte de ces éléments. La partie 3 détaille cette utilisation.

2.3 Reconnaissance du caractère multiplicatif d'un échantillon fissile

Une étude récente[5] de la répartition des neutrons de fuite d'une cascade de fission montre que la probabilité de détecter plus de 8 neutrons d'une même cascade devient accessible à la mesure même pour des amplifications inférieures à 2. Il est donc possible de reconnaître sans aucune hypothèse le caractère multiplicatif d'une source. La modélisation en l'absence de temps mort permet de traiter ce cas jusqu'à la caractérisation complète d'une source multiplicative[3], mais l'importance des détections multiples de rang élevé nous conduit à penser que les effets de temps mort sont considérables et ce point sera examiné après reprise complète de l'étude.

2.4 Reconnaissance des matériaux fissiles dans les déchets (prospectif)

Les utilisations précédentes consistent à vérifier la nature d'une source lorsque cette nature est annoncée. Une utilisation plus fine consisterait à reconnaître une source dont la nature n'est pas annoncée a priori. D'après les données actuelles, les sources existantes sont assez différentes les unes des autres pour que cette reconnaissance soit envisageable. Une condition nécessaire de cette utilisation est le réalisme du modèle de l'ensemble ralentisseur.

3. Détail d'une utilisation spécifique: contrôle de la nature et de la masse d'un échantillon de Pu (échantillon transparent aux neutrons)

3.1 Principe (l'organigramme figure 1 résume ce paragraphe)

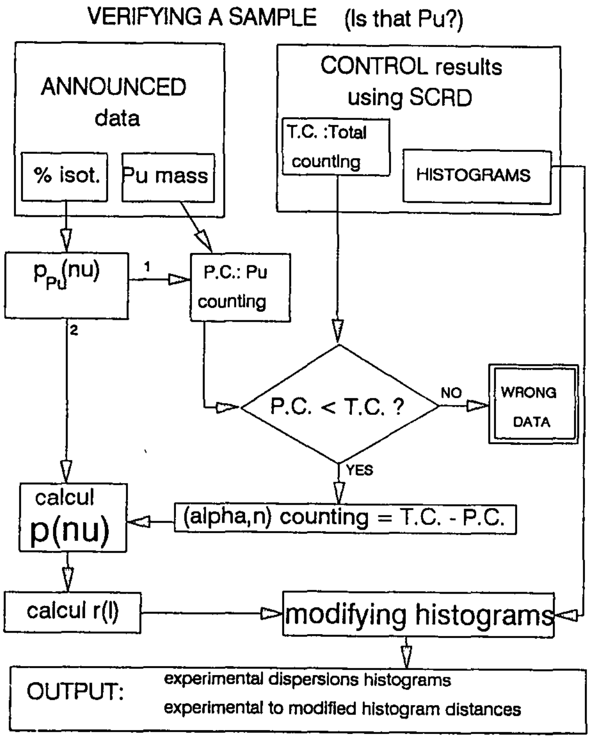

Figure 1

Les données à contrôler concernant le Pu réputé présent dans l'échantillon sont affichées. Ces données fournissent:

-le nombre S_{Pu} de fissions spontanées par seconde

-les probabilités $p_{Pu}(\nu)$ d'émission de ν neutrons lors d'une fission

Le taux de comptage dû aux neutrons de fission émis par le Pu est alors

$C_{Pu} = S_{Pu}\bar{\nu}\epsilon_f$ *coups/s*

Le premier résultat de mesure utilisé est le taux de comptage total. Il suffit, s'il est inférieur à la valeur attribuée aux neutrons de fission du Pu, pour refuser l'affirmation sur la source.

La différence C_p entre le taux de comptage total et la valeur attribuée aux neutrons de fission du Pu est $*^{t}$-attribuée, si elle est positive, à un comptage de neutrons (α ,n). Ce signal parasite, qui est poissonien, peut être considéré comme la réponse du système à une source parasite poissonienne d'intensité $S_p = C_p/\epsilon$ pour laquelle le système de détection aurait la réponse $P(t) = \epsilon\alpha e^{-\alpha t}$

L'ensemble "valeurs affichées + taux de comptage total" peut être remplacé par l'affirmation:

L'échantillon contient une source neutronique d'intensité $S_{Pu} + S_p$ cascades par seconde.

La probabilité d'avoir ν neutrons émis dans une cascade est:

$$p(\nu) = \frac{S_{Pu}\, p_{Pu}(\nu)}{S_{Pu} + S_p} \qquad pour \qquad \nu \neq 1$$

$$p(1) = \frac{S_{Pu}\, p_{Pu}(1) + S_p}{S_{Pu} + S_p}$$

La modélisation fournit les $r(l)$ correspondant aux $p(\nu)$

On utilise maintenant les résultats fournis par le SCRD et non accessibles à l'appareil initial: ces résultats sont 2 histogrammes A et $(R+A)$ comportant N coups et tirés sur les lois inconnues a et $(r+a)$. Ces lois vérifient le système (1). Le système

$$(R+A)^*(l) = \sum_{i=0}^{l} A^*(i).r(l-i) \qquad (1^*)$$

doit donc être vérifié par des quantités différant au plus de quelques écarts types des comptages obtenus.

Le comptage $A(l)$ suit la loi binomiale de moyenne $N.a(l)$ et d'écart-type $\sigma_A(l) = \sqrt{N.a(l)(1-a(l))}$. Ces valeurs peuvent être estimées en prenant $a(l) = A(l)/N$

De même pour le comptage $(R+A)(l)$ et la loi $(r+a)$

On modifie symétriquement les valeurs de $A(0)$ et $(R+A)(0)$ de k écarts types de façon à restituer la première équation du système (1*): k est défini par

$$r(0) = \frac{A(0) + k\sigma_A(0)}{(R+A)(0) - k\sigma_{(R+A)}(0)}$$

Les modifications:

$$A^*(0) = A(0) + k\sigma_A(0)$$

$$(R+A)^*(0) = (R+A)(0) - k\sigma_{(R+A)}(0)$$

sont effectuées si elles ne conduisent pas à des valeurs négatives (dans le cas contraire il y a impression d'un message et abandon du fichier en cours)

Ces modifications étant faites on modifie de même les $A(1)$ et $(R+A)(1)$ pour vérifier l'équation suivante.

En fin de traitement le programme affiche pour chaque histogramme:

-la valeur de k de plus grande valeur absolue utilisée, et la valeur de l pour laquelle elle a été utilisée.

-la dispersion expérimentale de chaque histogramme et la distance entre un histogramme traité et l'histogramme modifié correspondant.

3.2 Résultats expérimentaux

3.2.1. Ensemble de détection utilisé. Réalisme du modèle.

La figure 2 schématise l'ensemble de detection à 18 compteurs sur lequel nous avons effectué les mesures.

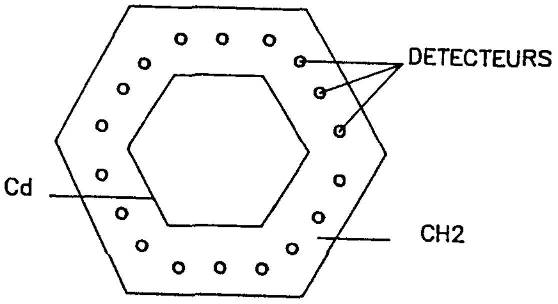

Fig 2 Ensemble de comptage

Pour cet ensemble de comptage nous avons pu effectuer une mesure directe de $P(t)$ en utilisant une source de Cf_{252} très mince et de très faible intensité: les fissions sont caractérisées par la détection d'un fragment de fission et on note la détection des neutrons dans les 3 ms qui suivent une fission (pas de 1 µs).

La figure 3 présente l'histogramme obtenu.

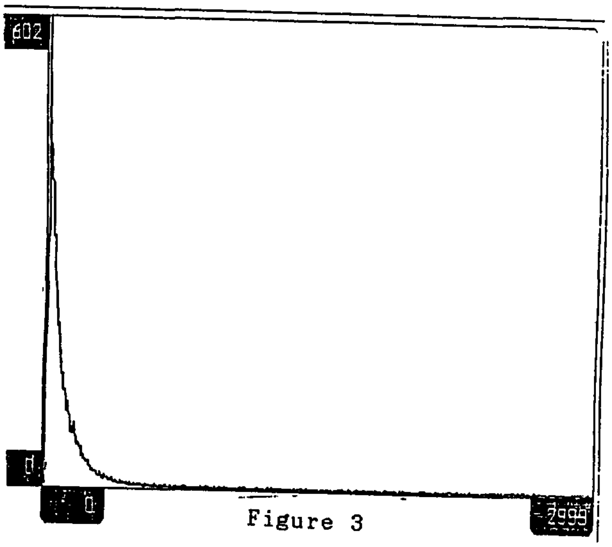

Figure 3

Pour cet ensemble de détection très hétérogène, il n'y a pas de décroissance exponentielle pure: ceci est assez net pour être visible avec un simple dépouillement graphique sur la courbe tracée en semi-log (Fig 4): le maximum est atteint 4 µs après le début du signal. On a ensuite une pseudo-pèriode de 42 µs pendant 27 µs, puis une pseudo-pèriode de 105 µs pendant 200 µs. Dans ces conditions on ne peut pas espérer du modèle autre chose qu'un accord dans une certaine zone après calage expérimental des paramètres non réalistes ϵ et α. Ce procédé est détaillé sur le premier exemple.

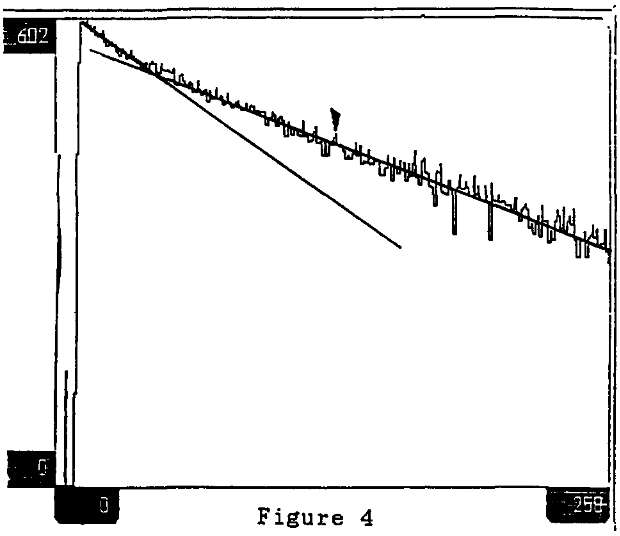

Figure 4

L'efficacité de détection pour les neutrons de fission est ϵ_f = 0.0725 coup/neutron

Pour chaque largeur T de fenêtre les valeurs initiales des paramètres de calage sont obtenues par lissage numérique de la forme $P(t)=\epsilon\alpha e^{-\alpha t}$ sur la portion de courbe centrée à 68 µs (point médian de la répartition) et de même largeur que la fenêtre. Ces valeurs sont:

$T=60.56\mu s$ $\epsilon=0.067$ $1/\alpha=71.2\mu s$

$T=121.6\mu s$ $\epsilon=0.067$ $1/\alpha=63.4\mu s$

3.2.2 Description d'une série de mesures

On a effectué avec une source de Cf252 et une source Am/B beaucoup plus intense une série de mesures consistant à placer la source Cf au centre de l'ensemble de comptage et la source Am plus ou moins loin à l'extérieur. Chaque mesure comporte 5 comptages de 100 s. Le tableau suivant donne les réglages utilisés: seul le retard varie. Dans chaque groupe de mesures on a la source Cf seule, puis 3 comptages avec des positions différentes de la source Am. On indique d'une part le comptage total et le comptage $R_s=(R+A)_s-A_s$ du dépouillement classique de BOEHNEL, et d'autre part la valeur $R(0)=(R+A)(0)/A(0)$, évaluation expérimentale de $r(0)$, qui est utilisée pour le calage du paramètre ϵ

RECAPITULATIF FICHIERS MESURES
Durée=500 s Tcoïn=121.6 µs

FICHIER	θ (µs)	TOTAL	R_s	$R(0)$
88CF	7.15	1 357 273	119 782	0.91582
88CF20	7.15	1 694 017	117 228	0.93452
88CF50	7.15	2 706 379	118 140	0.95962
88CFAM	7.15	3 702 987	114 531	0.97097
78CF	6.20	1 342 836	117 639	0.91681
78CF20	6.20	1 694 833	118 594	0.93368
78CF50	6.20	2 709 026	117 147	0.95968
78CFAM	6.20	3 704 354	118 016	0.96978
68CF	5.25	1 344 140	119 791	0.91494
68CF20	5.25	1 697 172	118 760	0.93334
68CF50	5.25	2 707 409	117 013	0.95945
68CFAM	5.25	3 701 750	111 305	0.97222

Remarque préliminaire sur la qualité de ces mesures:

La source Cf étant la même, les comptages TOTAUX devraient être identiques lorsque la source Cf est seule. Or les comptages TOTAUX de 88CF et 78CF sont 1 357 273 et 1 342 836

La différence 14 437 est 12.4 fois la racine du plus grand nombre.

Le dépouillement de chaque série est effectué indépendamment, sans tenir compte de cette remarque qui n'aurait pas pu être faite dans le cas opérationnel d'une seule série de mesure. Ce dépouillement suit le schéma classique auquel on est obligé de se plier lorsque l'on utilise des paramètres d'un modèle non réaliste, et/ou un ensemble de comptage mobile assez petit pour risquer d'avoir une réponse variable avec l'environnement:

a) On effectue une mesure sur une source bien connue, de préférence de même nature que les sources à contrôler

b) On ajuste le ou les paramètres du modèle pour obtenir le meilleur accord expérience-calcul

c) On regarde si le résultat théorique ainsi obtenu est à l'intérieur du domaine de la dispersion expérimentale. SI NON on déclare que la méthode testée ne peut pas être employée dans cet environnement.

d) SI OUI on utilise les paramètres obtenus par calage pour chercher les caractéristiques des sources à contrôler.

Pour l'ajustement à 2 paramètres utilisé ici nous avions constaté sur une mesure ancienne que l'on obtenait une plage étendue de couples (ε,α) donnant un accord de même qualité (C'est ce fait qui nous avait amenés à penser que le modèle exponentiel, universellement admis, est irréaliste pour cet ensemble de comptage, comme l'a confirmé la mesure directe) Dans le calage effectué ici on a donc conservé le α obtenu par lissage et ajusté seulement ε , ce qui conduit à des calculs plus courts.

DEPOUILLEMENT DE LA SERIE "88CF"

a,b,c) Calage sur le fichier 88CF: le bruit de fond étant négligeable dans cette série de mesures on considère que le comptage total est dû au Cf seul.

On connait la valeur du paramètre $\epsilon_f = 0.0725$

On en tire la masse de Cf de 1.621 10^{-8} g

On choisit la valeur de α obtenue par lissage de la réponse impulsionnelle: $1/\alpha = 63.4 \mu s$

On détermine la valeur de ε en restituant, dans le calcul des r(I), la valeur expérimentale R(0): on a obtenu $\epsilon = 0.136$ **(Cette valeur étant très différente de la valeur 0.067 obtenue lors du lissage le couple (α,ε) est dépourvu de signification physique)** La comparaison des dispersions expérimentales des histogrammes du fichier et de la distance entre les histogrammes expérimentaux et modifiés (voir premières lignes du tableau suivant) montre que les *r(I)* obtenus par le modèle sont compatibles avec les expériences à la précision où elles ont été effectuées.

d) Utilisation du modèle avec ces valeurs de paramètres

On effectue sur chaque fichier le traitement décrit figure 1: un fichier contient toutes les données, on a rentré l'isotope (Cf_{252}), et il ne reste plus qu'à indiquer la masse m. On a obtenu les résultats suivants (Disp= dispersion expérimentale de l'histogramme considéré; Dist= distance entre l'histogramme expérimental et l'histogramme obtenu après calcul)

FICHIER 88CF: Disp (R+A) = .00081 Disp A = .00061

$m(g)*10^8$	kmax	Imax	Dist(R+A)	Dist(A)
1.4	-2.5	0	.00479	.00407
1.6	+2.2	1	.00091	.00076
1.621	+2.1	1	.00067	.00054
1.8	+6.0	0	.00317	.00264

FICHIER 88CF20: Disp (R+A) = .00149 Disp A = .00150

$m(g)*10^8$	kmax	Imax	Dist(R+A)	Dist(A)
1.2	-7.0	0	.00392	.00353
1.4	+0.7	1	.00030	.00027
1.6	+6.1	0	.00324	.00290

FICHIER 88CF50: Disp (R+A) = .00148 Disp A = .00150

$m(g)*10^8$	kmax	Imax	Dist(R+A)	Dist(A)
1.0	-5.5	0	.00263	.00255
1.2	+1.5	6	.00032	.00030
1.4	+4.4	0	.00232	.00224

FICHIER 88CFAM: Disp (R+A) = .00126 Disp A = .00163

$m(g)*10^8$	kmax	Imax	Dist(R+A)	Dist(A)
1.0	-2.5	0	.00132	.00130
1.2	+1.1	0	.00061	.00060
1.4	+4.8	0	.00222	.00219

On constate que pour les 3 derniers fichiers on obtient une valeur de m permettant de restituer les *r(I)* calculés à l'aide de modifications des histogrammes nettement à l'intérieur de leur dispersion expérimentale. Ces masses sont plus faibles que celle choisie, mais on avait remarqué au départ que le comptage total du fichier 88CF était anormalement élevé

Le fait que l'accord pour cette mesure principale nécessite une distorsion des histogrammes voisine de la dispersion expérimentale affichée, alors que pour les autres fichiers on obtient moins de la moitié, pourra sans doute être utilisé comme un signal d'alarme lorsque nous disposerons d'une habitude suffisante de ce type de dépouillement. Nous allons maintenant examiner sur ce cas peu favorable la possibilité de discrimination de source que nous proposons:

PEUT-ON PRENDRE CETTE SOURCE Cf POUR UNE SOURCE Pu240?

88CF NON: R(0) est supérieur à 0.955, pour une source de Pu240, avec ces valeurs de paramètres.
88CF20 NON: même motif
88CF50 OUI: en annonçant 68g de Pu240 on obtient: kmax=-2.8, Imax=1, Dist(R+A)=.00086, Dist(A)=.00082
en annonçant 70g de Pu240 on obtient: kmax=-2.7, Imax=1, Dist(R+A)=.00095, Dist(A)=.00091
88CFAM OUI: en annonçant 60g de Pu240 on obtient: kmax=-1.4, Imax=1, Dist(R+A)=.00063, Dist(A)=.00062

On constate qu'à la précision de ces mesures l'examen des A(I) et (R+A)(I) pour I > 0 n'apporte pas de renseignements supplémentaires. Nous avons évalué la statistique nécessaire à la discrimination dans le cas du fichier 88CFAM, qui est évidemment le plus défavorable, de la façon suivante:

-on prend la répartition expérimentale A(I) obtenue lors du premier comptage

-on calcule les répartitions:

$$(R+A)(I) = \sum_{i=0}^{I} r(i)A(I-i)$$

obtenues en prenant les r(I) calculés pour le Pu et le Cf

-on calcule la distance d entre ces 2 répartitions. On a obtenu d=1.03 10^{-3}

-on évalue par la formule (2) la distance $\bar{d}$ à attendre entre la répartition (R+A) obtenue pour le Cf et la vraie répartition (r+a) correspondante. On a obtenu $\bar{d}$ = 1.69 10^{-3} pour la statistique utilisée de 739 349 coups. (Cette valeur est bien compatible avec la valeur expérimentale).

Il suffirait donc d'une statistique 16 fois plus forte que le fichier élémentaire, soit à peine 3 fois plus forte que celle du fichier somme, pour effectuer cette discrimination. On peut penser qu'à ce niveau on ne se heurterait pas encore à l'insuffisance du modèle.

Dans le cas d'échantillons multiplicatifs les r(I) correspondant à la même valeur moyenne de I(I-1) sont très éloignés de ceux obtenus ici.

DEPOUILLEMENT DE LA SERIE "78CF"

a,b,c) Calage sur le fichier 78CF:

Le comptage total donne une masse de Cf de 1.604 10^{-8} g

Le calage donne $\epsilon = 0.132$

d) Utilisation de ces paramètres. On a obtenu les résultats suivants

FICHIER 78CF: Disp (R+A) = .00157 Disp A = .00120

m(g)*10^8	kmax	Imax	Dist(R+A)	Dist(A)
1.4	-7.3	0	.00444	.00378
1.6	+1.1	1	.00045	.00037
1.604	+1.1	1	.00039	.00032
1.8	+6.3	0	.00352	.00295

FICHIER 78CF20: Disp (R+A) = .00091 Disp A = .00119

m(g)*10^8	kmax	Imax	Dist(R+A)	Dist(A)
1.4	-2.4	0	.00139	.00154
1.5	+1.1	1	.00052	.00045
1.6	+3.9	0	.00202	.00120

FICHIER 78CF50: Disp (R+A) = .00119 Disp A = .00174

m(g)*10^8	kmax	Imax	Dist(R+A)	Dist(A)
1.3	+0.9	0	.00055	.00053
1.4	+3.3	0	.00171	.00165
1.5	+5.6	0	.00286	.00276

FICHIER 78CFAM: Disp (R+A) = .00049 Disp A = .00113

m(g)*10^8	kmax	Imax	Dist(R+A)	Dist(A)
1.3	-1.3	4	.00062	.00061
1.4	+2.9	0	.00142	.00139

DEPOUILLEMENT DE LA SERIE "68CF"

a,b,c) Calage sur le fichier 68CF:

Le comptage total donne une masse de Cf de 1.605 10^{-8} g

Le calage donne $\epsilon = 0.132$

d) Utilisation de ces paramètres. On a obtenu les résultats suivants

FICHIER 68CF: Disp (R+A) = .00163 Disp A = .00115

m(g)*10^8	kmax	Imax	Dist(R+A)	Dist(A)
1.4	-7.6	0	.00501	.00425
1.6	+2.9	1	.00115	.00095
1.605	+2.9	1	.00109	.00089
1.8	+6.2	0	.00327	.00271

FICHIER 68CF20: Disp (R+A) = .00102 Disp A = .00147

m(g)*10^{8}	kmax	Imax	Dist(R+A)	Dist(A)
1.4	+1.6	1	.00111	.00100
1.5	+1.9	0	.00090	.00079
1.6	+5.0	0	.00227	.00254

FICHIER 68CF50: Disp (R+A) = .00121 Disp A = .00202

m(g)*10^{8}	kmax	Imax	Dist(R+A)	Dist(A)
1.2	+1.5	3	.00053	.00048
1.3	+1.7	0	.00092	.00090
1.4	+4.1	0	.00207	.00201

FICHIER 68CFAM: Disp (R+A) = .00136 Disp A = .00140

m(g)*10^{8}	kmax	Imax	Dist(R+A)	Dist(A)
1.1	+1.3	3	.00062	.00061
1.2	+2.0	0	.00107	.00106
1.3	+3.8	0	.00184	.00182

On constate que pour ces valeurs de calage on a souvent un accord bien meilleur. Des comptages réalisés plus tard dans les mêmes conditions ont permis de vérifier que le comptage total du fichier 88CF est anormalement élevé. Cette aberration n'est pas d'un type qui aurait pu être détecté dans une mesure de routine.

En se basant sur les qualités d'accord obtenues dans les autres mesures on pourrait dire que la statistique utilisée est suffisante pour refuser l'hypothèse d'une source de Pu. Nous pensons que ce point qui reste notre objectif à court terme demande des expériences plus précises. Un point important est de voir comment varient les paramètres de calage si celui ci est effectué à l'aide d'une source de Pu ou d'une source de Cf.

Conclusion

La première série d'expériences destinée à tester sur une source connue la qualité de la modélisation du SCRD avant d'utiliser cette modélisation pour caractériser des échantillons multiplicatifs, est nettement insuffisante et devra être reprise.

Par contre la mesure de la réponse impulsionnelle, qui fera l'objet d'une description détaillée, est précise et montre que le modèle exponentiel n'est pas réaliste pour l'ensemble de détection utilisé, même sous la forme élaborée que nous utilisons (prise en compte d'un transitoire avant la phase exponentielle). La prise en compte de ce fait peut a priori être envisagée de 3 façons:

a) **Développement du modèle pour une réponse quelconque**: tout à fait hors de question avec nos moyens actuels.

b) **Réalisation d'un ensemble de comptage à réponse plus proche du modèle** On sait que le modèle est réaliste pour un ensemble infini homogène. On peut donc s'en rapprocher en augmentant la taille du ralentisseur et en modifiant le nombre et la répartition des compteurs. Cette solution ne peut être envisagée que pour une installation fixe.

c) **Utilisation de l'ensemble de comptage et de la modélisation actuels**, en notant bien que toute série de mesures nécessite alors un calage et en prévoyant les sources calibrées nécessaires à ce calage.

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ACTIVE WELL COINCIDENCE COUNTER MEASUREMENTS OF ENRICHED URANIUM FUEL ASSEMBLIES IN SCANNING AND STATIONARY MODES*

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Abstract

Enriched uranium fuel assemblies were measured with an Active Well Coincidence Counter (AWCC) at the Beloyarskaya Nuclear Power Plant. Special AWCC inserts, electronics, and software were used. Stationary and scanning measurements were performed to establish calibrations and performance specifications for the assay of ^{235}U and ^{235}U/cm for BN600 fuel.

1. Introduction

A measurement exercise was conducted at the BN600 reactor facility at the Beloyarskaya Nuclear Power Plant to evaluate the Active Well Coincidence Counter (AWCC) /1/ for the nuclear safeguards verification of enriched uranium fuel assemblies. The normal AWCC technique is to induce fissions in the fuel with neutrons from americium-lithium (AmLi) isotopic neutron sources and to count the coincident neutrons from the fissions with coincidence circuitry. Once a calibration is established, the coincidence count rate determines the ^{235}U content of the fuel. However, because the enriched region of the fuel assemblies is much longer than the irradiation region of the AWCC, a single, stationary measurement of the fuel determines the ^{235}U mass per unit length; the length of the enriched region (the active length) must be measured separately to determine the total ^{235}U mass of the fuel.

One method for measuring the active length of the fuel is to manually scan the fuel assembly with a collimated gamma-ray detector. The disadvantage of the method for nuclear safeguards is that the gamma-ray measurement only determines the active length of the surface of the fuel. The operational disadvantages are that the safeguards inspector must handle measurement equipment close to the fuel assembly along its active length and must have a means for marking and measuring the distance along the assembly.

Another method of measuring the active length is to scan the fuel assembly with the AWCC. In this method the fuel assembly is lowered by crane through the AWCC and the total neutron count rate is measured in short counting intervals. The advantages of the method are that the fuel assembly does not need a wrapper for protection and for marking, inspectors and hand-held equipment do not need to be close to the assembly, and the measurement and analysis are largely automatic. The disadvantage is that the crane speed must be known or measured.

Instead of using a stationary measurement plus a scan for the verification of fuel assemblies, a single scan can suffice if it is slow enough that the error from counting statistics is as low as required for the type of verification being done. The active length is then not needed for the verification of the total ^{235}U content. (The scan, nevertheless, automatically provides the length as part of the analysis.) As before, the crane speed must be known or measured.

In this exercise both stationary and scanning measurements were evaluated. A more detailed report on this exercise will soon be available /2/.

2. Material and Methods

Facility and Fuel

The BN600 facility /3/ is a 600 MWe fast breeder reactor near the city of Sverdlovsk, USSR. The enriched uranium fuel assemblies have enrichments of 17%, 21%, or 26%. The enriched assemblies used for this exercise have 100-cm active lengths with adjacent 30-cm depleted regions at each end. A simplified drawing of a fuel assembly is shown in Fig. 1.

The assemblies have an hexagonal cross section with 9.6 cm across the flats. The outer cladding is stainless steel 0.2 cm thick. The enriched assemblies contain 127 UO_2 fuel pins with an outer diameter of 6.9 mm; the fuel-pin cladding is stainless steel 0.4 mm thick. Figure 2 shows the cross section of an enriched fuel assembly.

The ^{235}U masses of the assemblies vary from approximately 4700 g to approximately 7200 g.

Detector

The detector is a standard AWCC modified for use at the BN600 facility. The normal end plugs, nickel reflector, and cadmium liner were removed and replaced with a custom insert for the BN600 fuel. A 15-cm diam irradiation channel at one side of the polyethylene insert is for the fuel assembly. A 4.2-cm diam hole in the polyethylene insert is for the AmLi sources; 1.0 cm of polyethylene separates the AmLi sources from the 15-cm diam irradiation channel. Figure 3 shows vertical and horizontal cross sections of the modified AWCC.

The 15-cm diam hole is lined with cadmium 0.4 mm thick to prevent thermal neutrons from escaping the polyethylene and inducing fissions in the fuel assembly; i.e., the AWCC is operated in the fast mode. Because the ^{235}U mass in the fuel assemblies is high, the thermal mode

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*Los Alamos National Laboratory effort supported by the International Safeguards Project Office.

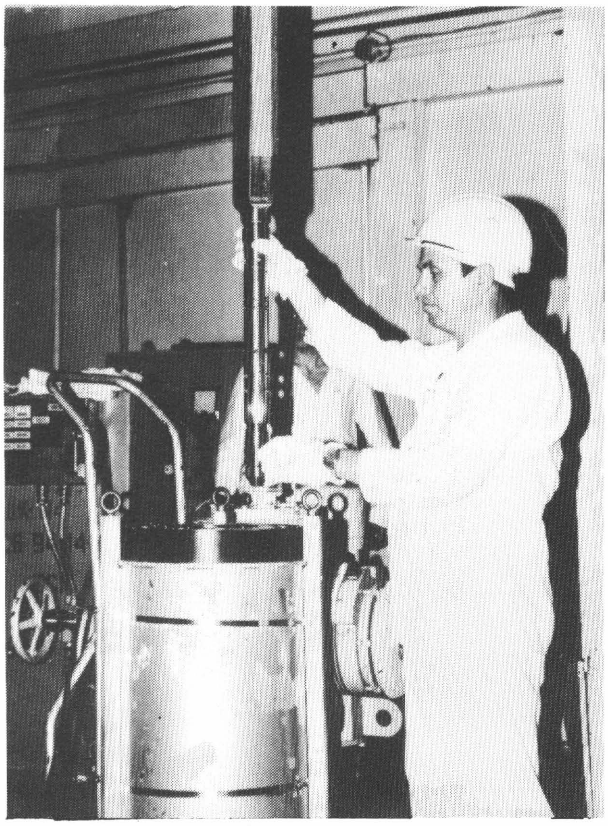

Fig. 4. BN600 fuel assembly being positioned for lowering into the AWCC.

measurements, the raw data from an SR4 circuit are almost the same as those from a JSR-11.

The SR4 was set to a predelay of 3 μs and a gate length of 64 μs.

A custom data collection and analysis program for IBM PCs was written to perform the stationary and scanning measurements. The SR4 coincidence module has no manual controls; it is operated from the computer via a 9600 baud RS-232-C serial port. Because the data transfer time is short, the raw data from the SR4 can be displayed on the computer display in near real time.

For stationary measurements, the counting rates, results, and errors are updated and displayed about once a second as the measurement proceeds; for scanning measurements, the total count rate is plotted on the computer display as the scan proceeds.

Experimental Fuel Handling

Two cranes were used for the scanning measurements; each crane had a fast and a slow speed. The small crane (5-tonne capacity) is the normal fuel-handling crane and has a downward slow speed of 3.6 cm/s. The large crane (30-tonne capacity) has a downward slow speed of 0.23 cm/s. The fast speeds of the cranes are about an order of magnitude faster than the slow speeds.

All scanning measurements were made using the slow crane speeds while the fuel was being lowered through the AWCC. For this exercise the AWCC was positioned over a fuel-transfer hole in the floor of the room. The 100-cm enriched region of an assembly passes through the center of the AWCC in about 30 s with the fast crane and in about 7 min with the slow crane.

Stationary Measurements

Two fuel assemblies of each enrichment were centered in the AWCC and measured in the stationary position. Most measurements were 1000 s long. A 1000 s measurement produces a coincidence-count-rate standard deviation from counting statistics of 2.0% for a 17%-enriched assembly and 1.5% for a 26%-enriched assembly.

The six calibration data points were fit by least squares using the Deming code /6/ and a calibration equation of the form

$$R = \frac{am}{1 + bm} ,$$

where R is the coincidence rate, m is the ^{235}U mass/cm, and the calibration constants are a and b. The calibration curve is plotted with the data points in Fig. 5.

Slow Scans

For the slow scanning measurements, the fuel assemblies were lowered through the AWCC with the slow crane at a speed of 0.23 cm/s. The SR4 coincidence circuit was cycled with a 3-s measurement time and the raw data were stored for each cycle. During the scan the totals count rate was plotted vs time on the computer display, so the progress of the scan could be observed. The totals count rate distribution for a slow scan of a 26%-enriched assembly is shown in Fig. 6; the totals rates are averaged over three cycles to smooth the distributions.

Eight slow scans were done with six different assemblies; all three enrichments were used.

The totals rate is not used directly to assay the ^{235}U mass of the fuel because the totals rate depends strongly on the scattering and absorption effects of the fuel on the AmLi source neutrons as well as on the ^{235}U induced fission rate. The totals rate scan, however, easily identifies the enriched region of the fuel and determines the active length; the coincidence counts obtained in the active region can then be used to assay the total ^{235}U mass of the fuel.

The enriched region of the fuel assembly is indicated in Fig. 6. The lower count rate above and below the enriched region is caused by neutron scattering and absorption in the depleted uranium, which does not contribute a significant number of induced fission neutrons. The complex distribution above the depleted region is caused by scattering and absorption effects from the end of the fuel assembly and the fuel-handling hardware.

Figure 7 shows the coincidence counts measured for the same scan; there are statistically significant coincidence counts only from the enriched region of the fuel assembly. The integral of the coincidence counts determines the total ^{235}U mass; an effective coincidence rate is calculated for the scan and the mass is obtained from the stationary calibration curve.

The shape of the curve of the totals rate vs time during the transitions between the enriched and depleted regions of the fuel is determined by the AWCC irradiation characteristics and the crane speed; the average absolute slope of the totals rate curve in the transition regions is used to calculate the crane speed. The width of the totals rate distribution, calculated as the distance between half-height positions in the transition regions, determines the active length.

Fast Scans

The short measurement time for fast scans (about 30 s for the active region) produces large measurement errors, but the fast scans can still be useful for verifying the active

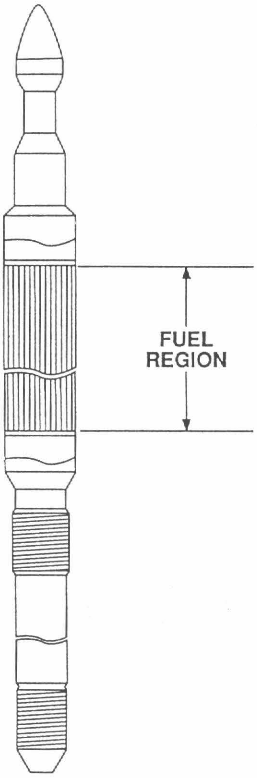

Fig. 1. Simplified drawing of a BN600 fuel assembly.

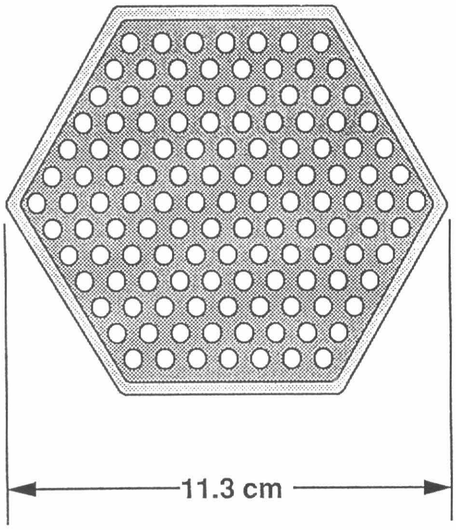

Fig. 2. Cross section of a BN600 fuel assembly with enriched uranium fuel.

is not satisfactory because of the poor thermal-neutron penetration into the fuel.

Cadmium is not used as a liner around most of the polyethylene insert so that the detection efficiency of the AWCC for induced-fission neutrons is high; however, a small rectangle of cadmium (about 10 cm wide by 20 cm long) is glued to the outside of the insert next to the AmLi sources to reduce the count rate from the AmLi sources.

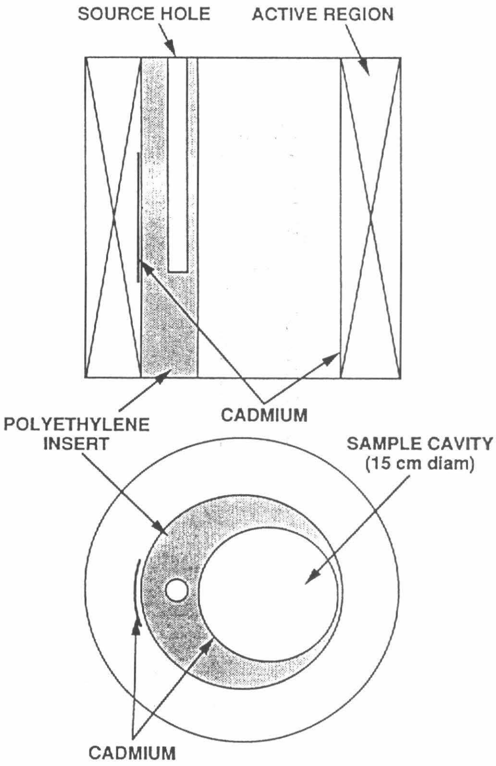

Fig. 3. Diagram of vertical and horizontal cross sections of the modified AWCC.

The AmLi neutrons do not produce real coincidence counts, but increase the error of the real coincidence counts by increasing the random neutron count rate.

A photograph of the modified AWCC in use at the BN600 facility is shown in Fig. 4; a fuel assembly is being positioned for a scanning measurement.

Two AmLi neutron sources are used to induce fissions in the fuel; each has a yield of about 50 000 neutrons per second. The two sources are placed in a custom, polyethylene source rod that slides into the source hole shown in Fig. 3.

Electronics and Software

The AWCC has AMPTEK /4/ preamplifiers in its junction box, so the AWCC requires +5 V to power the preamplifiers and +1680 V for the ^{3}He proportional counters; the output is standard TTL logic signals, consisting of neutron pulses 50 ns wide.

The standard JSR-11[*] coincidence counting circuit /5/ was not used for this exercise because short measurement and readout times were needed for the scanning measurements; the shortest measurement time is 1.0 s for the JSR-11 and the readout time is about 2 s.

Instead of the JSR-11, a new Los Alamos prototype NIM coincidence circuit, designated the SR4 (Shift Register; 4 MHz), was used because it has a minimum measurement time of 0.1 s and a readout time of 22 ms. The SR4 circuit will be described in a future publication, but the operating principle is the same as that for the JSR-11. At low count rates, such as obtained from AWCC

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[*] Jomar Systems, Los Alamos, NM, USA, 87544

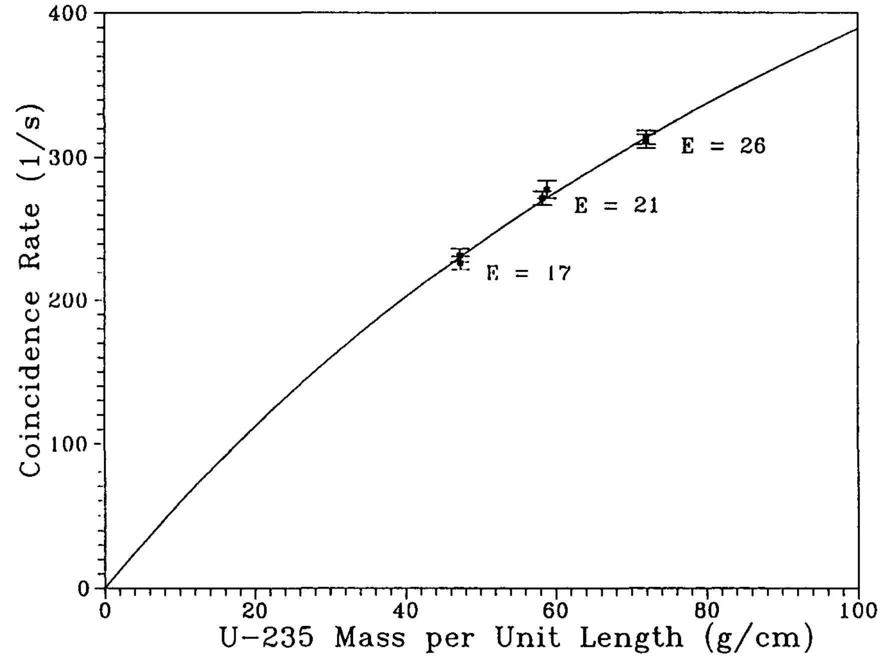

Fig. 5. Calibration curve for stationary assays of BN600 enriched uranium fuel assemblies.

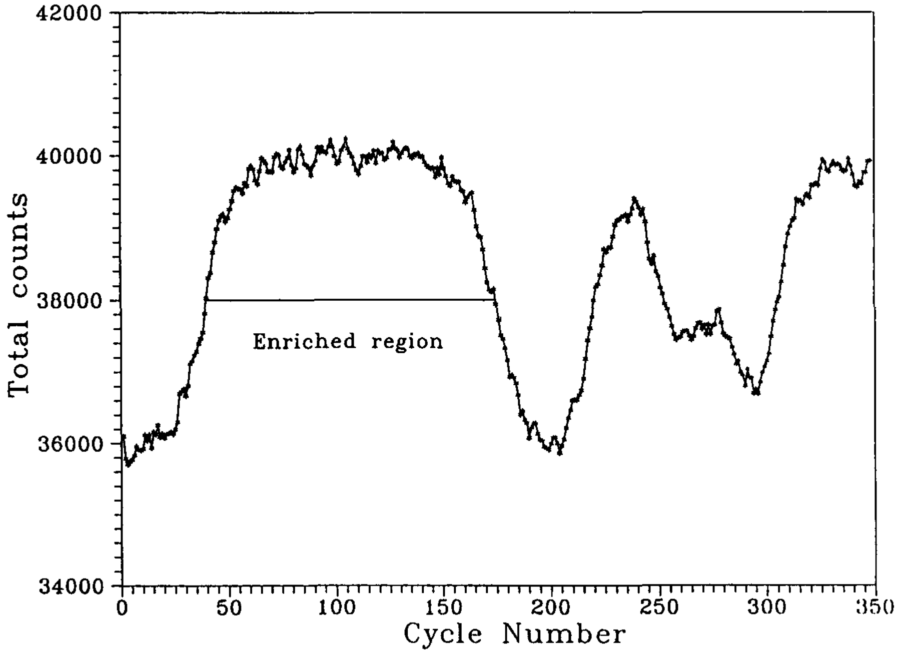

Fig. 6. Plot of the totals count rate for a slow scan of a 26%-enriched fuel assembly. The totals rate is plotted vs the cycle number; each cycle is a 3-s measurement.

lengths in connection with stationary measurements or for performing gross defect measurements. In general, the measurement and analysis of fast scans is the same as that for slow scans. The main differences are that the crane speed is 3.6 cm/s instead of 0.23 cm/s and the measurement time per cycle is 0.5 s instead of 3 s.

Seventeen fast scans were performed, ten of which were consecutive repetitions on a single 21%-enriched assembly.

The average active length initially measured from the 17 scans was 96.8 ± 1.2 cm, so the half-height to half-height analysis of the totals distribution underestimates the

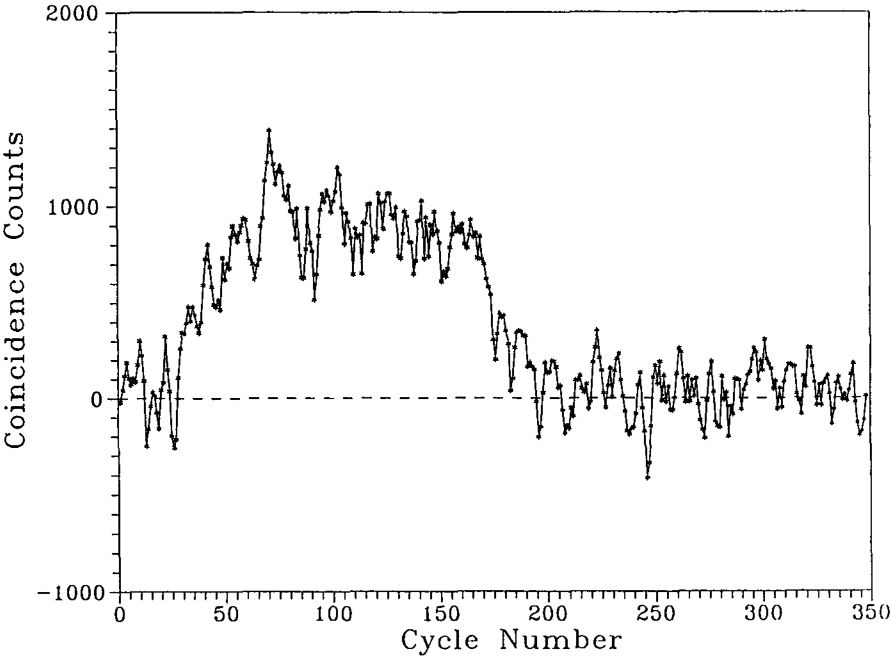

Fig. 7 Plot of the coincidence count rate vs the cycle number for the same scan as shown in Fig. 6 for the totals count rate.

active length by about 3% for fast scans. A correction factor of 1.033 was introduced to account for this bias.

3. Results

Stationary Measurements

Calibration constants were established for the stationary assay of enriched uranium BN600 assemblies. The standard deviation of the assay mass for a 1000-s measurement is 2.8% for a 17%-enriched assembly, 2.5% for a 21%-enriched assembly, and 2.4% for a 26%-enriched assembly.

Slow Scans

The mass, active length, and crane speed results for the eight slow scans are shown in Table I.

The standard deviations for the mass measurements are 3.9% for 26%-enriched fuel, 4.2% for 21%-enriched fuel and 4.6% for 17%-enriched fuel. The average absolute mass difference between the measured and accepted values is 3.9% and the average mass difference is -0.9%.

The average absolute active length difference between measured and accepted values is 1.1% and the average active length difference is 0.6%. The sample standard deviation of the active length measurements is 1.4%.

The eight slow scans were used to calibrate the crane speed measurements for slow scans, so the average crane speed in Table I is approximately the reference value of 0.23 cm/s. The sample standard deviation of the crane speed measurements is 6.4%.

Fast Scans

The standard deviation of the mass measurements is 22% for 26%-enriched fuel, 24% for 21%-enriched fuel and 26% for 17%-enriched fuel. The average absolute mass difference between measured and accepted values is 14.9% and the average mass difference is -0.4%. The sample standard deviation of the active length measurements is 5.1% and the sample standard deviation for the crane speed is 13%.

Summary

Table II summarizes the final results by giving the standard deviation in percent for the measurement of mass, active length, and crane speed for the three measurement types: stationary, slow scan, and fast scan. For stationary measurements, the mass refers to the ^{235}U mass per unit length of fuel, whereas for the scanning measurements the mass refers to the total ^{235}U mass in the assembly. The mass standard deviations are given for the three enrichments: 17%, 21%, and 26%.

If the crane speed is known, a partial defect measurement of the fuel assemblies for total ^{235}U mass can be done either by making a single slow scan for the total ^{235}U mass or by making a stationary measurement for the ^{235}U mass per unit length of fuel and a fast scan for the active length.

In either case the measurement of the crane speed from a single scan is not accurate enough for a partial defect measurement. If there is a possibility of crane speed variations, the crane speed can be checked with a stop watch during the scan.

More accurate measurements can be performed by combining a stationary measurement with a slow scan.

A single, fast scan can be used for a gross defect measurement.

Similar, but passive, scanning measurements could be used for the assay of MOX fuel assemblies. Because of the higher count rates, the precision of the measurements would be better than those for enriched uranium assemblies.

Table I. Slow Scan Results

Sample	Enrichment (%)	Active Length (cm)	Crane Speed (cm/s)	Accepted ^{235}U Mass (g)	Measured ^{235}U Mass (g)	σ (Meas. ^{235}U Mass) (g)	Δm^a (%)
1	26	100.3	0.216	7198	6997	276	−2.79
2	17	99.5	0.238	4733	4818	222	1.80
3	21	98.9	0.245	5832	5938	247	1.82
4	26	103.2	0.237	7197	6922	270	−3.82
5	21	101.0	0.223	5832	5655	236	−3.03
6	17	100.3	0.216	4733	5125	225	8.28
7	21	101.7	0.216	7176	6512	259	−9.25
8	21	99.6	0.254	7176	7183	284	0.10

a

$$\Delta m = \frac{\text{measured } ^{235}\text{U - accepted } ^{235}\text{U mass}}{\text{accepted } ^{235}\text{U mass}} \times 100$$

Table II. Standard Deviation Summary

Relative Standard Deviation (%)			
Quantity	Method		
	Stationary (1000 s)	Slow Scan (~600 s)	Fast Scan (~60 s)
Mass (E^a = 17%)	2.8	4.6	26
Mass (E = 21%)	2.5	4.2	24
Mass (E = 26%)	2.4	3.9	22
Active length	---	1.4	5.1
Crane Speed	---	6.4	13

^{a} E = Enrichment

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ANALYSIS OF THE VDC RESULTS IN THE PASSIVE NEUTRON ASSAY.

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Abstract

Analysis of the variable dead-time logic for neutron coincidence counting has been performed. An analytical expression for the coincidence rate was derived that defines its relationship with the fission rate, variable dead time counter characteristics and neutron multiplicity distribution taking into account induced fissions. Usefulness of results correction for methodical error was tested at count rates up to about 30 kHz.

Application of the known formalism was examined for multiplication correction using the ratio of coincidence rate to the total count rate. The proposed assay algorithm was used for plutonium measurements in oxide fuel samples containing up to 100 g plutonium.

1. Introduction

For non-destrictive assay of nuclear materials by passive neutron counting various techniques and instruments have been developed which allow fission neutrons to be distinguished from neutrons arising from other processes using the time-correlated nature of fission neutrons. One of the simple instruments used for such purpose is a thermal neutron detector with a variable dead-time circuit (VDC) of neutron coincidence counting. The principle of neutron coincidence selection by the VDC-technique consists in counting of pulses from the neutron detector in two counting channels simultaneously: in the fast channel with a small dead time and in the slow one with the present dead time which value is higher than the neutron lifetime in the detector. The method is based on the assumption that several of simultaneously emitted neutrons in the fission event can be counted in the fast channel and only one of them in the slow one. Thus, the counting losses in the slow channel include a component depending on fission rate.

Because of the limitation in allowable counting rates this method has not been widely adopted in practice in comparison with the shift register coincidence technique. But the simplicity of hardware design justifies its application for measuring of small amounts of spontaneously fissioning materials.

2. Analysis of the VDC results

In the passive counting of fission neutrons using the VDC logic the neutron coincidence rate is defined by the following expression

$$F = N_\tau/(1-\tau N_\tau)-N_T/(1-TN_T), \qquad (1)$$

where N_τ and N_T are the count rates of neutron detector pulses in the fast counting channel (by fast scaler) with small dead time τ and in the slow channel (by slow scaler) with preset dead time T, correspondingly. In accordance with the terminology of the neutron coincidence techniques this quantity can be defined as a real coincidence count rate (reals).

Let us consider the dependence of the observed reals on the characteristics of counting system. Let us assume that Q neutrons of the background and (α,n)-reaction and $f\sum_m mB(m)$ fission neutrons are detected per time unit when there is no dead time counting losses. Where f is fission rate, B(m) is probability for detection of m neutrons per fission event. The count rate of fission neutrons can be presented as two components: $f\sum_m B(m)$ and $f\sum_m (m-1)B(m)$.

Let us believe that in case when a fission event is followed by detection of several neutrons the first of them will be referred to the first component and all the rest to the second one. As spontaneous fissions as well as (α,n)-reactions represent the Poisson´s event flow, the dead-time losses in the slow counting channel are defined as

$$L = [f\sum_m B(m)+Q]TN_T+[f\sum_m (m-1)B(m)]\, g(T),$$

where g(T) - probability of losses for the second component of the fission neutron count rate. From the evident relations

$$N_T+L = Q+f\sum_m mB(m) = N_\tau/(1-\tau N_\tau),$$

there follows

$$N_\tau/(1-\tau N_\tau)-N_T/(1-TN_T) = Gf\sum_m (m-1)B(m), \qquad (2)$$

where $G = [g(T)-TN_T]/(1-TN_T)$.

The probability to detect m neutrons per fission event is defined as

$$B(m) = \sum_v q(v)C^m_v \epsilon^m (1-\epsilon)^{v-m},$$

where q(v) - probability that v neutrons are emitted per fission event,
C^m_v - binomial coefficient m over v,
ϵ - detector efficiency.

Using an expansion of the $(1-\epsilon)^{v-m}$ complex in a power series in ϵ one can obtain

$$\sum_m (m-1)B(m)=\sum q(v)[v(v-1)\epsilon^2/2!-v(v-1)(v-2)\epsilon^3/3!+..]$$

It is suitable to express the terms of the obtained series via the factorial moments of v. As a result of this expression (2) for reals is transformed to

$$F = fG \sum_{j=2} [(-1)^j W_j \epsilon^j/j!], \qquad (3)$$

where W_j is the j-th factorial moment of fission neutron multiplicity distribution.

An assumption that not more then one neutron from the fission event is counted in slow scaler is the basis of the VDC-technuque. This assumption is valid for condition g(T)=1, which is satisfied if the lifetime of fission neutrons in the detector does not exceed the preset dead time T. In this case a factor G=1 and reals are proportional to the fission rate.

In the detecting system formed from the thermal neutron counters embedded into a moderator the probability for a neutron to survive after a fission event to time t is proportional to $\exp(-t/\beta)$, where β is the detector die-away time. The probability to count more than one neutron per fission event in slow scaler can be neglected if preset dead time $T \gg \beta$. Implementation of this condition in practice is usually unsuccessful due to a sharp decrease of permissible count rates with dead time increasing. It leads to a disturbance of proportionality between observed reals and fission rate and causes a methodical error of measurements.

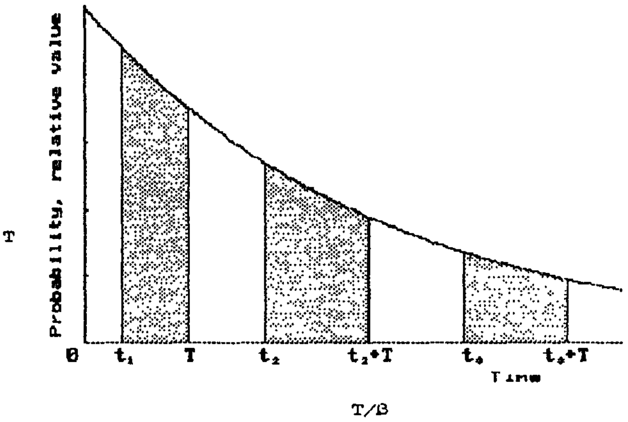

Fig. 1. Probability of detecting within dead-time intervals for neutrons of the same fission event.

For estimation of factor G let us consider the probability g(T) for pulses caused by fission neutrons to get into the dead time intervals. Let us set the beginning of the time scale at the beginning of one of such intervals

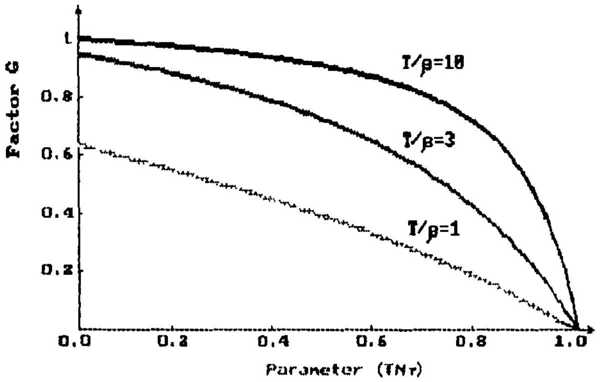

Fig. 2. The correcting factor G as a function of (TN_T) and (T/β) parameters.

and denote the initial moments for the next intervals as: t_2 - for the second, t_3 - for the third and so on (Fig.1). Let the first neutron of a fission be detected at the time moment t_1. The detection probability for the rest neutrons of this fission is approximately described in time by a function $U(t)=(1/\beta)\exp[-(t-t_1)/\beta]$. The probability for detection moments to get into the dead time intervals is

$$V(t_1) = \int_{t_1}^{T} U(t)dt + \sum_{i=2} \int_{t_i}^{t_i+T} U(t)dt = 1-D\exp(t_1/\beta),$$

where $D = e^{-T/\beta} - (1-e^{-T/\beta}) \sum_{i=2} e^{-t_i/\beta}$

From the analysis of the time interval distribution between the pulses counted in the slow scaler it follows that the values for items of the last sum are represented by the terms of the decreasing geometrical progression. Its sum is determined by the expression

$$\sum_i e^{-t_i/\beta} = \beta Z e^{-T/\beta} [1+\beta Z(1-e^{-T/\beta})],$$

where $Z = N_T/(1-TN_T)$.

The first detected fission neutron with probability $(1-TN_T)$ is counted in the slow scaler. In this case the detection moment coincides with the beginning of the time scale and then $V(t_1=0) = 1-D$. If the moment t_1 is within the interval (0,T), then the average probability

$$\bar{V}(t_1=0) = (1/T)\int_{0} V(T_1)dt_1 = 1-D\beta(e^{T/\beta} - 1)/T.$$

Thus, the probability g(T) can be presented as

$$g(T) = (1-TN_T)(1-D) + TN_T[1-D\beta(e - 1)T].$$

After substitution of g(T) the expression for factor G takes the form of

$$G = (1-e^{-T/\beta})/[1+\beta Z(1-e^{-T/\beta})]. \qquad (4)$$

Fig. 2 shows a dependence of G on the (TN_T) parameter at different values of (T/β)-ratio. We would note that the obtained expression for factor G seems to be preferable in comparison with the correction given in ref. [1] which is valid only in the range of low (TN_T) values and with increase it transfers into the negative values region.

3. Implementation of the method

The experimental examination of the VDC-technique was performed at the facility designed for measurement of plutonium in the mixed dioxide fuel samples. Four identical detector units situated around a vessel for sample location were used for neutron detection. Each of these units comprises five ^{3}He-counter tubes embedded into moderator. The pulse signals from the detector units are mixed and come to the digital one-shot multivibrator. Its dead time can be set from the value sequence of 8, 16, 32 and so on to 512 µs. The output pulses from the mixer and one-shot multivibrator are counted in the corresponding scalers. The data from scalers are periodically transferred to the computer memory for proper processing.

The dead time of neutron counting in the fast channel (from mixer output) was determined by the known two-sources method. Its value is 0.43 μs. Actual values of the preset dead time in the slow counting channel were defined more precisely by direct measurement of the duration of pulses from the one-shot multivibrator with an accuracy of about 0.02 μs. The efficiency of the detection system for fission neutrons is about 18%.

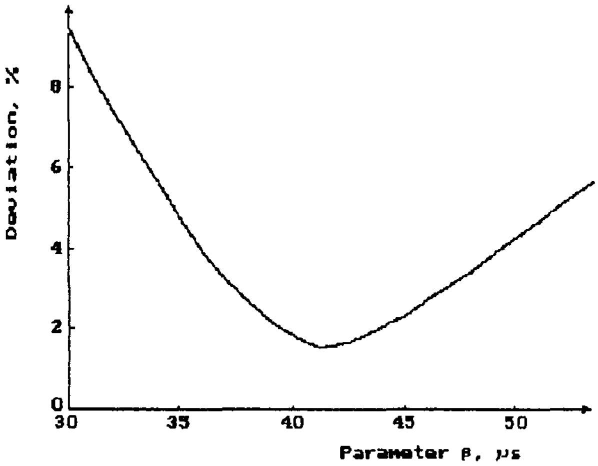

Fig. 3. Standard deviation of measurement results after correction for methodical error as a function of β value.

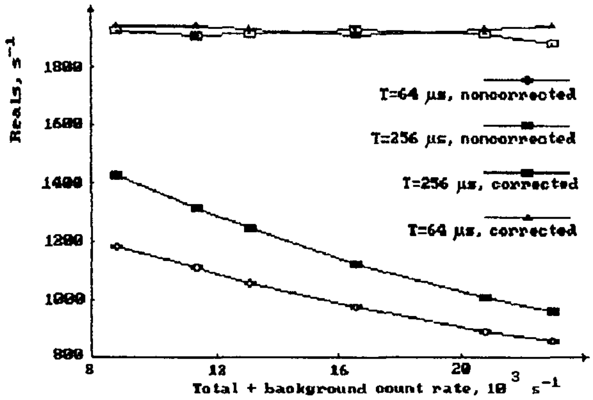

Fig. 4. Dependence of reals on background count rate before and after correction for methodical error.

A set of ^{252}Cf fission neutron sources and Pu-Be (α,n)-sources have been used in measurements. The proportionality of the observed response to fission rate and its independence of (α,n)-neutron count rate are the obvious requirements that the method has to meet. The reals defined in accordance with expression (1) satisfy these requirements only in the region of low counting rates (up to about 10^3 s^{-1}). Expression (4) reveals the methodical error of measurements that rises with count rate increase.

In order to estimate the correction factor G one should know the value of the parameter β. Proceeding from the functional sense of correction, the optimum value is β that minimizes the variation of corrected reals $F_0=F/G$ at different values of the preset dead time and the background count rate. Fig. 3 shows the relative standard deviation of corrected reals as a function of β value in measurements with ^{252}Cf neutron source when the preset dead time was changed from 32 to 256 μs and (α,n)-neutron sources were used for generation of background count rates up to about $2\cdot10^4 s^{-1}$.

The reals change before and after correction depending on background
count rate is shown in Fig. 4.

4. Multiplication effect correction

Correction for induced fission caused by primary neutrons presents the main difficulty in the interpretation of measurement results. Estimation of the induced fission effect on reals is connected with definition of factorial moments for neutron multiplicity distribution taking into account the neutron multiplication. The solution of this problem is described in ref./2/, based on the use of probability generating functions for the number of neutrons emitted in fission events.

In a single-group energy assumption the factorial moments in expression (3) are defined via the factorial moments for spontaneous and induced fissions by the following relations:

$$W_2 = M^2 Z_1, \qquad (5)$$

$$W_3 = M^3(Z_2+3XW_{2in}Z_1),$$

$$W_4 = M^4[Z_3+6XW_{2in}Z_2+XZ_1(4W_{3in}+15W^2_{2in})]$$

etc.,

where $Z_j = W_{(j+1)sp}+X(1+\alpha)W_{1sp}W_{(j+1)in}$,

$X = (M-1)/(W_{1in}-1)$,

M - neutron multiplication factor,

α - ratio of the (α,n)-neutron to spontaneous neutron yields,

W_{jsp} and W_{jin} - the j-th factorial moments of neutron multiplicity distribution for spontaneous and induced fissions correspondingly.

When M values are close to 1 simplification of awkward expressions for factorial moments of a high order is possible. Their analysis shows that if the terms with X parameter to the power of 2 and higher are neglected we obtain:

$$W_j = M^j[W_{jsp}+\alpha XW_{1sp}W_{jin}+X\sum_{k=2}^{j} C^k_j W_{kin}W_{(j-k+1)sp}] \qquad (6)$$

for all j>=2.

The known formalism /3/ is usually applied in practice for multiplication correction in measurements by the shift register coincidence technique. The ratio of reals to the total count rate serves as an informative parameter of multiplication. A similar approach was used for correction of measurement results by the VDC technique.

The correction factor K for reals can be presented in the following form:

$$K = F_0/F_{cor} = F_0(1+\alpha)M/(rN_0), \qquad (7)$$

where $F_0=F/G$ - the reals value corrected for the methodical error of measuring,

$N_0=N_T/(1-\tau N_T)$ - the total count rate corrected for the dead time losses,

F_{cor} - the unknown value of reals

corrected for neutron multiplication,

r - an auxiliary parameter that represents the ratio of reals to total count rate in the absence of multiplication at $\alpha=0$ and $G=1$.

Using expression (6) obtained for W_j, a dependence of the ratio F_0/F_{cor} on the multiplication factor M was analyzed by calculation modeling performed for different fissile nuclides. The analysis shows that in the limited range of efficiency values at M values approaching 1 an approximation of the multiplication factor M can be obtained as a solution of the following quadratic equation

$$A(M-1)^2 + B(M-1) + C = 0, \qquad (8)$$

where

$$A = [(1+\alpha)/\bar{v}_{sp}-r(2-r)/(\bar{v}_{sp}-1)]\bar{v}_{in}^2/(\bar{v}_{in}-1),$$

$$B = 1-r+[(1+\alpha)/\bar{v}_{sp}-r/(\bar{v}_{sp}-1)]\bar{v}_{in}^2/(\bar{v}_{in}-1),$$

$$C = 1-F_0(1+\alpha)/(rN_0),$$

$\bar{v}_{sp}=W_{1sp}$ and $\bar{v}_{in}=W_{1in}$ are the average numbers of neutrons emitted in the spontaneous and induced fission acts, correspondingly.

Numerical calculation has been performed in order to estimate an error of M definition in this way. The reals calculation was done according to expression(3) at preset ϵ, α and M values with determination of factorial moments (5) to the sixth inclusive. The calculated reals and totals values were further applied for estimation of the multiplication factor from equation (8).

At the efficiency values up to 20% the discrepancy between the obtained and input values of M ranging from 1 to 1.1 did not exceed 0.01.

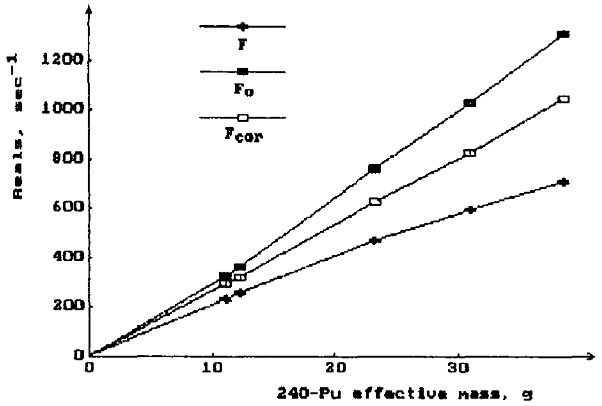

Fig. 5. Dependence of coincidence count rate on ^{240}Pu effective mass before and after correction for methodical error and multiplication.

Fig.5 shows the measurement results for the samples before and after multiplication correction as a function of ^{240}Pu effective mass (m_{ef})

The proposed algorithm was used for multiplication correction in measurements of fuel samples containing up to 100 g of plutonium. A set of five samples with the known data on mass and plutonium isotopic composition was applied for calibration of the detector. Two of them contain about 50 g of PuO_2 powder with different isotopic composition; three others contain 300 g, 400 g and 500 g of electrodeposited mixture of plutonium and natural uranium dioxides (25% of PuO_2).

calculated from plutonium isotopic data. It can be seen that the measured reals F and the reals F_0 corrected for methodical error have a nonlinear dependences on m_{ef}. After multiplication correction a calibration function $F_{cor}(m_{ef})$ for detector response is a straight line.

5. Conclusions

In summary, it should be noted that in spite of the simplicity of the VDC logic the interpretation of measurement results by this technique is rather complicated. With increase of the detector count rate the methodical error of measurements rises that distorts the correlation of detector response to fission rate. The proposed correction for the methodical error allows the proportional correlation to be ensured in the range of count rate up to about $3\cdot10^4$ s^{-1}. The multiplication correction can be implemented to obtain a linear calibration function for the detector response.

However, the VDC-technique demonstrates its value for assay of small amounts of spontaneously fissioning materials only.

6. References

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NON-DESTRUCTIVE ISOTOPIC URANIUM ASSAY BY MULTIPLE DELAYED NEUTRON MEASUREMENTS

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Abstract

The high accuracy and precision required in nuclear safeguards measurements can be achieved by an improved neutron activation technique based on multiple delayed fission neutron counting under various experimental conditions. For the necessary ultrahigh counting statistics required, cyclic activation of multiple subsamples has been applied. The home-made automated flexible analytical system with neutron flux and spectrum differentiation by irradiation position adjustment and cadmium screening, permits the non-destructive determination of the U-235 abundance and the total U element concentration needed in nuclear safeguards sample analysis, with a high throughput and a low operational cost. Careful experimental optimization led to considerable improvement of the results.

1. Introduction

Several more or less sophisticated methods have been developed for nuclear safeguards and reference material analysis in order to meet the high specifications required in this type of measurements. The conventional delayed fission neutron technique has been applied successfully with sufficient accuracy and precision to the determination of natural uranium not only in geological but also in nuclear safeguards and reference material samples covering a wide concentration range, by calibrating the analytical system with certified natural uranium standards (1-3).

Since however in nuclear safeguards and reference material applications not only the uranium element concentration but also the U-235 isotopic abundance is required, multiple measurements under different experimental conditions were necessary in order to determine the two unknown values as well as the correction factors due to neutron flux depression and other interferences.

2. Method description

The method is based on reactor neutron activation of uranium or other fissile material samples of various isotope abundances and the measurement of the delayed neutrons from the fission products. The uranium element concentration and the U-235 isotope abundance were determined by performing two measurements differentiating the neutron flux spectrum and the irradiation position at the nuclear reactor. The neutron flux spectrum was modified by cadmium screening resulting in thermal neutron flux depression. Initially separate closed or partially open small cadmium containers around each sample were used for variable neutron flux spectrum differentiation. This however necessitated the preparation of two samples, one bare and one cadmium covered, besides the difficulty of proper cadmium container preparation and the probability of capsule damage at the irradiation and counting position stops of the capsule transfer system, with subsequent sample leakage, resulting in possible pneumatic transfer tube contamination. Therefore a permanent cadmium sleeve was installed around one of the two pneumatic tube irradiation terminals. The flexible terminals were positioned at different distances from the reactor core of the 5 MW swimming pool reactor of N.C.S.R. Demokritos, to compensate for the difference between the thermal and epithermal neutron flux and the fission cross-section of the uranium isotopes U-235 and U-238, in order to optimize the counting rates during the measurements (4-6).

Initially the simple uranium fission balance expression was used:

$$\alpha U^5+\beta U^8 = C \qquad (1)$$

$$\text{or } \alpha U^5+\beta(U-U^5) = C \qquad (2)$$

$$\text{or } (\alpha-\beta)U^5+\beta U = C \qquad (3)$$

$$\text{or } BU^\alpha+\Gamma = \frac{C}{U} \qquad (4)$$

where

U^5 = U-235 concentration

U^8 = U-238 concentration

U = total U element concentration

$$U^\alpha = \frac{U^5}{U} = \text{U-235 abundance} \qquad (5)$$

C = neutron counting rate per mass unit

B,Γ =coefficients

Applying equ.(4) for the bare and cadmium covered cases respectively, the following system is obtained

$$\begin{aligned} BU^\alpha+\Gamma &= \frac{C}{U} \\ B'U^\alpha+\Gamma' &= \frac{C'}{U} \end{aligned} \qquad (6)$$

where the coefficients B,Γ,B',Γ' can be determined by using two standards of different element concentration U_1,U_2 and isotope abundance U_1^α,U_2^α, from which

$$B = \frac{\frac{C_1}{U_1} - \frac{C_2}{U_2}}{U_1^\alpha-U_2^\alpha} \qquad (7)$$

$$\Gamma = \frac{C_1}{U_1} - BU_1^\alpha \qquad (8)$$

$$U^\alpha = \frac{\Gamma' C - \Gamma C'}{BC' - B'C} \qquad (9)$$

$$U = \frac{BC' - B'C}{B\Gamma' - B'\Gamma} \qquad (10)$$

However if the thermal neutron flux depression is taken into account, which can be approximated by the percentage expression

$$d = 1 - \delta U^\alpha , \qquad (11)$$

then equ. (4) becomes

$$BU^\alpha(1-\delta U^\alpha)+\Gamma = \frac{C}{U} \qquad (12)$$

or $$AU^{\alpha^2}+BU^\alpha+\Gamma = \frac{C}{U} \qquad (13)$$

and for the cadmium covered case:

$$A'U^{\alpha^2}+B'U^\alpha+\Gamma' = \frac{C'}{U} \qquad (14)$$

The coefficients A,B,Γ,A',B',Γ' can be determined by using three standards of different element concentration U_1,U_2,U_3 and isotope abundance $U_1^\alpha,U_2^\alpha,U_3^\alpha$, and thus from equ. (13)

$$A = \frac{\dfrac{\frac{C_1}{U_1} - \frac{C_2}{U_2}}{U_1^\alpha - U_2^\alpha} - \dfrac{\frac{C_2}{U_2} - \frac{C_3}{U_3}}{U_2^\alpha - U_3^\alpha}}{U_1^\alpha - U_3^\alpha} \qquad (15)$$

$$B = \frac{\frac{C_1}{U_1} - \frac{C_2}{U_2}}{U_1^\alpha - U_2^\alpha} - A(U_1^\alpha+U_2^\alpha) \qquad (16)$$

$$\Gamma = \frac{C_1}{U_1} - BU_1^\alpha - AU_1^{\alpha^2} \qquad (17)$$

For A',B',Γ' similar equations hold. The system of equ. (13),(14) can also be solved in the normal matrix way for A,B,Γ,A',B',Γ'.

Then from equ. (13),(14)

$$(A - \frac{C}{C'}A')U^{\alpha^2}+(B - \frac{C}{C'}B')U^\alpha+(\Gamma - \frac{C}{C'}\Gamma') = 0 \qquad (18)$$

or $$A''U^{\alpha^2}+B''U^\alpha+\Gamma'' = 0 \qquad (19)$$

from which

$$U^\alpha = \frac{-B''+\sqrt{B''^2-4A''\Gamma''}}{2A''} \qquad (20)$$

and from (13)

$$U = \frac{C}{AU^{\alpha^2}+BU^\alpha+\Gamma} \qquad (21)$$

In order to improve the counting statistics, because of the short-lived nature of the delayed neutron emitting fission products, cyclic activation has been applied combined with timed intermediate sample storage to avoid radiation build-up and dead-time losses for counting rate optimization. Cyclic activation reduces also timing and sample position uncertainties during irradiation.

A programmable logic controller is being installed for automatic system operation while a special compute software program facilitates data evaluation.

5. Results

Intercomparisons of nuclear safeguards uranium samples from the Safeguards Analytical Laboratory of the International Atomic Energy Commission gave the following results.

Using two standards with

$U_1 = 84.76\%$ $U_1^\alpha = 2.38\%$, $U_2 = 87.36\%$, $U_2^\alpha = 0.25\%$

the following counting rates were obtained for the standards and the unknown sample x, with the cadmium covered terminal closer to the reactor core than the bare one:

C_1=9433 c/mg, C_2=1097 c/mg, C_x=2301 c/mg

C_1'=5865 c/mg, C_2'=1955 c/mg, C_x'=2193 c/mg

Inserting these values into equ. (7) and (8) we got

$B = 46.34$, $\Gamma = 1$, $B' = 21.97$, $\Gamma' = 16.89$

and from equ. (9) and (10):

$U_x^\alpha = 0.718\%$, $U_x = 67.14\%$

Using three standards with

$U_1 = 84.76\%$ $U_1^\alpha = 2.38\%$

$U_2 = 86.79\%$ $U_2^\alpha = 0.711\%$

$U_3 = 87.36\%$ $U_3^\alpha = 0.25\%$

we got

C_1 = 9433 c/mg, C_2 = 2919 c/mg, C_3 = 1097 c/mg

C'_1 = 5865 c/mg, C'_2 = 2792 c/mg, C'_3 = 1977 c/mg

C_x = 2301 c/mg, C'_x = 2193 c/mg

and from equ. (15) to (17)

$A = 0.3917$, $B = 45.32$, $\Gamma = 1.2187$

$A' = 0.4545$, $B' = 20.784$, $\Gamma' = 17.16$

and from equ. (18) to (21):

$U_x^\alpha = 0.7157\%$, $U_x = 67.957\%$

which compare quite well with the true values

$U_x^\alpha = 0.711\%$, $U_x = 68.306\%$

6. Discussion

Various combinations of bare and cadmium covered samples have been tried to optimize the experimental conditions. Thus bare samples at the bare tube and up and down cadmium covered samples at the cadmium covered tube gave the following similar results for various standard combinations:

$U_{x_1} = 66.78\%$, $U_{x_2} = 66.78\%$, $U_{x_3} = 66.83\%$

$U_{x_1}^\alpha = 0.7184\%$, $U_{x_2}^\alpha = 0.7185\%$, $U_{x_3}^\alpha = 0.7179\%$

with a small descrepancy of the total uranium element concentration compared to the true value

$U_x = 68.306\%$, $U_x^\alpha = 0.711\%$.

Another measurement gave:

$U_{x_1} = 68.87\%$, $U_{x_2} = 68.86\%$, $U_{x_3} = 66.49\%$

$U_{x_1}^\alpha = 0.6958\%$, $U_{x_2}^\alpha = 0.6958\%$, $U_{x_3}^\alpha = 0.7240\%$.

Approaching the tube terminals closer to the reactor core for counting statistics improvement and trying various bare(B) and cadmium(C) covered sample(S) and tube(T) combinations at different distances(DD) from the reactor core, the following results were obtained:

BS in BT, BS in CT: $U = 69.19\%$, $U^\alpha = 0.7098\%$

BS in BT at DD: $U = 68.96\%$, $U^\alpha = 0.6982\%$

BS in CT, CS in BT: $U = 68.59\%$, $U^\alpha = 0.7033\%$

BS in BT, CS in CT: $U = 69.03\%$, $U^\alpha = 0.7015\%$

compared to the IAEA values: $U = 68.306\%$, $U^\alpha = 0.711\%$

7. Conclusion

It can be concluded that most results are quite close to the true values giving an error of less than 1%, with poor counting statistics. It is expected that careful sample preparation, repeatable experimental conditions and mainly high counting statistics will improve the results considerably.

8. Acknowledgments

The authors would like to thank S. Deron and his staff of IAEA-SAL for providing the intercomparison samples and for valuable discussions, as well as the colleagues C. Perisinakis, C. Spyrou, G. Lefkopoulos, C. Gatsos and the Reactor and Machine Shop staff for their valuable contribution to the work.

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CALIBRATION OF THE NEUTRON COINCIDENCE COLLAR FOR THE ASSAY OF FUEL ASSEMBLIES CONTAINING THERMAL NEUTRON POISONS

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Abstract

The paper describes the calibration of the neutron coincidence collar using fresh, low enriched uranium oxide and mixed (U/Pu) oxide mock-up Light Water Reactor (LWR) type fuel assemblies. The paper also summarises studies on the sensitivity of the neutron coincidence collar to measurements on LWR fuel assemblies after the removal or substitution of fuel pins and the addition of various types of neutron poisons. Finally monte carlo calculations used to simulate some of the observed experimental results are discussed along with a relationship for applying the results to full scale production LWR fuel assemblies.

1. Introduction.

The neutron coincidence collar (NCC) is a non-destructive assay (NDA) instrument designed by the Los Alamos National Laboratory (LANL), Los Alamos, USA for either the measurement of the ^{235}U or the ^{240}Pu eff. mass loading (g/cm) of fresh LWR fuel assemblies [1,2,3]. The instrument is a member of the high-level neutron coincidence counter (HLNCC) family [4]. An additional problem in the assay of LWR fuel assemblies is the presence of thermal neutron poisons in the fuel assembly. Poisons may be present in various forms in a fuel assembly. A detailed study has already been performed by the LANL and the Euratom Safeguards Directorate into improving the calibration of the NCC for the measurement of low enriched uranium (LEU) type LWR fuel assemblies containing burnable thermal neutron poisons such as Gd_2O_3 [5]. In pressurized water reactors, for example the type designed by Fragema/Framatom, various poison materials may also be present in the form of clusters of rods which are completely separate from the fuel pins and are inserted into the open guide channels of the fuel assembly as opposed to the burnable poisons such as Gd_2O_3 which are homogeneously mixed with the LEU fuel material. The types of materials typically used are stainless steel, alloys of silver indium and cadmium, boron carbide and silicon oxides. Measurements have been performed by Euratom NCC detector systems using LWR reference mock-up fuel assemblies of fissile material originating and present outside and within the European Community. This paper concentrates on studies performed for these second type of poisons for both LEU and MOX type LWR fuel assemblies. The paper also reports on the sensitivity of the NCC to the assay of fresh LWR MOX type fuel assemblies for various diversion scenarios by the removal or substitution of fuel pins and goes on to demonstrate the practical application of MCNP [6] techniques to supplement the experimental measurements and the creation of correction factors for establishing the relationship between the mock-up and full size LWR MOX fuel assemblies.

2. Materials and Methods

2.1. Measurement Technique.

NCC for LEU LWR fuel

The NCC is configured as an active interrogation device equipped with an Am-Li (^{241}Am-^{7}Li) source and an array of 18 ^{3}He thermal neutron detectors embedded in high density polyethylene. The 18 detector tubes are positioned in sets of 6 in 3 panels of a rectangular arrangement with the 4th panel being used to hold the interrogation source. The polyethylene panels provide for the moderation of the source neutrons, the moderation of the induced fission neutrons deriving from the fissile content of the fuel and the structural support.

The assay technique is essentially a two step measurement operation. The measurement response of the NCC is a net coincidence (Reals) count rate $Ri_{(net)}$, being the result of two separate measurements on the same fuel assembly.

The first measurement is the passive measurement (without interrogation source) and the second the active measurement (with interrogation source inserted in the 4th panel). The net Reals count rate is thus,

$$Ri_{(net)} = Ri_{(active)} - Ri_{(passive)}$$

The best calibration algorithm which has been shown to represent the measurement technique is,

$$K.Ri_{(net)} = \frac{aM}{1 + bM}$$

where $M = {}^{235}U$ (g/cm) and
a and b are the calibration coefficients for the type of fuel assembly and K is a combination of correction factors which account for various differences between the measurement conditions at the time of the calibration and the application to field measurement conditions [5].

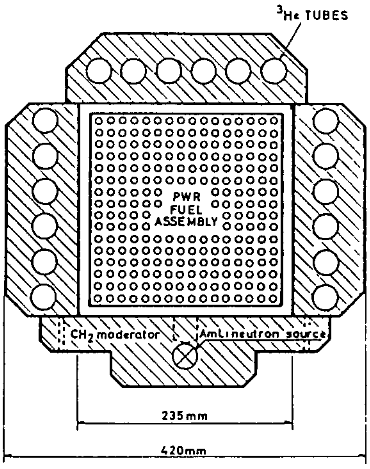

Fig.1.

K = K0 * K1 * K2 * K3 * K4 * K5 and

K0 represents changes in the AmLi source strength
K1 accounts for possible electronic drift
K2 corrects for detector efficiency
K3 corrects for thermal neutron poisons.
K4 corrects for differences in the heavy metal loading
K5 can account for other small effects such as extra assembly wrapping (plastic, cardboard, etc.)

NCC for LWR MOX type fuel.

The NCC used for the measurement of fresh LWR MOX fuel assemblies is similar to that used for the LEU type fuel except that the 4th panel which holds the interrogation source is replaced by another polyethylene panel with 6 ^{3}He tubes. The application of the NCC in this configuration is analogous with the HLNCC-II for the measurement of any plutonium based item. The NCC has previously been demonstrated for the assay of Fast Reactor MOX sub-assemblies [7,8]. For the assay of LWR MOX fuel assemblies the cadmium liners are not used due to dimensional restrictions. The basic equations used for data reduction and evaluation have been described elsewhere but are summarised below:

$$R_{mc} = a.{}^{240}Pu_{eff}.(g/cm) \quad \text{where}$$

$${}^{240}Pu_{eff.} = 2.49.f_{238} + f_{240} + 1.57.f_{242}$$

and f_i is the weight percent fraction of the appropriate plutonium isotope.

R_{mc} is the multiplication corrected Reals counting rate and is calculated using the following expressions:

$$R_{mc} = R/CF \qquad CF = M.r$$

$$r = [(R/T)/p_o].(1+\alpha)$$

$$p_o = (R/T)_o.(1+\alpha) = 0.0814$$

$$M = [-B+(B^2-4AC)^{1/2}]/2A$$

$$A = 2.074(1+\alpha)$$

$$B = -(2.074\alpha+1.0174) \qquad C= -r$$

$$\alpha = \frac{(13400f_{238}+38.1f_{239}+141f_{240}+1.3f_{241}+2f_{242}+2690f_{241Am})}{1020(2.54f_{238}+f_{240}+1.69f_{242})}$$

where p_o is for a non multiplying sample and α is the ratio of (α, n) neutrons to spontaneous fission neutrons.

2.2. Description of Reference Material.

Various types of fuel pins were available for creation of the LWR mock-up fuel assemblies. The fuel pins are all of the same length with small variations in the diameter of the fuel pellets and thickness of the cladding. The fuel cladding is a stainless steel alloy for all of the reference materials except for the ^{235}U (3.3%) fuel which is zircalloy. The 3.3% material was only used for the experiments concerning the LEU calibration whereas all the other reference materials were used in the LWR MOX experiments. The characteristics of the VENUS LWR mock-up fuel are given below:

Characteristics of the VENUS LWR mock-up fuel

TYPE		LEU		MOX		
		3.3/0	4.0/0	2/2.7	1.7/3.2	0.7/5.1
Chemical Composition (w/w)%	UO_2	100	100	97.3	96.82	94.95
	PuO_2	-	-	2.7	3.18	5.05
Isotopic Composition (15.11.1966)						
(w/w)%	^{234}U	0.029	0.022	0.016	0.011	-
	^{235}U	3.311	3.971	2.002	1.785	0.72
	^{236}U	0.017	0.030	0.013	0.010	-
	^{238}U	96.644	95.977	97.970	98.194	99.28
	^{238}Pu	-	-	0.068	Idem	Idem
	^{239}Pu	-	-	79.247	..	..
	^{240}Pu	-	-	17.197	..	..
	^{241}Pu	-	-	3.044	..	..
	^{242}Pu	-	-	0.445	..	..
	^{241}Am	-	-	0.075	..	..
Mass loading (g/cm)	Pu_{tot}	-	-	0.1424	0.1795	0.2881
	^{235}U	0.1570	0.2234	0.1026	0.0976	0.0390
Fuel diameter (cm)		0.819	0.890	0.902	0.897	0.896
length (cm)		50.0	50.0	50.0	50.0	50.0
cladding type		Zr-4	SS304	SS304	SS304	SS304
diameter (cm)		0.950	0.978	0.978	0.978	0.978
thickness (cm)		0.057	0.038	0.038	0.038	0.038

2.3. Poison Materials.

Four types of rods which are typically used with LWR reactor fuel assemblies for the purpose of criticality control and fine adjustment to the operation of the reactor have been used for the experiments. The rods used have similar dimensions to the mock-up fuel and their characteristics are given below:

Type	Pyrex	AgInCd	Inox	B_4C
form	(SiO_2/B_2O_2) pipe	%(80/15/5) solid alloy	SS304 solid	compressed compound
diameter (cm)	0.905 (ext) 0.605 (int)	0.903	0.978	0.902
length (cm)	50	50	50	50
cladding	type all stainless steel SS304			
diameter	0.978	0.978	-	0.978
thickness	0.038	0.038	-	0.038

2.4. Experimental set-up.

The VENUS mock-up assembly array is designed to simulate as close as possible a typical 17 by 17 LWR fuel assembly structure. The array consists of 264 positions for insertion of fuel rods and 25 open positions. The central position is always left open but the remaining 24 open positions are available for insertion of the various rods of thermal neutron poison materials. The external dimensions of the grid are 21.42 cm by 21.42 cm and the fuel pin spacing is 1.26 cm. The fuel pins are slightly more densely pact together than is the case for full size fuel assemblies. The NCC was placed around the mock-up so that the fuel stack was centrally positioned relative to the axial linear counting response of the detector during all experiments. The detectors were supported by a metal and thin wooden plate for that purpose. Euratom detector systems EUR 3 and EUR 5 were used for the studies. The performance of both the instruments were monitored regularly in order to be able to compare and supplement calibration work carried out by them at the LANL, USA [5].

2.5. Case Studies.

2.5.1. Addition of clusters.

Tests were performed to investigate the influence of adding various number and combinations of the different poison material rods to the measured counting response. The combinations chosen followed as closely as possible typical loading deployed in full scale fuel assemblies. Due to restricted availability of the fuel pins and time for performing the experiments not all possible combinations were fully explored. The studies were however selective enough to be able to optimise the results required to draw conclusions from the tests. Clusters of poison rods were introduced to both a LEU mock-up (264 fuel pins at 3.3 % ^{235}U) and a MOX mock-up (264 fuel pins 2/2.7 % $^{235}U/Pu$). Detector EUR 3 was used for the experiments on the LEU mock-up and detector EUR 5 was used for the experiments on the MOX mock-up. A sectional view of the mock-up assembly is given below.

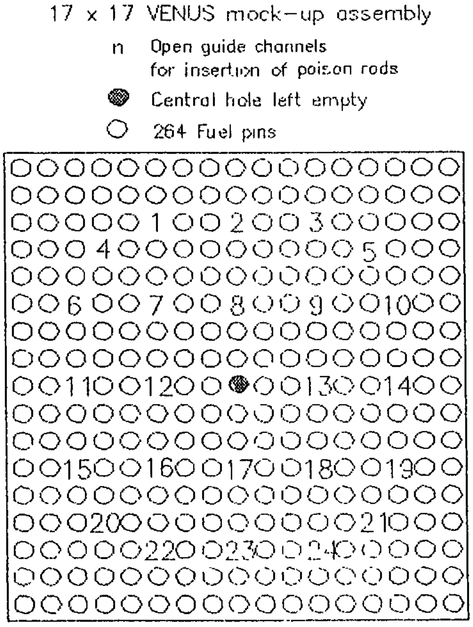

Fig.2.

The numbers associated with the open guide holes are shown for reference to indicate the positions of the poison rods for the various cluster combinations. For the measurements using the AmLi source the face with the guide holes 6,11 and 15 was always closest to the source panel of the NCC. For measurements on MOX the face with guide holes 1,2,3 was always closest to the door panel of the NCC. The combinations tested were as follows:

Low Enriched Uranium mock-up.

Material Type	Number of rods	Positions in cluster
AgInCd	4	4,5,20,21
	8	1,3,6,10,15,19,22,24
	12	1,3,6,8,10,12,13,15,17,19,22,24
	16	1,3,4,5,6,8,10,12,13,15,17,19,20,21,22,24
	20	1 to 24 less 7,9,16,18
Inox	8	1,3,6,10,15,19,22,24
	16	1,3,4,5,6,8,10,12,13,15,17,19,20,21,22,24
	24	1 to 24
Pyrex	3	5,8,13
	8	1,3,6,10,15,19,22,24
	9	1,2,3,4,6,8,11,12,15
	9	1,2,3,5,8,10,13,14,19
	12	1,3,4,5,6,10,15,19,20,21,23,24
	16	1,3,4,5,6,8,10,12,13,15,17,19,20,21,22,24
Mix 1		
AgInCd	8	2,4,5,11,14,20,21,23
Inox	16	1,3,6-10,12,13,15-19,22,24
Mix 2		
AgInCd	8	1,3,6,10,15,19,22,24
Inox	8	4,5,8,12,13,17,20,21
Pyrex	8	2,7,9,11,14,16,18,23

Mixed oxide fuel mock-up.

Material Types	Number of Rods	Positions in cluster
[B_4C	4	4,5,20,21
AgInCd	8	2,4,5,11,14,20,21,23
Inox	12	1,3,4,5,6,10,15,19,20,21,22,24
Pyrex]	16	1,3,4,5,6,8,10,12,13,15,17,19,20,21,22,24
[B_4C	4	8,12,13,17
Inox		
Pyrex]		
[AgInCd	19	1,3,4,5,6-10,12,13,15,16,18,19,20-22,24
Inox]		
[B_4C	20	1,3,4-10,12,13,15-22,24
Pyrex]		
Pyrex	24	1-24
	3	13,17,21
	9	1-3,5,8,10,13,14,19
Inox	1	8
Mix 1		
Inox	16	1,3,6-10,12,13,15-19,22,24
AgInCd	8	2,4,5,11,14,20,21,23

2.5.2. Fuel pin removal and substitution.

The fabrication of LWR MOX type fuel assemblies is increasing more and more as a result of fuel cycle management policies within the European Community. From a nuclear safeguards stand point the measurement of this new generation of LWR fuel assembly is very important considering a typical assembly contains around 22 Kg of plutonium. Tests have been performed to investigate the sensitivity of the NCC measurement technique to scenarios of diversion of material by either removal of fuel pins, the substitution of fuel pins with Inox rods which are normally used in the clusters mentioned previously and the substitution of MOX type fuel pins with LEU type fuel pins in an attempt to maintain the heavy metal loading of the assembly constant. In addition the removal and substitution of fuel pins with Inox rods was carried out at in areas corresponding to an inner and outer zone of the fuel assembly. The number of pins ranged between 2 and 20. The 25 open guide holes were free of any other neutron poison rods for the purpose of the tests thus simulating a standard LWR MOX fuel assembly with 264 fuel pins of 2/2.7 % U/Pu type fuel. The locations of the zones where fuel pins were either removed or substituted are indicated in the following diagram.

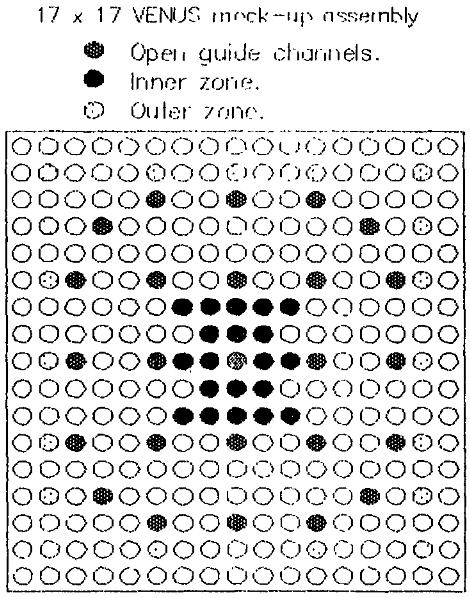

Fig.3.

2.5.3. MOX Calibration.

Combinations of the various types of reference MOX fuel pins were selected in order to create several mass loading for the 264 fuel pin mock-up. The 25 guide holes were always left open without insertion of thermal neutron poison rods. Five combinations were configured by replacing up to 120 of the 2/2.7 % type fuel pins with up to 64 of the 1.7/3.2 % and 56 of the 0.7/5.1 % type fuel pins. The range in the Pu mass loading was 38.81 to 47.12 (g/cm). The measurement data accumulated during some of the experiments for fuel pin removal were also used to extend the lower range of the calibration down to 34.0 (g/cm). The position of missing fuel pins or the distribution of fuel pins of different plutonium mass is not expected to affect the measurement results. The evaluation of measurement results described in the next chapter and MCNP calculations [9] confirm this theory.

2.5.4. MCNP Calculations.

A limited number of the experiments on the VENUS LWR MOX mock-up were also simulated using an adapted version of the Monte Carlo Code for neutron transport set up at A.E.A., Harwell. The model previously set up [9,10] was changed to allow for the VENUS mock-up fuel and poison rod characteristics. The calculations were able to provide the following data:

- A correction to the Totals, uncorrected Reals and multiplication corrected Reals response as a function of the fuel height.
- A correction for the difference between stainless steel and zircalloy fuel pin cladding.
- The theoretical effects to the Totals and Reals measurement response for the insertion of increasing numbers of B_4C rods in the open guide channels.
- The theoretical effects to the Totals and Reals measurement response for the insertion of various different clusters of 16 poison rods.

2.5.5. Cadmium Ratio Measurement.

A limited number of measurements for long counting times were carried out using the NCC in the active mode before and after deploying cadmium liners between the assembly and the detector panels. The measurements were made on the 17 by 17 LWR LEU mock-up before and after the insertion of Pyrex poison rods. Measurements were also performed on a 15 by 15 LWR LEU configuration for comparison between the VENUS and the LANL reference mock-up assemblies. The ratio of the measured Reals counting rate with and without the use of cadmium liners has been used to independently verify the number of poison rods present in LWR LEU fuel assemblies [5].

3.Results.

3.1. Addition of Clusters.

The relative net Reals counting response for the addition of the various combinations of poison rods in clusters previously outlined in point 2.5.1. are graphically displayed in fig.4. It should be noted that there is a considerable perturbation for the addition of the Pyrex rods. The results show that the relative location of the poison rods with respect to the position of the interrogation source to be very significant. The results for the combinations of poison rods which are more symmetrically distributed are more consistent.

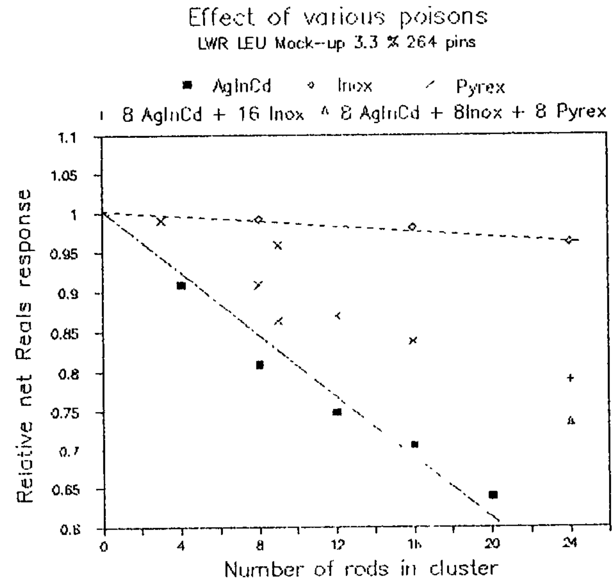

Fig.4.

A comparison between theoretical and experimental results for the LWR MOX mock-up with addition of B_4C rods case is shown graphically in fig.5. The results are in good agreement considering the errors associated with the MCNP calculation. A comparison between the theoretical and experimental results for the addition of poisons in clusters of 16 rods for the different poison types is given in the table below. The computer processing run times for the MCNP calculations were in the order of 1000 minutes for each data point.

Poison Type	T_{mcnp} (sT)	T_{exp} (sT)	R_{mcnp} (sR)	R_{exp} (sR)
B_4C	.955(.003)	.962(.001)	.916(.005)	.899(.002)
AgInCd	.985(.005)	.977(.002)	.945(.009)	.941(.003)
Pyrex	.998(.004)	.992(.002)	.980(.011)	.973(.003)
Inox	1.01(.004)	1.01(.001)	1.03(.008)	1.03(.002)

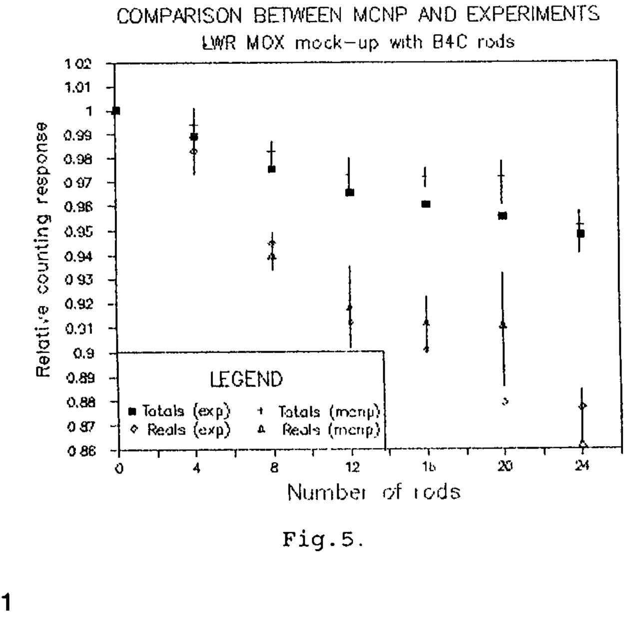

Fig.5.

Considering that the resulting multiplication corrected Reals rates (R_{mc}) are strongly dependent on the corresponding Totals rates the overall effect on the multiplication corrected Reals is small. The poison type which influences the measured response the most was the B_4C rods which caused a reduction of 0.11 % per rod. The position of the poison rod in the assembly is not as important as with the technique for the assay of LEU due to the absence of perturbations caused by source sample coupling. A correction algorithm for the presence of poison rods in clusters is suggested.

$$K_{(Thp)} = \frac{R_i \text{ (no poison)}}{R_i \text{ (poison)}}$$

$$\text{for LEU } R_i = R_{net}$$
$$\text{MOX } R_i = R_{mc}$$

$$K_{(Thp)} = a.n + b$$

where n is number of rods and a and b are the correction coefficients.

The correction coefficients for all cases are given in the table below.

	Poison Type	a	b	σ^2_a 10^{-8})	σ^2_b (10^{-6})	Covab (10^{-7})
LEU	AgInCd	0.0278	0.998	66.9	66.2	-48.0
	Inox	0.0016	0.998	4.3	8.7	- 4.8
	Pyrex	0.0129	1.001	6.2	6.1	- 4.8
		(10^{-4})				
MOX	B_4C	10.0338	1.001	1.09	1.73	-1.05
	AgInCd	6.9046	0.999	1.71	2.16	-1.52
	Inox	-4.3026	1.000	3.22	2.68	-2.05
	Pyrex	0.5924	1.000	0.68	1.09	-6.82

The correction factor $K_{(Thp)}$ corresponds to the factor K4 used in the general correction algorithm outlined in 2.1. and [5]. It has also been proven that multiples of the correction factors can be used to account for cases where the cluster comprises of several different types of thermal neutron poison. For example a cluster inserted into a LWR LEU fuel assembly which contains 8 AgInCd rods, 8 Inox rods and 8 Pyrex rods would require a correction to the measurement response corresponding to K4 where K4 is calculated as follows.

$$K4 = K_{AgInCd} \cdot K_{Inox} \cdot K_{Pyrex}$$

K_{AgInCd}, K_{Inox} and K_{Pyrex} are the correction factors for the number of pins of each poison type present.

3.2. Fuel Pin Removal and Substitution.

The relative multiplication corrected Reals measurement responses for the removal and substitution of MOX fuel pins are shown graphically in fig.6.

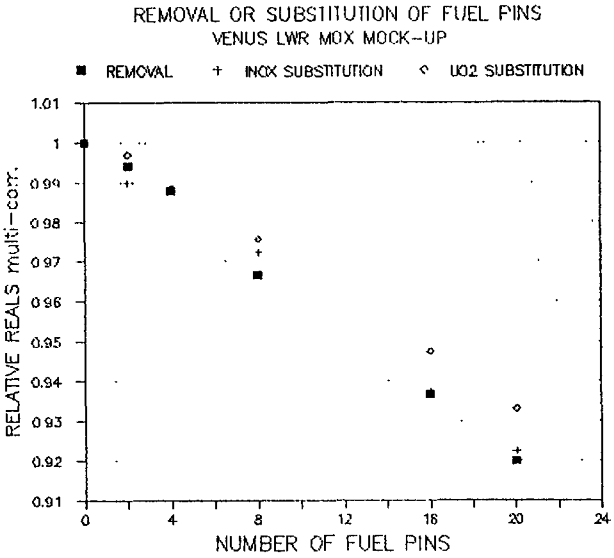

Fig.6.

The results show that the removal or substitution of fuel pins with stainless steel (Inox) rods have virtually the same effect whereas the substitution of MOX fuel pins with UO_2 fuel pins gives rise to lower reduction in the measured response. This effect is most probably due to the increase in the ^{235}U loading while also maintaining the heavy metal ratio in the assembly. The reduction in the measured R_{mc} rates for the various diversion scenarios are as follows:

UO2 pin	Substitution	0.36 % / pin.
MOX pin	Removal	0.41 % / pin.
Inox rod	Substitution	0.43 % / pin.

The reduction in the measurement response for removal or substitution of fuel pins from the inner or the outer zones of the fuel assembly is equal. The ratio of reduction in response between zones is 0.998 +/- 0.002. Considering the overall effect on the measured ^{240}Pu mass loading for the various diversion scenarios it shows that for a typical counting period of 600 seconds the accuracy of the NCC is not better than 3 to 4 fuel pins (1.5 %). It should however be noted that the measured Totals rates greatly influences the correction to the Reals and if the uncorrected REALS alone could be used for the calibration and subsequent assay of the assemblies the accuracy could be improved to 1 to 2 fuel pins (0.8%).

3.3. MOX Calibration.

The measurement results relative to the combinations of several fuel pin types described in 2.5.3. are used to establish a calibration for LWR MOX fuel assemblies. Before the calibration established for the VENUS mock-up can be used for the assay of full size fuel assemblies it is necessary to normalise for differences in their physical characteristics. The main differences between the VENUS mock-up and typical full size assemblies are listed as follows:

Characteristic	VENUS mock-up	Full size Assembly
Fuel length	50 cm	366 cm
Pin diameter	0.90 cm	0.82 cm
Cladding type	Inox	Zr-4
Cladding thickness	0.38 cm	0.57 cm
Pitch (fuel pin)	1.26 cm	1.26 cm
Heavy metal loading	1622 g/cm	1260 g/cm

MCNP calculations have been performed to investigate the importance of the different characteristics relevant to the NCC performance specifications and some correction factors have been established. It is estimated that the overall effect to the multiplication corrected Reals for the difference in Zr-4 and Inox fuel cladding types is not greater 1.5 %. The effect of the fuel length is shown in fig.7. The errors associated with the calculated data points are of the order of 1 % for a 1000 minute run time per data point.

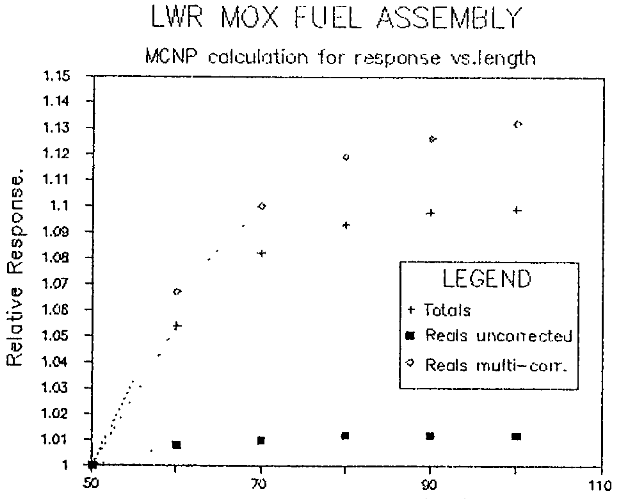

Fig.7.

The results show that the Totals and Reals response increases relative to the increase in the fuel length until the fuel is more than 100 cm. Once again the influence of the measured Totals response to the multiplication corrected Reals is apparent. The overall correction to the multiplication corrected Reals for normalisation of measurements on the VENUS mock-up to full size fuel assemblies is estimated to be 13.5 %. The overall effect of the differences in fuel length and cladding is estimated as 12 %. The measurement results used for the VENUS calibration have been normalised to a selection of results for measurements on full size fuel assemblies using the same NCC detector after application of the corrections for fuel cladding and length. The results are shown graphically in fig.8.

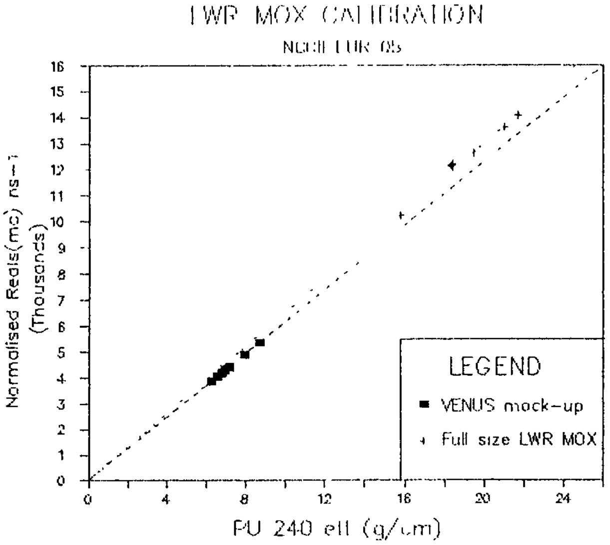

Fig.8.

The calibration coefficient for the LWR MOX, established using measurements on full size fuel assemblies [11] was 647.9. The corresponding coefficient for the VENUS reference calibration is 601.4 +/- 0.5 which is 7.2 % smaller. It is interesting to note that a recent calibration established for Fast Reactor fuel sub-assemblies using the NCC in a thermal mode (without cadmium liners) produced a coefficient of 624 +/- 5 [8].

The reduction in the measured response is thought to be related to the heavy metal loading of the VENUS reference assembly. The linear heavy metal loading should be corrected for in much the same way the linear U loading is corrected for in the NCC technique for the assay of LWR LEU fuel assemblies [5]. MCNP calculations have not been carried out to estimate the perturbation caused by the heavy metal loading but a factor will be established in the near future. The VENUS calibration was used to re-evaluate the measurement results to estimate the expected operator-inspector differences. The results were as follows:

Average Operator-Inspector Diff's. Signed	Absolute
-0.05(0.39)	0.32(0.37)

3.4. Cadmium ratio method.

Before discussing the results of the measurements carried out on the LEU mock-up with and without using cadmium liners it is useful to note the absolute relationship between the reference material used for the calibration experiments at LANL, USA and the material used at VENUS, Mol, Belgium. After application of the relevant normalisation factors [5] to the NCC(EUR 3), which was used for both calibration exercises, the expected measured net Reals for the 15 by 15 (204 fuel pin) LWR LEU mock-up at VENUS was 156.2 c/s. The actual measured net Reals was 154.8 +/- 0.8 c/s. The 1 % reduction in the measurement response is due to a small loss of detection efficiency (end effects) for the shorter active length of the fuel pins. The measurement results for the insertion of clusters of Pyrex rods with (R_{cd}) and without (R_{no-cd}) using cadmium sheets to line the inner faces of the NCC detector panels are shown graphically in fig.9.

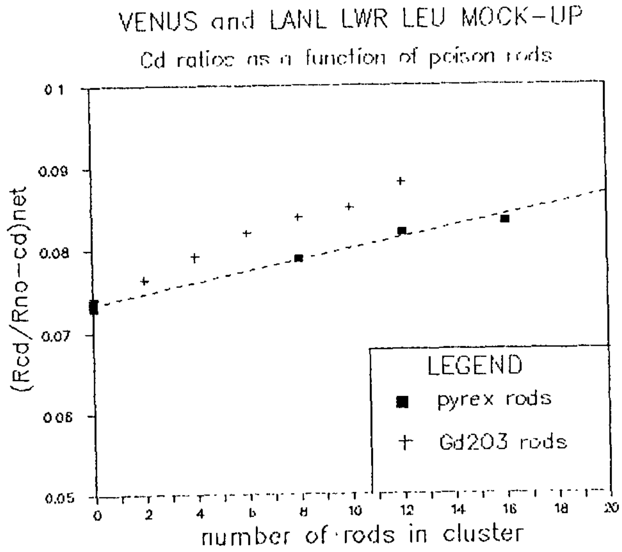

Fig.9.

The ratio R_{cd}/R_{no-cd} for the 17 by 17 VENUS mock-up before insertion of the poison rods agrees well with the ratio established for the 15 by 15 LANL mock-up [5] the values being 0.0731 and 0.0742 respectively. The cadmium ratio for the 15 by 15 VENUS mock-up was 0.0736. A calibration function is proposed for the purpose of independently verifying the number of Pyrex poison rods present in the cluster. The linear relationship is represented by the equation

$R_{(cd/ncd)}$ = a.n + b where n is the number of poison rods and a and b are the calibration coefficients. When no poison rods are present the cadmium ratio is equal to b. The coefficients with their associated errors are given below. The cadmium measurement results were established using long counting times of 10 times 5000 seconds for each poison loading.

a = 0.000671 +/- 0.000058
b = 0.0732 +/- 0.0006
Cov ab = $- 2.8.10^{-8}$

The fuel material and time was not available to make calibrations for all types of poison material during the exercises but either future experimental work or MCNP calculations could be performed to obtain the necessary coefficients.

4. Conclusions.

Euratom NCC's have been calibrated for the assay of LWR LEU and MOX fuel type assemblies and correction factors have been established for the presence of several types of thermal neutron poisons.

The measurement results have shown that although the NCC is not sensitive to the position of poison rods within the LWR MOX assembly a certain amount of perturbation in the measurement response can arise with NCC LEU assembly due to the AmLi source to sample poison rod coupling.

A calibration function has been proposed for the independent verification of the presence of thermal neutron poisons in the form of Pyrex rods using the cadmium ratio method established at LANL, USA.

The agreement between MCNP calculations and experimental data indicates that future calibration work can be reduced and in some cases replaced by mathematical modelling alone, especially when only relative effects are being investigated.

The NCC is not sensitive to the removal or substitution of less than 4 LWR MOX fuel pins from a 17 by 17 assembly for counting periods shorter than 600 seconds.

More mathematical modelling and experimental measurements are recommended in the future to investigate the requirements for a correction factor for the heavy metal loading of LWR MOX assemblies in order avoid possible bias effects.

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CALIBRATION OF UFBC COUNTERS AND THEIR PERFORMANCE IN THE ASSAY OF LARGE MASS PLUTONIUM SAMPLES.

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Abstract.

The paper reports on the cross-calibration of four Universal Fast Breeder reactor assembly coincidence (UFBC) counters using multi-can containers of Plutonium oxide powders with masses between 2 and 12 Kg. of plutonium and a parametric study on the sensitivity of the detector response to the positioning or removal and substitution of the material with empty cans. The paper also reports on the performance of the UFBC for routine measurements on large mass, multi-can containers of plutonium oxide powders and compares the results to experience previously obtained in the measurement of fast reactor type fuel assemblies in the mass range 2 to 16 Kg.of plutonium.

1.Introduction.

The UFBC counter is a member of the HLNCC family of passive neutron counting systems [1] and was originally designed specifically for the assay of Fast Breeder Reactor type fuel assemblies. The present paper reports on the application and the performance of the UFBC for the assay of large, multi-can containers of plutonium oxide powders. Four Euratom UFBC counters, three of the standard design and one of a modified larger design, have been cross-calibrated using a multi-can container of reference plutonium oxide powders similar to those produced at the output stage (PuO_2 product) of spent fuel reprocessing plants and recieved at the input of plutonium fuel fabrication plants. Measurements have been performed using one UFBC as the reference counter for calibration purposes and to study the effect on the measurement response after changing the position of the cans of plutonium powders within the multi-can container, characterising it's sensitivity to the removal or substitution of plutonium material. The purpose of the study is to monitor the UFBC's sensitivity to various diversion scenarios associated with empty or part-filled cans or by introduction of spacers in the container packing. A selected set of cans of reference materials and californium-252 spontaneous fission neutron sources was measured by all four UFBC counters to establish their relative detector efficiencies. The paper goes on to report on the performance of the UFBC for routine measurements on multi-can containers of plutonium oxide under in-field conditions and compares the results with experience of measurements on various types of plutonium and mixed (U/Pu) oxide fast reactor type fuel assemblies.

2. Materials and Methods.

2.1. Measurement Technique.

The UFBC is a non destructive assay (NDA) instrument designed by the Los Alamos National Laboratory (LANL), Los Alamos, USA. The instrument resembles the HLNCC [2] in that it uses similar digital electronics and detects the coincidence neutrons from the spontaneous fission of the 240 Pu eff. mass. It is however designed to operate for much taller samples and for much higher counting rates. The maximum sample diameter for the standard version is 17 cm and 23.2 cm for the modified version. The uniform counting (flat response) region in the detector is around 105 cm. The original instrument design and performance characteristics have been reported in detail elsewhere [3] but a sectional drawing and table of specifications summarising the basic features of both versions of the UFBC are given in fig.1. and table.1. respectively.

Table.1.

	Dimensions (cm)								
Version	A	B	C	D	E	F	G	H	I
UFBC II	141	31	17	-	-	9	17.5	15	-
UFBC IImod	137	38	23	15	17	9	17.5	15	3

The differences in operating characteristics between the 4 Euratom UFBC instruments are summarised in table.2.

The UFBC is calibrated in the same way as any other HLNCC-II based measurement system through the use of a series reference samples of differing $^{240}Pu_{eff}$

mass. The calibration function takes the form

$$R = a.(^{240}Pu_{eff.})^2 + b.^{240}Pu_{eff.}$$

for the uncorrected Reals response and

$$Rmc = a.^{240}Pu_{eff.}$$

for the multiplication corrected Reals response.

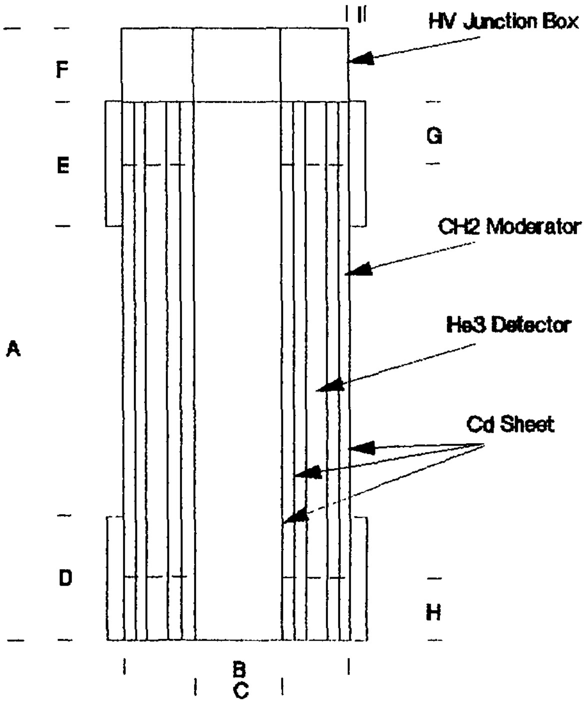

Fig.1.

Table.2.

Detector N_o.	EUR01	EUR02	EUR03	EUR04
H.V.(volts)	1680	1680	1680	1680
Dead-time Coefficients				
a (10^{-6}s)	0.62	0.775	0.726	0.62
b (10^{-12}s)	0.20	0.250	0.234	0.20
Rho 0	0.4992	0.4992	0.4992	0.0591
Linear Response Zone(cm)	95.0	95.0	95.0	100.0
Coincidence Gate Length (10^{-6}s)	64	64	64	64
Die-away time (10^{-6}s)	22	22	22	25
Relative Detector Counting Efficiencies				
(Point source)	1	1.186	1.188	1.119
(Large sample)	1	1.250	1.280	1.140

2.2. Reference material.

A series of plutonium oxide reference powders was available to fill the multi-can containers for the purpose of creating a set of samples for calibration of the UFBC. The containers were typical of containers used in the nuclear fuel cycle. The cans are constructed of stainless steel, 34 cm in height and 12 cm diameter with a wall thickness of 0.3 cm. Up to five cans could be placed one after the other in the container which is also constructed from steel and has a diameter of 14 cm with a wall thickness of 0.3 cm. The results outlined in this paper concentrate on measurements on a maximum of 3 fuel cans placed in the container. The characteristics of the reference material selected for the studies are given in table.3.

Table.3.

		Isotopic Composition (6^{th} Feb.1989) (wt/wt % Pu)					
Can	Pu mass(g)	^{238}Pu	^{239}Pu	^{240}Pu	^{241}Pu	^{242}Pu	^{241}Am
A	2199.6	1.11	70.07	18.36	7.93	2.53	0.6437
B	2467.4			idem.			
C	2301.2			idem.			
F	1852.4	1.76	56.48	25.07	10.51	6.17	0.6256
H	1892.7	0.60	65.02	23.10	8.11	3.17	1.0986
I	1819.5	1.04	70.72	18.90	6.76	2.58	0.5378
J	2190.5	1.30	59.85	24.02	9.97	4.85	1.7539
K	1852.4	1.76	56.48	25.07	10.51	6.17	0.6256

2.3. Measurement set-up.

The UFBC was placed on a metal table support which has a hole bored in it's centre to permit the container to pass through it. In this way the relative position of the cans of plutonium with respect to the linear counting zone of the UFBC could be appropriately adjusted. For the purpose of the results described in this paper the container was permanently positioned with 5 cans as shown in fig.2.

The cans of plutonium oxide were changed within the container to vary the plutonium mass. The cans of plutonium were always placed in positions 2,3 or 4 to assure that the material lay within the linear counting zone of the UFBC. In some cases positions 2,3 and 4 were also occupied by either 5 or 10 cm aluminium spacers or multiples thereof. The relative positions of the cans of reference material for calibration measurements are listed in table.4. The position numbers correspond to those indicated in fig.2. "X" indicates an empty can.

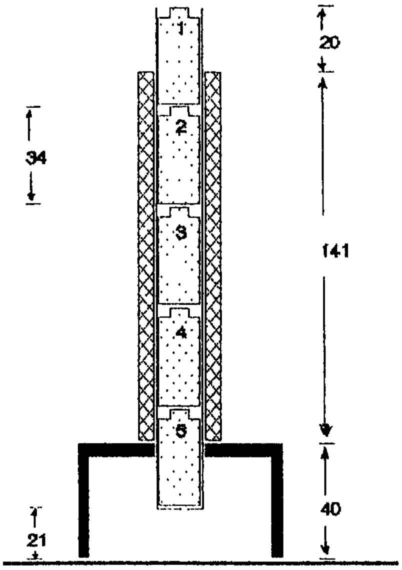

Fig.2.

Table.4.

Measurement i.d.	Position/Can i.d . 1	2	3	4	5
I	X	X	X	A	X
II	X	X	A	X	X
III	X	A	X	X	X
IV	X	X	B	A	X
V	X	B	A	X	X
VI	X	C	B	A	X
VII	X	H	I	F	X
VIII	X	I	H	X	X
IX	X	X	F	X	X
X	X	X	I	X	X
XI	X	X	K	X	X
XII	X	K	H	I	X

3. Results.

3.1. Calibration Measurements.

The Euratom UFBC 01 was used for the calibration measurements. The results are summarised in table.5.

Table.5.

Measurement	$^{240}Pu_{eff}$	alpha	R	sR	R_{mc}	sR_{mc}
		i.d.	(g)			
I	551.9	0.844	3482.6	39.0	1961.1	21.1
II	551.9	0.844	3179.5	36.0	1849.0	20.9
III	551.9	0.844	3236.7	37.7	1906.0	22.2
IV	1170.9	0.844	7173.7	90.0	3996.0	50.6
V	1170.9	0.844	7140.1	82.4	3966.5	45.8
VI	1748.3	0.844	10788.0	129.4	5997.0	71.9
VII	1749.6	0.700	9643.9	117.7	6066.0	74.0
VIII	1043.2	0.659	5653.8	62.6	3458.0	38.3
IX	725.1	0.762	3750.5	45.6	2439.3	29.7
X	464.6	0.786	2532.5	29.4	1555.9	18.1
XI	329.5	0.844	1671.8	21.3	1116.0	14.2
XII	1353.9	0.702	7542.8	87.9	4634.6	54.0

A calibration for plutonium oxides in large multi-can containers in the mass range from 330 to 1750 grams of $^{240}Pu_{eff.}$ has been established using this data. The calibration coefficients corresponding to the measured Reals response before and after neutron multiplication corrections are as follows:

		Uncorrected	Corrected
a	=	0.00022679	3.4047
b	=	5.4573	-
Va	=	0.00000006	0.0004
Vb	=	0.0675	-
Covab	=	-0.0000581	-

where Va, Vb and Covab are the variances and co-variance of a and b respectively.

Using the calibration functions to evaluate the measurement results produces the following comparison between declared and measured plutonium masses for the large mass samples.

Uncorrected Reals calibration

Average Declared-Measured Diff's. Signed	Absolute
-0.63(6.4)	5.8(6.5)

Corrected Reals calibration.

Average Declared-Measured Diff's. Signed	Absolute
-0.05(0.3)	1.5(1.8)

Measurements were performed on the container for cans A and B positioned in various locations inside the linear response zone in order to investigate any perturbation of the measurement results related to the de-coupling of the samples. The relative results for measurements on cans A and B with 5 and 10 cm aluminium spacers and empty cans used to separate the fuel cans are compared in table.6.

Table.6.

Combination	Relative Response
XABXX	1.000 +/- 0.016
XXABX	0.995 +/- 0.015
X35ABX	0.988 +/- 0.016
X30A5BX	1.009 +/- 0.015
X25A10BX	0.987 +/- 0.016
X20A15BX	0.985 +/- 0.015
X15A20BX	0.992 +/- 0.014

The results do not indicate that the de-coupling of the plutonium masses effects the measurement response providing that the total mass of the sample lies within the linear response zone of the UFBC.

3.2. In-Field Measurements.

Measurements on large single cans of plutonium oxide and small fast reactor and zero energy reactor type fuel assemblies have been evaluated using the above calibration for the multi-can containers of plutonium oxide. The average operator-inspector differences are given for comparison in table.7.

Table.7.

Sample Type	Avg. Diff. %
PuO_2	
Multi-can ref.	-0.05 (0.4)
Single can	-0.89 (1.7)
MOX Fast Reactor	
Type 1	-1.16 (0.7)
Type 2	-3.21 (1.0)

The type of multi-can containers that are used in the nuclear fuel cycle at the present time comprise of 4 cans that are smaller in height than those used during the calibration exercise. The cans are also contained in sets of two in an intermediate container. The two intermediate containers are welded together to form one large container which is in turn encapsulated in another outer container made from stainless steel. The individual cans are approximately 24.0 cm in height and the overall length of the outer container is 112 cm. The layout of the cans is shown graphically in fig.3.

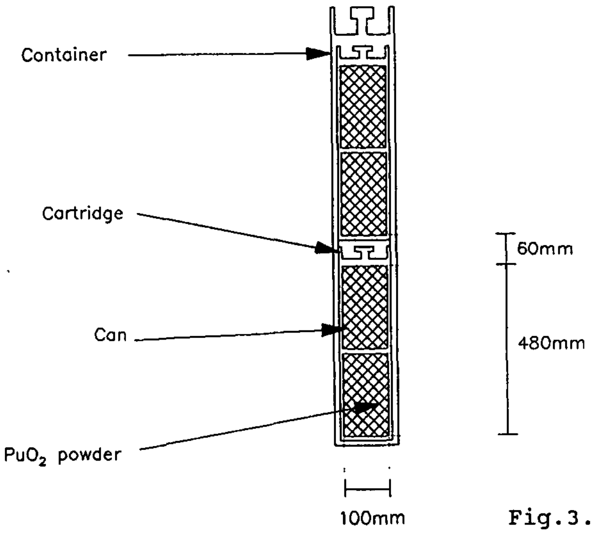

Fig.3.

Euratom have already carried out in-field measurements on a number of multi-can containers of large plutonium masses using both the standard and modified UFBC counters. The measurement results have been normalised for small differences between detector efficiencies and the value of Rho 0 used in the neutron multiplication correction algorithm.The results are compared to the calibration measurements on the 3 large can containers. The results are shown graphically in fig.4.

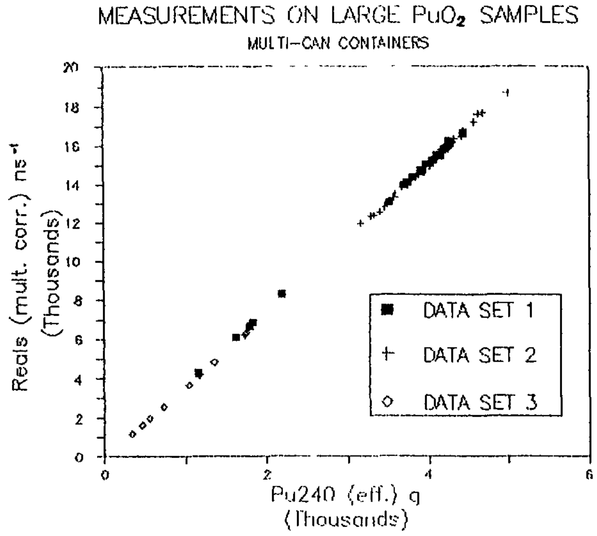

Fig.4.

The calibration used for in-field measurements is similar to the calibration established for the 3 large can containers. The coefficient a for the multiplication corrected Reals response has been established as 3.7626 +/- 0.0036 . The increase in the slope of the calibration is accounted for by the increased mass range of the samples which are effectively compressed into the same counting geometry (more dense) coupled with the slightly longer linear counting region of the Totals and Reals for the UFBC with respect to the position of the sample . Evaluation of the measurement results for various data sets using data set 1 as the origin for the in-field calibration of large plutonium oxide samples are compared in table.8.

Table.8.

Sample Type	Avg. Operator-Inspector Diff. %
PuO_2 Powders	
Data set 1	-0.28 (0.58)
Data set 2	0.02 (0.70)
Data set 3	4.95 (1.44)
MOX Fast Reactor Fuel Assemblies.	
Data set 4	3.85 (0.93)
Data set 5	1.90 (0.95)

Data set 1 and 2 represent results of measurements on 17 and 54 of the 4 can multi-can type containers respectively. Data set 3 represents the results of the measurements on the 12 larger can type containers. Data sets 4 and 5 correspond to measurements on 5 and 15 different fast reactor type fuel assemblies.

4. Conclusions.

Four universal fast breeder coincidence (UFBC) counters have been calibrated for the measurement of plutonium oxide powders in multi-can containers up to 13 Kg. of plutonium metal content.

The correction for differences in detector efficiency between instruments is dependent on the size of the sample to be assayed due to the increased totals efficiency at the extremities of the detectors.

The measurement response is not effected by the de-coupling of fuel cans within the container provided that the total mass of plutonium lies within the flat counting region of the UFBC.

The slope of the multiplication corrected Reals calibration for plutonium oxide is similar to that calculated for fast reactor type fuel assemblies. The value being 3.76 for PuO_2 and 3.70 for MOX fuel assemblies [4].

The performance of the UFBC can be characterised by the average operator-inspector differences for in-field measurements on large quantities of plutonium oxide powders. The overall average mass residual was 0.072 % +/- 0.63 %. The measurement precision for 300 second counting times is 1 %.

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UNATTENDED MODE MONITORING OF PASSIVE NEUTRON COINCIDENCE DETECTOR SYSTEMS USING A COMMERCIAL DATA LOGGER

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Abstract

A commercial Data Logger for unattended passive neutron coincidence data acquisition is described. This consists of an inexpensive commercial Data Logging equipment attached to a neutron coincidence electronics and a software package for data review. The Data Logger permits both the flexible configuration of a passive neutron coincidence measurement system for unattended mode monitoring and the storage of the measured Totals and Reals count rates.

An additional feature of the Data Logger is a custom software package providing for the complete analysis of the stored data and yielding an assay of each item passing through the measurement cavity. The analysis includes an input for different isotopic compositions, the calculation of the multiplication corrected Reals rates, the inclusion of a calibration functions, and the determination of $^{240}Pu_{eff.}$ masses.

The software package for data review displays the Totals and Reals count rates logged by the Data Logger as a function of time. In addition the custom software provides input files to the data review package to display the multiplication corrected Reals count rates and the measured $^{240}Pu_{eff.}$ masses as a function of time.

Information on the Data Logger is presented along with the monitoring mode specifications. The analysis functions implemented are described as is the data review software. Results are presented for a specific application.

1. Introduction

The Euratom Safeguards Directorate is being increasingly called upon to develop and install non-destructive assay (NDA) systems for unattended operation in new large scale nuclear facilities in the European Communities. The introduction of neutron coincidence detectors and acquisition electronics for unattended operation becomes obligatory:

- when nuclear material is handled automatically in remote controlled nuclear facilities
- where nuclear material is inaccessible for measurement or sampling by inspectors in the normal way
- to reduce the radiation dose to operator staff and inspectors caused by the increased number of measurement needed on large quantities of plutonium, i.e. higher throughputs and larger quantities in a single item
- when the investment in new measurement systems for unattended operation becomes cost effective due to the considerable increase in manpower overheads associated with the use of conventional attended measurement systems in new nuclear facilities.

The first substantial effort to design safeguards specific neutron coincidence detector systems for unattended operation was initiated in the Los Alamos National Laboratory (LANL) for the PFPF project/1/. LANL thus laid down the fundamental guide-lines for future development of unattended safeguards measurement systems. These principles can be summarised as such:

- redundancy in neutron coincidence electronics and computers
- continuous collection with short measurement times, e.g. 1 minute
- infrequent data review, e.g. 1 per month
- sealed cabinets
- continuous cable runs
- software self-diagnostics
- data collection in electronics cabinets near to the detectors.

These principles were translated by LANL in to a functioning system consisting of:

- local data acquisition, i.e. the "COLLECT" in the LANL applications, based upon a continuous collection of raw data for short acquisition times and the recording of that raw data after "marking" according to the type of data acquired
- off-line graphing of data, i.e. the "REVIEW" in the LANL applications, based upon a custom written software package
- preparation of data for subsequent evaluation using a separate additional software package to obtain an assay result.

The Euratom Safeguards Directorate took these guide-lines and investigated the possibility to add the following additional principles:

- integrated data evaluation including assay results
- use of commercial electronics and improved graphing software
- reduce size and weight of data acquisition hardware
- improve reliability by using battery backed-up RAM and not a hard disk
- reduction of the data to be stored through some form of "front-end data processing" involving:
    - storage of the Totals and Reals rates and not the raw data registers, t, T, R+A, and A, from the neutron coincidence electronics package
    - user definable thresholds on both the Totals and Reals rates to detect data of interest and to eliminate background recording
    - user definable frequency for the recording of the background.

Once these principles were established the Euratom Safeguards Directorate decided to invest in the modification of a commercial data logger system which was already being evaluated for the unattended data logging of tank volumes. The selected data logger

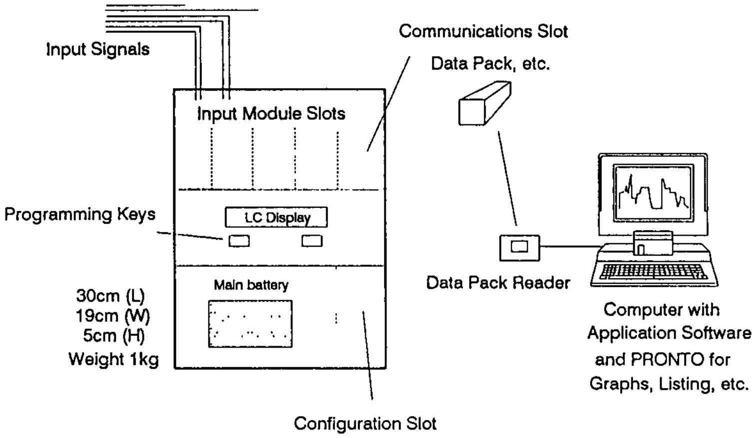

Fig. 1 The RANGER Data Logger System

is called the RANGER and comes complete with an applications software called PRONTO.

The RANGER* (Model: Rustrak Ranger II) is an advanced microprocessor-based data collection system permitting recording, analyzing, and reporting of physical and electrical information on an unattended basis. The basic components of the system include the data logger, input signal conditioning modules, communication modules, configuration modules and the PRONTO application software program.

The data logger functions as a paperless recorder storing data in memory at a rate and length of time specified by the user. Once the recording process has been completed, the data can be transferred to a computer. The PRONTO application program provides all the tools necessary to graph, review, analyze and report the data without the necessity for any programming on the part of the user.

Fig. 1 shows the RANGER data logger, its essential components, and the applications software PRONTO running on a separate computer. Fig. 2 shows the functions of the application software PRONTO.

The Euratom Safeguards Directorate considers that the selection of the RANGER-PRONTO combination has the following advantages:
- small size (30cm (L), 19cm (W), 5cm (H))
- light weight (approx. 1kg)
- easy to use and to configure
- no moving parts (data stored in battery backed-up RAM)
- high resistance to temperature, humidity, shock and vibration
- high flexibility with different input signal modules, etc.
- battery back-up system set-up
- software provides a common user interface for different logging applications
- flexible commercial graphing package
- professional user descriptions/manuals
- low cost for purchase and maintenance
- flexible, reasonably prices, development engine for safeguards applications.

The complete project, see Fig. 3, thus involved

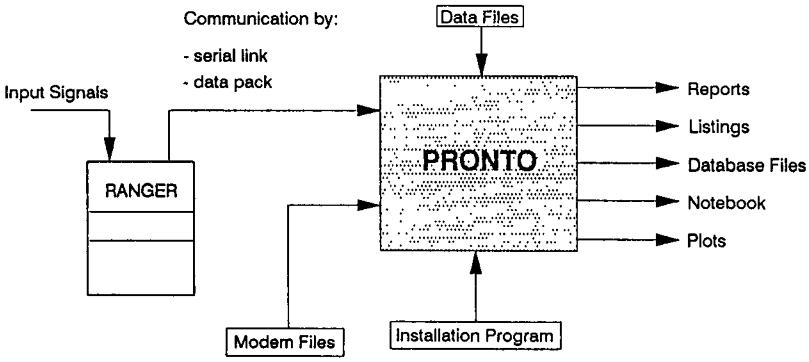

Fig. 2 Schematic of Link Between the RANGER and PRONTO

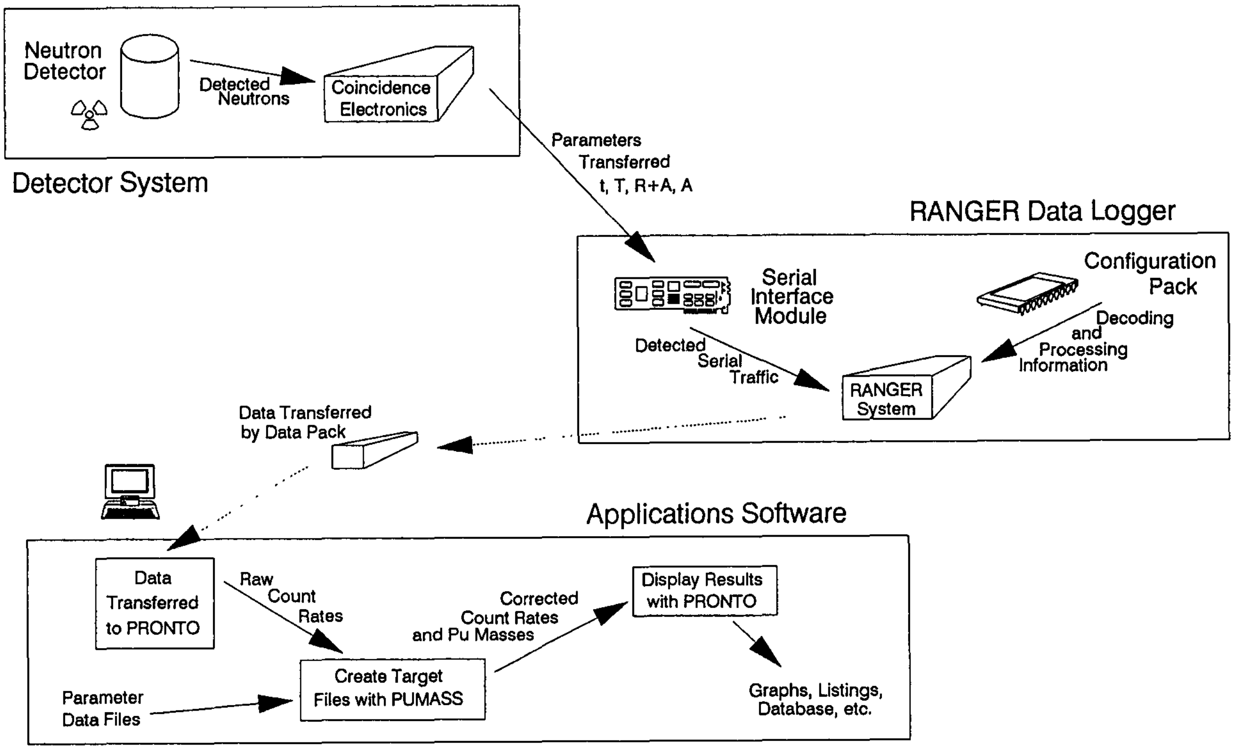

Fig. 3 Data Acquisition and Processing System

the following:
- a detector system based upon standard neutron coincidence detector designs/2/
- a modified RANGER data logger with both a serial input signal conditioning module and a custom built configuration module
- neutron coincidence data analysis algorithms/3/, in a custom written program called "PUMASS", integrated in to the data loggers application software "PRONTO".

2. The Data Logger

The RANGER data logger (see Fig. 1) consists of 4 separate slots for input signal conditioning modules (single or dual), a communications slot for a communications module or data pack for data storage, and a configuration slot for configuration modules.

The Euratom Safeguards Directorate commissioned the design of a special EURATOM configuration module which when coupled with an appropriate input signal conditioning module and data pack provides an autonomous data logger for connection to a neutron coincidence electronics package JSR-11**.

The RANGER data logger itself has the following basic features:-

A two button programming system in conjunction with a two line, 16 character/line LCD provides a clear and quick set-up and recording sequence.

Menus that are dependent on specific modules, e.g. communication or configuration modules, etc., or functions that are dependent upon certain set-up conditions only appear when those devices and/or functions are present.

Intelligent self-identifying input modules allow automatic configuration for the number of channels in use and the type of signals to be measured. Thus a one key startup procedure is available. In addition a set-up mode allows the user to define:
- scales and engineering units
- calibration values
- alarm levels
- recording methods
- mathematical operations
- communications.

Five different modes for storing data, adaptive store, point store, enhanced point store, store on alarm, and manual store. The implementation described here is "store on alarm" conditioned by a new custom built configuration pack.

Data logger memory that is battery backed up to insure against loss of valuable data or set-up information for up to 2 weeks. During that time all programming, data storage information, system set-up parameters, and data in memory are maintained.

Several optional modules are available to further enhance the data logger's capabilities. They include configuration modules, data packs, and other types of communication modules.

Input Modules

Input modules are required in order to condition incoming signals to be compatible with the data logger. Four types of input modules are available, isolated single input (some types of isolated dual input modules also exist) and non-isolated single and dual input. Up to 4 different types of input modules can be installed in a RANGER at one time.

A 25 pin RS-232 serial interface module is used to provide the link between the neutron coincidence electronics package (type: JSR-11 from Jomar Systems) and the RANGER. The neutron coincidence electronics, JSR-11, is set to recycle and the front panel commands set to the correct gate width and measurement time.

There also exists an Alarm Output Module which provides for 8 sets of outputs based upon alarm levels programmed on any channel on the RANGER. This functions as an electronic switch when an alarm level is reached.

Communication Modules

Communication modules permit the RANGER to transmit data from internal memory or a Data Pack directly to a computer printer as well as over phone lines via modem.

Data Pack

Data Packs are transportable memory devices designed to permit retrieval of data from the RANGER's internal memory. A Data Pack option allows for transportation of the data from the collection site to the computer while leaving the data logger in place to continue recording data. The Data Pack is battery backed up for a minimum of 10 years under normal conditions (minimum 2.5 years under adverse conditions). An AutoWrite feature allows the RANGER to transfer data to the Data Pack automatically.

Configuration Modules

Configuration modules expand the versatility of the RANGER by adding additional capability to the system. Additional storage capacity and additional math channel capabilities can be added. Custom dedicated applications can also be implemented.

The EURATOM Configuration Module

A specific EURATOM configuration module has been developed which implements the following specific features:

- receipt of data from the JSR-11, the neutron coincidence electronics package
- determination of the Totals and Reals rates from the raw data, t, T, R+A, and A
- user definable rescaling of Totals rate by 1, 10, or 100 so as to contain figure in integer range 0-59999
- user definable rescaling of Reals rate by 1, or 10 so as to contain figure in integer range 0-59999
- user definable selection of data units, kTs for the Totals rates, and kRs for the Reals rates
- user definable thresholds for both the Totals and Reals rates determine if the raw data is to be stored for further analysis, i.e. if either of the Totals or Reals rate thresholds are exceeded the RANGER goes in to an "alarm" condition and stores both the Totals and Reals rates until the "alarm" condition is removed by both the Totals and Reals rates dropping below their respective thresholds
- user definable frequency for sampling of the data input traffic in the RANGER internal memory, the RANGER internal memory is updated independently through the serial link with the neutron coincidence electronics package
- user definable frequency for sampling and recording the "background" data, i.e. the data is sampled and recorded at a user definable frequency irrespective of the Totals and Reals rate thresholds thus providing an indication that the whole monitoring and logging system is working correctly
- implementation of three diagnostic alarms, i.e. neutron detector failure, JSR-11 neutron electronics failure, and serial input module failure
- storage of alarm conditions, both diagnostic and threshold alarms, only upon occurrence
- sleep function with the logger being programmed to start at some user definable time in the future.

The set-up of the EURATOM configuration module can be checked against a review function in the data logger once the set-up has been completed. Upon removal of EURATOM configuration module the RANGER reverts to its normal configuration and can be used for other applications such as tank monitoring, etc..

Storage potential of the RANGER is a function of:

- storage capacity for the total and Reals rates, normally 2 bytes each
- frequency of sampling of the input neutron coincidence signal
- number of samples per "event"
- frequency of "events", number of items passing through the measurement cavity
- frequency of background sampling
- total storage capacity of the data logger, 57 k bytes for a basic logger.

The storage capacity of the RANGER is thus a function of a number of variables. Three examples are described below to show the potential storage capacity:-

Example A:

- a sampling frequency of 10s
- an event lasting 10 minutes, i.e. 600s
- a background frequency of 15 minutes, i.e. 900s
- a storage capacity of 57k bytes plus a data pack of 256k bytes, equals 313k bytes
- 10 items per day
- with approx. 3k bytes/day storage **the storage capacity of the RANGER would be >100 days**

Example B:

- a sampling frequency of 10s
- an event lasting 5 minutes, i.e. 300s
- a background frequency of 60 minutes, i.e. 3600s
- a storage capacity of 57k bytes plus a data pack of 256k bytes, equals 313k bytes
- 100 items per day
- with approx. 12k bytes/day storage **the storage capacity of the RANGER would be near 25 days**

Example C:

- a sampling frequency of 1 minute, i.e. 60s
- an event lasting 10 minutes, i.e. 600s
- a background frequency of 1 minutes, i.e. 60s
- a storage capacity of 57k bytes plus a data pack of 256k bytes, equals 313k bytes
- 20 items per day
- with approx. 5k bytes/day storage **the storage capacity of the RANGER would be about 60 days.**

The RANGER Menu

The RANGER main menu is as follows:

- LOG for
- PLAYBACK
- DISPLAY
- SET-UP
- REVIEW
- POWER.

The "LOG for" is a mode where the RANGER functions as a paperless recorder for a user definable period. The "LOG for" can be changed to a "LOG every" depending upon the type of storage mode selected. A delayed start recording option also exists as does a feature incorporating auto synchronization which automatically aligns the sample and storage process for multiple data loggers.

The "PLAYBACK" mode controls the transfer of data between the RANGER and external computer, printers, etc. The "PLAYBACK" to a computer, printer, and Data Pack, are implemented as in Data Pack to computer, and via modem.

The "DISPLAY" mode shows the input signals on the LCD.

The "SET-UP" mode controls the RANGER configuration. The "SET-UP" includes setting of time and date, programming of high and low alarms, setting of print-out intervals, programming the delayed start time, etc.. In addition "SET-UP" provides access to the specific set-up parameters in the EURATOM configuration module.

The "REVIEW" mode permits the user to rapidly check the set-up parameters, and to inspect some summary information about each input channel. The LCD builtin to the RANGER is used to display the average, maximum and minimum values for each active channel along with the start time and recording duration. This is very useful for in-field system set-up.

The "POWER" mode allows the user to switch off the RANGER.

In addition to the main menu the RANGER has also a fully implemented error message system. Error messages cover battery conditions, input module stability, memory conditions, configuration module instability, invalid set-up conditions, etc.

The RANGER Technical Specification

Below are listed the technical specifications for the RANGER:-

Inputs: 12 channels with 8 analog, or D digital, 8-D analog D=<4, via input modules, and 4 builtin math channels
Analog Conversion: 16 bits
Analog Accuracy: +/-0.05% of full scale
Resolution: 0.1mVdc
Linearity: +/-0.03% of full scale
Crosstalk: -80db typical channel to channel
Digital Accuracy: +/-0.001% of full scale
Clock Accuracy: max. error 30 seconds/month
Environmental Conditions:
Temperature: -10°C to +60°C
Relative Humidity: 5% to 90% (non-condensing)
Shock and Vibration: MIL-STD 810 Method 514.2
Battery Charge: min. 32 hours (nominal 48 hours) from full discharge to 100%
Memory Back-up: min. 30 days memory protection after main batteries fully discharged
User Interface: 2 button membrane with panel and 2 line 16 character/line LCD
External Interface: RS-232 port 300 baud to 19.3 kbaud
RAM Storage: 64k, expandable to 256k
Enclosure: G.E. Polycarbonate (Lexan)
Dimensions: 30.18 cm (L), 19.3 cm (W), 4.76 cm (H)
Weight: 1.13 kg
Approvals:
FCC Compliance - Class A, paragraph 15 subpart J
UL, CSA, Factory Mutual
Input Modules (examples):
General Purpose DC Voltage and Current: 0-100mV, 0-100mAdc, +/-1Vdc, etc.
Process DC: 1-5 Vdc, isolated and non-isolated 4-20mA
AC Voltage and Current: 0-30Vac, 0-360Vac, 0-1000Aac, etc.
Temperature/Thermocouple: -200 to +760°C, 0-1500°C, etc.
Dual Channel: non-isolated 4-20mA, etc.
Pulse Counting: 0-800kHz, 0-59999 counts, etc.
Configuration Modules (examples):
Additional Storage Modes
Additional Math Functions
User Configuration Pack
Communication Modules (examples):
Serial Interface Cable (25 and 9 pin)
Modem Controller Module
Data Pack: 64k, 128k, and 256k

3. The Software

Software can be divided in to two separate sections, the first being "PUMASS" the custom software that takes measured Totals and Reals rates and produces corrected count rates and plutonium assay results, the second being "PRONTO" the applications software for the RANGER that provides graphs, listings, data archival, etc.

The Analysis Module PUMASS

As has been described above the RANGER records the Totals and Reals rates as calculated from the output of the JSR-11 neutron coincidence electronic package. The files of the Totals and Reals rates can be read by the applications package PRONTO, and in addition they can be analysed using a special custom written program called PUMASS. The program PUMASS takes the raw Totals and Reals rates and produces an additional series of files which can also be read by the applications package PRONTO. These additional files consist of time varying values of:

- background and normalization corrected Totals and Reals rates
- $^{240}Pu_{eff.}$ and Pu mass, the uncertainty, and the difference between declared and measured values, before the multiplication correction
- background, normalization, and multiplication corrected Reals rates
- $^{240}Pu_{eff.}$ and Pu mass, the uncertainty, and the difference between declared and measured values, after the multiplication correction.

Once these new files are created they are stored and can be accessed by the applications package PRONTO for graphing, listing, data archival etc. Fig. 4 shows the assay mass calculation data flow according to the standard procedure for data analysis/3/.

Input files to the program PUMASS include (i) a file containing neutron detector constants and calibration constants, and (ii) a file containing measurement variables specific to each item.

The first file contains fields for the following detector constants:

- neutron detector dead times
- gate width of the neutron coincidence electronics package
- K constant relating the moments of the induced and spontaneous fission neutron multiplicity distributions
- p_0 relating the Totals and Reals for a non-multiplying sample
- calibration constants before and after multiplication correction

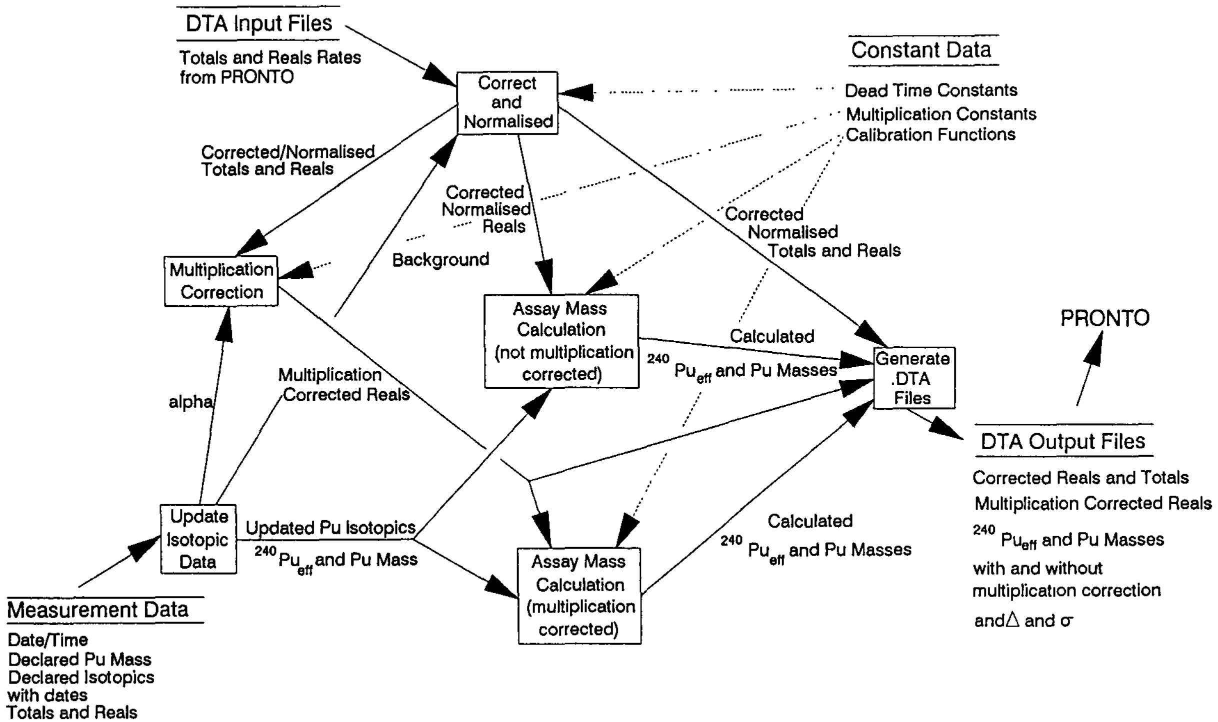

Fig. 4 Assay Mass Calculation Data Flow Diagram

- associated variances and covariances.

The second file contains fields for the following variables, item by item:
- date and time of entrance of item in detector taken from either the raw data review or/and from other information such as a review of surveillance films, etc.
- a normalization factor and uncertainty taken from an initial of the review of the raw data
- a Totals and Reals background taken from an initial raw data review
- the Pu mass taken from operator declarations
- the Pu isotopic composition and valid date taken either from operator declarations or independent safeguards measurements
- the ^{241}Am (ppm) and valid date taken either from operator declarations or independent safeguards measurements.

The Graphics Package PRONTO

The PRONTO application software is uniquely configured to match the data format of the RANGER. The software PRONTO provides the user with a complete set of tools for plotting, annotating, analysing, and printing reports on the data collected by the RANGER.

PRONTO implements the following functions:
- transfer and save remotely collected data
- create, name, and describe data files
- convert data to graphs
- plot graphs from raw or processed data
- compare data from up to 12 graphs on the same screen
- change graphics in format, size, and position
- analyse points and areas of graphs
- annotate graphs with labels/text
- window in and amplify sections of a graph
- print out graphs or lists for reports
- create compatible files for output to spreadsheets, e.g. Lotus 1-2-3.

The PRONTO display is laid out in the form of a menu bar with pull down sub-menus activated by hot keys. The PRONTO application software can display graphs in four formats: line plot, max./min. plot, average plot, and cumulative plot. Help and error massages are also implemented.

The software PRONTO comes with a separate installation program, for example, a full IBM VGA screen driver is available with 640x400 colour resolution, and an auto detect option is also available.

The PRONTO Menu

The PRONTO main menu is as follows:
- HELP
- DATA FILE
- PLOT
- ANALYZE
- PRINTSCRN
- SET-UP
- ESCAPE.

The "HELP" will provide context sensitive help to the user.

The "DATA FILE" permits the user to retrieve data, file data, select data for subsequent analyse and review, and save processed data.

The "PLOT" provides a screen plot of user selected files. Up to 12 traces may be shown on the screen simultaneously.

The "ANALYZE" function provides the following additional features:
- zoom
- text annotation in graph
- statistical analysis
- change size and location of graph
- remove, or duplicate graph

- different presentation options
- notebook option.

The "PRINTSCRN" sends the screen layout to the printer.

The "SET-UP" function provides access the default configuration for the disc drives, serial ports, etc.

The "ESCAPE" function provides a exit route from any given instruction and permits access to DOS commands.

The two most important commands from the point of view of data presentation and analysis are "DATA FILE" and "ANALYZE". The first, "DATA FILE" has a quick plot mode under the file selection command which permits the user to quickly review, analyze, and print without searching through historical data. The "ANALYZE" function provides a complete suite of commands for the presentation of graphs, e.g. zoom on a user definable region or window, modification of graph layout, combination of several traces in one graph, determination of maximum, minimum, mean, and integral values over a user definable set of data, etc.

The PRONTO Technical Specifications

PRONTO requires a standard IBM-PC, or 100% compatible, with preferably a good video board, i.e. VGA. Below are listed the technical requirements and specifications for PRONTO.

Computers (examples):
IBM PC, XT, AT and 100% compatibles
IBM PS/2 models 30, 50 ,70
Toshiba 1000, 1100, 1600, 3100, 5200 laptops
RAM Memory: 512k
DOS: 2.11 or later
Video Board: graphics interface needed, Hercules, CGA, EGA, VGA or 100% compatibles
PRONTO Program Size: >300k
Graph:
Maximum Number of Traces on Screen: 12
Maximum Time from Left to Right Axis: 1500 days
Range of Values on Graph: -99999 to +99999
Time Resolution:
Minimum Time Span Across Graph: 1 second
Resolution of Left and Right Axis: 1 second
Resolution of Division on Time Axis: 0.1 second
Printers (examples):
Epson FX, MX, RX series
HP Laser Jet series II
HP Paint Jet
Mouse (examples):
Microsoft mouse
Logitech mouse

4. An Implementation

The overall performance of the RANGER-PRONTO configuration can best be described through some examples of the hardware operating in an unattended mode. The first involves the connection of a pair of RANGER's to a pair of neutron coincidence electronic packages (JSR-11's) which are connected to a neutron detector with the objective to detect the rapid passage of a item through the detector even when part of the electronics chain fails (monitor mode). The second is the detection a number of plutonium items in an unattended mode and the subsequent analysis of the data to obtain assay results (assay mode).

Monitor Mode

The monitor mode requires that each plutonium item passing through the detector is detected and logged, and that the chain from the neutron detector through the coincidence electronics to the data logger has a high level of redundancy. The hardware configuration consisted of the following:

- one set of ^{3}He thermal neutron detectors embedded in polyethylene
- use of the Amptek A-111 amplifier/discriminators
- half the neutron detectors receiving the high voltage from one neutron coincidence electronics package, the other half from a second package
- half the Amptek A-111 receiving the +5V from one neutron coincidence electronics package, the other half from a second package
- the two partial signals from each group of detectors and Ampteks added and duplicated for two separate neutron coincidence electronic packages
- connection of one RANGER data logger to each of the neutron coincidence electronic packages.

The neutron coincidence electronics package was set to recycle with a 5s measurement time. The existing model of the neutron coincidence electronics package used, i.e. JSR-11, requires about 8s to transfer data before resetting the counting cycle so the RANGER sample time was set to 1s thus producing a series of small measurement "plateau" each about 15s long.

Fig. 5 shows the tests performed:

- arrival and positioning of an item in the detector cavity at 10:36

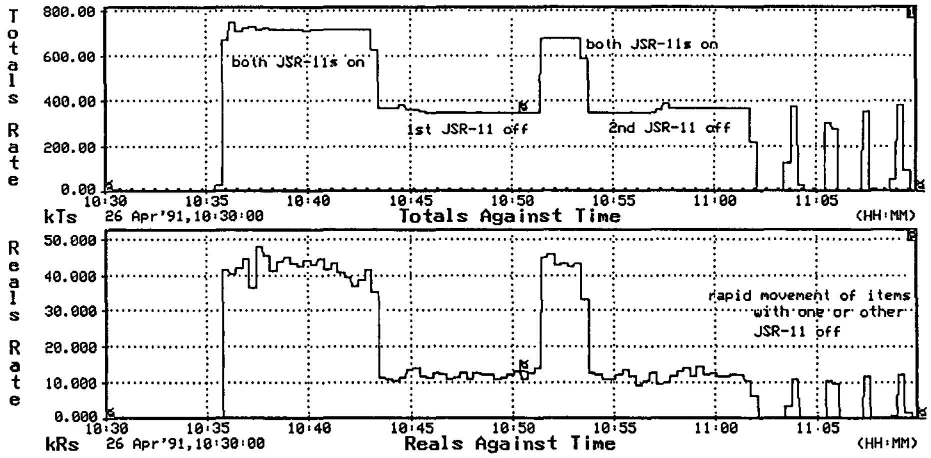

Fig. 5 The Totals and Reals Rates as a Function of Time for a Reduntant Detector Configuration

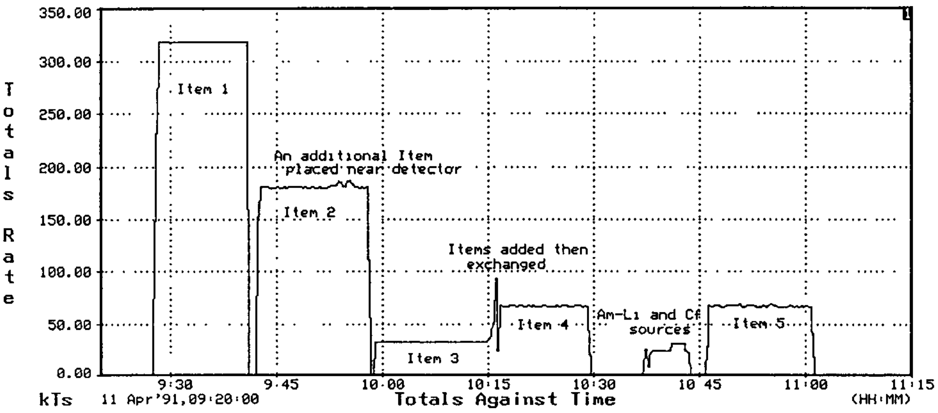

Fig. 6 The Totals Rates as a Function of Time for Five Plutonium Items

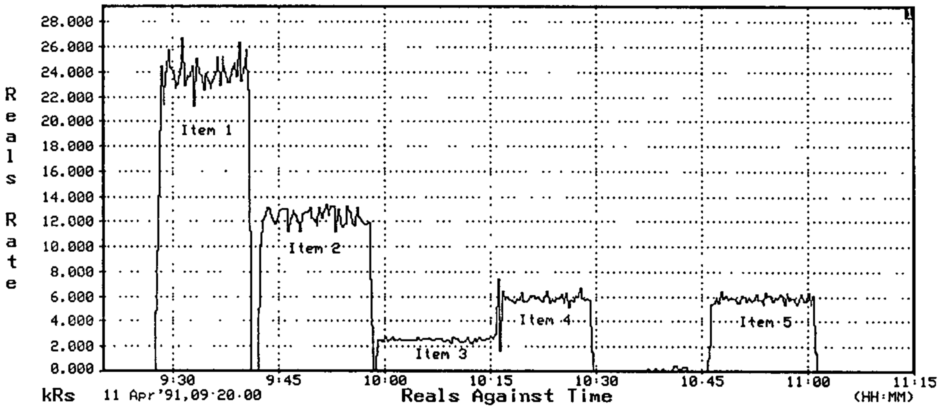

Fig. 7 The Reals Rates as a Function of Time for Five Plutonium Items

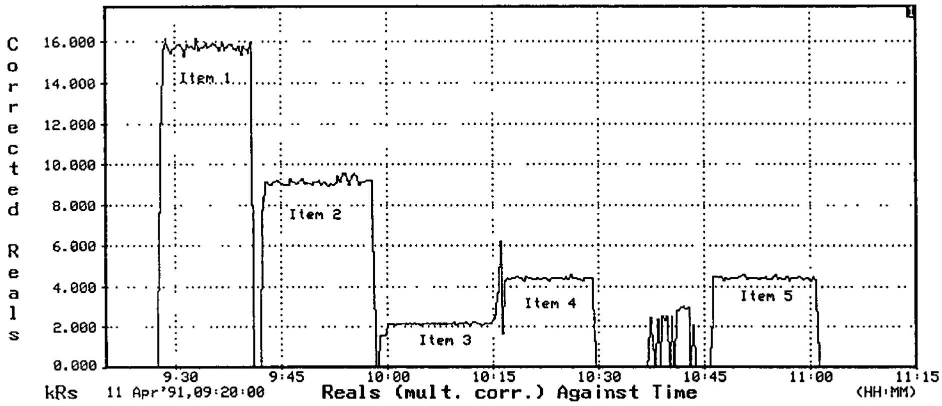

Fig. 8 The Reals Rates (multiplication corrected) as a Function of Time for Five Plutonium Items

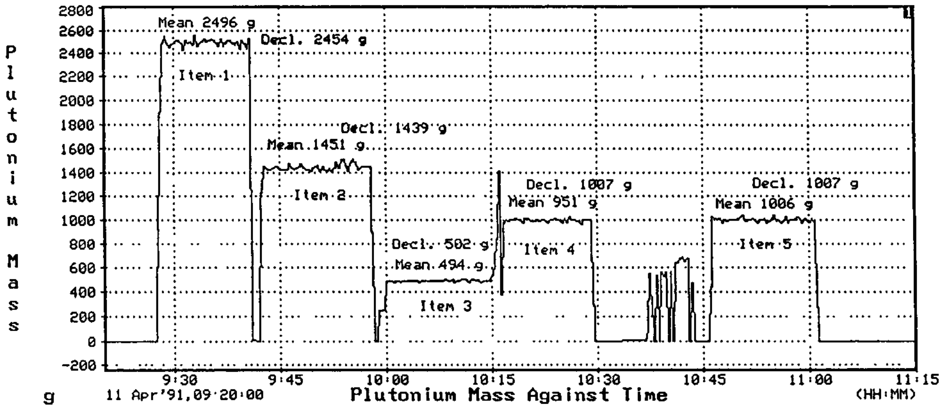

Fig. 9 The Plutonium Total Mass as a Function of Time for Five Plutonium Items

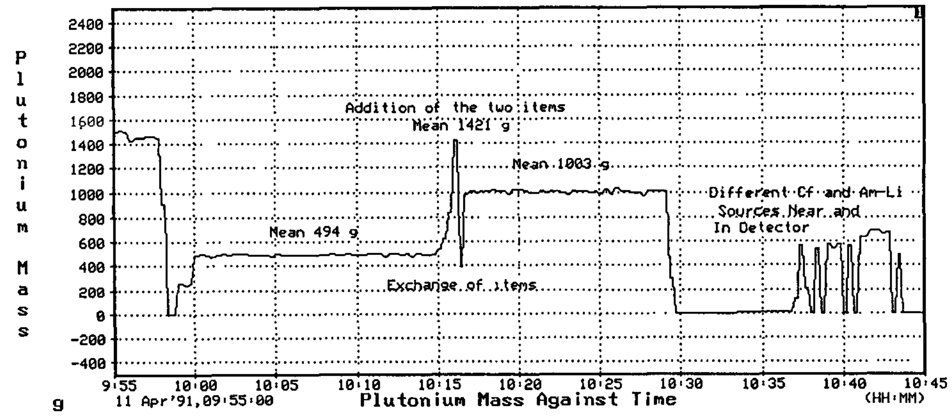

Fig. 10 The Totals and Reals Rates as a Function of Time

- failure of power supplies (high voltage and +5V) of first neutron coincidence electronics package at 10:43
- re-establishment of power supplies at 10:52
- failure of power supplies of second neutron coincidence electronics package at 10:54
- removal of item from detector cavity at 11:02
- rapid passage in both directions of item through the detector cavity with a power failure of one or other of the neutron coincidence electronics packages.

It can be clearly seen that the combination of neutron detector and coincidence electronics is sufficiently sensitive to the rapid passage of an item through the detector cavity and has a high level of redundancy. In addition the combination RANGER-PRONTO can be seen to be able to log and present the data in a practical and easy to interpret format.

Assay Mode

The assay mode is an addition to the monitor mode in that first data must be acquired in the monitor mode then an assay result must obtained off-line by the user with PUMASS. The assay mode thus requires that each plutonium item passing through the detector is detected and logged, and that the item remains in a correct and fixed position for a sufficient time to provide data for a complete assay. There are situations where the monitor and assay modes are combined to produce an assay system with monitoring functions included. The hardware configuration consisted of the a set of ^{3}He thermal neutron detectors embedded in polyethylene, Amptek A-111 amplifier/discriminators, and a neutron coincidence electronics package. The RANGER data logger is connected to the neutron coincidence electronic package.

Fig. 6 and Fig. 7 show the Totals and Reals rates acquired for a series of 5 different PuO2 powder samples ranging in mass between 502 g and 2454 g Pu. These Totals and Reals rates are already a product of the PUMASS analysis since they have already been corrected for the dead time, normalization (k=1), and background. The data logger was set to sample every 1s and the JSR-11 was set to recycle every 10s. The 5 different items can be clearly distinguished as can a series of tests with Am-Li and ^{252}Cf sources performed between 10:30 and 10:44. In addition with Item 2 a small perturbation can be seen in the plateau of the Totals rates corresponding to the passage of an addition plutonium item in the vicinity of the neutron coincidence counter. Another interesting feature is the perturbation noted between Item 3 and Item 4 corresponding to the introduction of both items in to the measurement cavity before Item 3 was removed.

Fig. 8 shows the multiplication corrected Reals rates corresponding to same time period as shown in the previous graphs. Fig. 9 shows the total plutonium mass again for the same time period. Again the PUMASS analysis program has been used to obtain the multiplication corrected Reals rates and, using existing calibration parameters for the neutron coincidence counter, the total plutonium mass. In Fig. 9 is clearly shown the declared plutonium mass and that obtain by using the standard averaging function in the analysis option of PRONTO.

Fig. 10 shows a zoom of a small portion of Fig. 9 where the addition of Items 3 and 4 can be clearly seen. It is possible even to perform an approximate assay during the 20-30s that the two items remained together in the measurement cavity. The presence of a "fictitious" plutonium mass for the Am-Li and ^{252}Cf is due simply to the allocation of a declared mass and isotopics to that particular period. It is however important in a future version of the PUMASS program to introduce a test of the ratio R/T in order to distinguish between different neutron emitters/1/.

5. Discussions

The RANGER-PRONTO combination coupled with the EURATOM configuration module and special software PUMASS provides both monitoring and assay functions for neutron coincidence data. The position of the Euratom Safeguards Directorate is that this solution provides one approach to the problem of unattended neutron coincidence data acquisition and subsequent off-line data analysis. The Euratom Safeguards Directorate views this solution as being particularly effective when:

- unattended neutron coincidence data acquisition is required
- either or both monitoring and assay functions are needed
- when immediate data analysis is not required, i.e. when the inspector only expects to service the data acquisition system every week or every month
- when simple neutron coincidence detector systems are installed, i.e. when no combined neutron and gamma-ray system is employed
- where a high level of redundancy is required, i.e. relatively low cost solution, battery backed-up components, etc.
- when subsequent off-line data analysis is acceptable,

i.e. when there are only a limited number of items passing through the detector cavity.

Areas where the Euratom Safeguards Directorate is concentrating its future developments for unattended measurement systems are:

- unattended operation of combined neutron and gamma-ray measurement systems
- networking data acquisitions systems to provide rapid data analysis and failure detection
- automatic recognition and analysis of events of safeguards importance.

Future areas of development for the RANGER-PRONTO configuration are as follows:

- improved user interface to PUMASS
- a new configuration module for the new computer controllable neutron coincidence electronics JSR-12
- use of R/T ratio to help automatically identify the different types of items being monitored
- use of T^2g to help automatically identity the measurement plateau for assay
- introduction of graphs for R/T, measurement statistics, and declared-measured differences
- completion of a detailed user manual for the specific application
- testing the connection of the data logger to a communications network.

6. Conclusions

This paper describes a simple, inexpensive, and flexible solution to the unattended mode acquisition of passive neutron coincidence measurement and monitoring data.

The solution consists of a dedicated commercial Data Logger connected to a commercial neutron coincidence electronics package.

Raw data from the Data Pack is automatically loaded in to a custom written software package and processed with time-specific data on the isotopic compositions to yield multiplication corrected Reals count rates for all items passing through the measurement cavity. The uncorrected and multiplication corrected Reals count rates are then interpreted, through user specified calibration functions, as $^{240}Pu_{eff.}$ and $Pu_{tot.}$ masses.

Both the raw data and the results from the Recovery Software, i.e. $^{240}Pu_{eff.}$, $Pu_{tot.}$ etc., can be examined in detail, as a function of time, through comprehensive graphics facilities thus providing a basis for the monitoring mode review.

One particular application was discussed where the Data Logger was installed for unattended mode monitoring on a passive neutron coincidence counter.

The solution of the Data Logger for the unattended mode acquisition and analysis of passive neutron coincidence measurements and the acquisition of the same type of data for monitoring purposes has proved to be a very versatile, low cost, user friendly solution.

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THE MEASUREMENT CAPABILITIES OF PHONID 3b

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1. ABSTRACT

The newest model of the PHONID series of nondestructive assay instruments is described. The measurement technique is active interrogation of the sample by a photoneutron source. The measured signal is the induced fission neutrons. The measurement samples are product materials from the nuclear fuel cycle such as uranium or plutonium oxide. Results from an evaluation using the PERLA calibration materials at the JRC-Ispra demonstrate that straight line calibration functions are adequate for most users. Long term stability of 0.2% was achieved in laboratory conditions.

2. INTRODUCTION

The PHONID series of photoneutron-based active-interrogation instruments have been used for inspections of low enriched uranium facilities in Europe. Previous work has documented 2-3% measurement uncertainties in field conditions/1,2/. Statistical analyses have indicated that these uncertainties are larger than those predicted by counting statistics/3/. The present work had two objectives. We reviewed the measurement procedures and the instrument components with the objective of reducing the measurement uncertainties. We also continued the evaluation/4/ of applying this technique to other isotopes.

3. INSTRUMENT

PHONID 3b is the most recent model of a family of nondestructive assay instruments designed and built by the Joint Research Center, Ispra, Italy/5,1,2,6/. PHONID 3b uses active interrogation of the sample with photoneutrons from an Sb(Be) source to induce fissions in fertile material. High energy fission neutrons are the assay response. The interrogating neutron spectrum has an average energy of 12keV and a narrow bandwidth. In particular, the neutron energies are below the fission barriers of the even-mass uranium and plutonium isotopes and they are above the large thermal cross-sections of the odd-mass isotopes. The fast-neutron detectors are 5cm diameter, cylindrical ^{4}He proportional counters. Energy discrimination is used to block the source (neutron and gamma-ray) components in the measured signal. The sample chamber contains no hydrogenous materials, consequently the neutron energy spectrum is not thermalized.

A new transfer container is used to transport the Sb sources separately from the rest of the hardware. It is a 540kg lead shield with interlocks that is ruggedized for transport. Figure 1 shows the transfer container on top of the PHONID 3b in the source loading position. The drawer underneath the PHONID 3b is part of the source unloading mechanism. During the source transfer activity, the transport container and PHONID 3b shield prevent any exposure to radiation from the Sb source.

4. SAMPLES

The PERLA calibration materials cover a wide range of properties of interest in NDA measurements/7/. The uranium samples of interest for this work are UO_2 powder in 4mm thick aluminum containers. The sample diameter is 87mm. The containers are 90, 150, or 300mm high as required by the different quantities of oxide. A minimum of 5 samples spanning a wide mass range (0.1 - 1.5 kg elemental uranium) from each of four different enrichments (20, 35, 60, & 92% ^{235}U) were measured. Three uranium metal samples and two UF_4 samples were also measured.

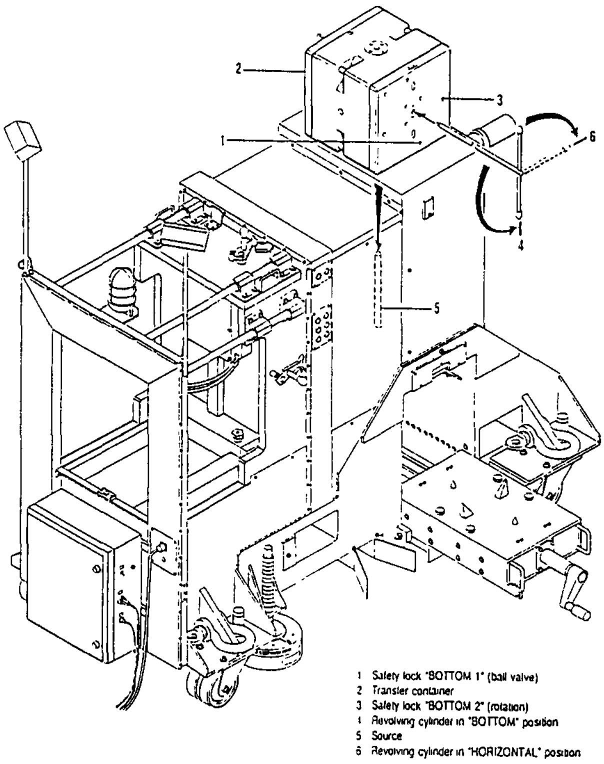

Figure 1 The PHONID 3b with the Sb source transfer container in the source loading position.

The plutonium oxide samples chosen for this evaluation are doubly contained in steel containers with a plastic bag between the two containers. Characteristics of the containers for the different mass ranges are given in table I. The smallest possible container is used for each quantity of oxide, except for a few special cases. These special cases use the large diameter containers for the small masses of plutonium. They are for the study of multiplication effects.

Table I. PuO_2 container characteristics

largest mass (g Pu)	sample diameter (mm)	container height (mm)	combined wall thickness (mm)
200	34.8	124	2.6
1000	58.3	204	4.1
2500	82.8	238	5.0

The elemental mass range of the plutonium samples was 0.5 - 2.5 kg. Three different burnups are represented (58, 60, & 70% ^{239}Pu).

5. DATA ANALYSIS - DETERMINATION OF MASS

The data reduction procedure is the same for all samples. This makes the PHONID 3b simple to use. The raw data consists of two count rates: the passive has the sample in the counting chamber but the photoneutron source is off, and the active has the sample in the chamber with the photoneutron source on.

1. A simple deadtime correction is made to all count rates. This correction is less than 4% for the largest samples of high burnup plutonium.

2. A background subtraction is performed to remove the passive response from the active measurement of the sample. This correction is negligible for most uranium samples.

3. A correction is made for the decay of the Sb in the photoneutron source. ^{124}Sb decays 1.16% per day.

4. Other corrections to the data may then be applied. This is an active area of research/8/ whose status will be summarized below. Our present recommendation is to use representative standards with respect to sample geometry and enrichment (no correction).

5. The corrected net active response is then related to the effective sample mass with a calibration function. A linear function is used whenever possible. The advantages of the linear function include easier application of corrections and cross calibration. The effective sample mass is computed from knowledge of the isotopic composition:

$$235_{effective} = 1.000\ {}^{235}U + .197\ {}^{238}Pu + .957\ {}^{239}Pu + .018\ {}^{240}Pu + 1.614\ {}^{241}Pu + .003\ {}^{242}Pu + .013\ {}^{241}Am + .028\ Cm$$

The coefficients in this formula are based on MCNP calculations/9,10/. $235_{effective}$ is analogous to the $240_{effective}$ defined for the thermal neutron counter assay of plutonium/11/.

6. DATA REDUCTION of the PASSIVE SIGNAL

If we make the same assumptions typically used for the analysis of thermal neutron coincidence counters/12/, the passive count rate can be expressed in the same way for the PHONID:

$$\text{passive count} = (1032\ n/s/g240)\ m_{240}\ M\ (1+alpha)\ \epsilon$$

where

1032 = spontaneous fission rate of ^{240}Pu

m_{240} = mass of ^{240}Pu (effective)

M = multiplication

α = (α,n) reaction rate / spontaneous fission rate

ϵ = average detection efficiency

7. MEASUREMENT RESULTS

First we present stability results, then we discuss the calibration data. The long term stability of the PHONID 3b is now better understood. A ^{252}Cf source was measured for 14 days in a series of passive counts each with 0.2% precision. The standard deviation of the ≈389 measurements was 0.2%. Repeated active measurements of three large mass 92% enriched UO_2 samples during 1 month agreed to better than 1%. The temperature variation of the PHONID 3b response is less than 0.1% per °C. However, we note that some applications have experienced more than 5°C daily variation.

The variation in the PHONID 3b response was measured along the sample axis with small sources in 2.5cm steps. The data were normalized by the maximum value and averaged from a height of 7.5 to 42.5cm above the floor of the sample chamber. The passive response was 0.954 ± 0.038 (measured with a small ^{252}Cf source), while the active response was 0.926 ± 0.065 (measured with a small Pu source). The effect of a 2cm radial offset for a 1kg UO_2 sample was less than 1% in the active response. However, density effects can be at least 5%. Table II lists the results of a measurement sequence during which the density of the sample contents was intentionally altered. We recommend that all users tap the sample containers on a hard surface before measurement if they would like to reduce this effect.

Figure 2 plots the corrected net active response as a function of elemental mass for all of the samples measured in this campaign. Results from uranium oxide, metal and fluoride are shown as well as results from plutonium oxide samples. These data were taken with an Sb source intensity of ≈2Ci. The measurements were performed over a 4 month period. We draw your attention to the linear behaviour of each material type (except the uranium metal). Geometry variations caused additional effects in the metal samples. Typical counting precision in the corrected net active response was 0.7% for the uranium samples but increased to 3% for the plutonium samples as a consequence of the passive background. A stronger Sb source (10 Ci has been traditional) or longer count times would yield improved counting precision.

Table II. Density and Position Effects in the PHONID (1kg UO_2 sample)

position	date/time	net active rate (s^{-1})
remove from store put into PHONID	5-mar 9:15	107.91 ± 0.46
turn upside down	5-mar 9:34	110.91 ± 0.63
turn upright	5-mar 9:48	111.14 ± 0.63
radial offset 2cm	5-mar 10:00	110.50 ± 0.63
tamp sample (increase density) radial offset 2 cm	5-mar 10:11	105.63 ± 0.63
center sample	5-mar 10:21	106.72 ± 0.61

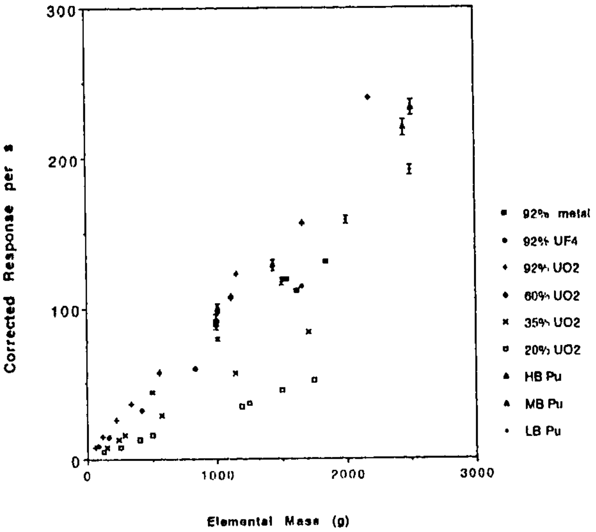

Figure 2 The corrected net active response per second of the PHONID 3b as a function of special nuclear material mass. Data from four different uranium enrichments and three different plutonium enrichments are shown.

Table III lists the uranium measurement data. For the 92% enriched uranium, samples 10-16 are oxide, 21 & 22 are fluoride, and 22-24 are metal. Samples 30-34 are 60% enriched UO_2, samples 40-45 are 35% enriched UO_2, and samples 50-58 are 20% enriched UO_2. Note that samples 14, 15 and 16 were measured twice, ≈1 month apart. After the decay correction is made, the results agree to better than 1% for these three samples.

One possibility for the calibration of this instrument is a straight line fit to the data as a function of ^{235}U mass. Figure 3 shows the results for the lowest enrichment, 20% UO_2. Table IV lists the coefficients for weighted, least squares, straight line fits to the four sets of UO_2 samples in this evaluation. The small samples not included in the linear fitting procedure are indicated in column five.

A second possibility for the calibration of this instrument is under study/8/. Figure 4 shows the corrected net active response as a function of the uranium mass times the enrichment raised to the 0.8 power. All of the data from four enrichments

Table III. PHONID 3b Measurement Data for UO_2 Samples

ID	U mass (g)	^{235}U mass (g)	time of measurement	passive rate (s^{-1})	active rate (s^{-1})
10	55.57	51.4	16-jan 9:42	1.30 ± 0.05	9.05 ± 0.08
11	111.16	102.7	16-jan 10:08	1.35 ± 0.05	16.32 ± 0.13
12	222.28	205.4	6-mar 11:09	1.18 ± 0.11	15.61 ± 0.07
13	333.41	308.1	8-mar 10:04	1.12 ± 0.11	21.34 ± 0.10
14	555.67	513.6	6-mar 10:45	1.19 ± 0.11	33.32 ± 0.16
14	555.67	513.6	9-apr 9:49	1.23 ± 0.07	23.14 ± 0.17
15	1111.37	1027.1	5-mar 10:21	1.28 ± 0.11	62.10 ± 0.35
15	1111.37	1027.1	9-apr 10:10	1.25 ± 0.07	42.32 ± 0.23
16	1667.03	1540.7	6-mar 10:27	1.51 ± 0.12	89.06 ± 0.33
16	1667.03	1540.7	18-mar 10:43	1.50 ± 0.12	78.31 ± 0.31
20	2183.33	2017.8	18-mar 10:21	21.25 ± 0.21	139.12 ± 0.42
21	1152.14	1064.8	18-mar 9:54	11.10 ± 0.15	71.11 ± 0.29
22	1849.	1722.	18-mar 11:01	1.10 ± 0.10	64.82 ± 0.28
23	1544.	1438.	18-mar 11:52	0.86 ± 0.09	58.92 ± 0.27
24	1622.	1510.	18-mar 16:01	1.29 ± 0.11	55.83 ± 0.26
30	1662.58	998.6	4-mar 14:34	1.40 ± 0.08	67.21 ± 0.25
31	831.29	499.3	4-mar 14:56	1.17 ± 0.11	35.65 ± 0.13
32	415.76	249.7	4-mar 15:37	1.15 ± 0.08	19.66 ± 0.09
33	166.50	100.0	8-mar 11:09	0.93 ± 0.10	9.05 ± 0.11
34	83.27	50.0	8-mar 11:45	1.03 ± 0.10	5.63 ± 0.08
40	1710.89	598.8	4-mar 9:36	1.23 ± 0.06	49.95 ± 0.16
41	1147.00	401.5	4-mar 10:20	1.28 ± 0.07	34.07 ± 0.12
42	573.41	200.7	4-mar 11:35	1.19 ± 0.11	17.91 ± 0.13
43	235.90	82.6	5-mar 10:40	1.09 ± 0.07	8.29 ± 0.06
44	286.70	100.4	8-mar 11:28	0.93 ± 0.10	9.51 ± 0.11
45	143.50	50.2	6-mar 15:27	1.05 ± 0.07	5.67 ± 0.04
50	1751.51	348.2	8-mar 9:31	1.29 ± 0.11	29.98 ± 0.19
51	1501.02	298.2	28-feb 11:14	1.11 ± 0.10	28.31 ± 0.08
52	1251.11	248.7	8-mar 9:48	1.14 ± 0.11	21.50 ± 0.16
53	1193.53	237.3	8-mar 10:52	1.34 ± 0.11	20.53 ± 0.16
54	500.46	99.4	28-feb 12:42	1.11 ± 0.10	10.57 ± 0.03
55	500.49	99.5	5-mar 13:55	0.95 ± 0.07	9.83 ± 0.06
56	400.65	79.7	5-mar 11:25	0.93 ± 0.07	8.25 ± 0.06
57	250.19	49.7	6-mar 12:01	1.10 ± 0.10	5.72 ± 0.03
58	125.08	24.9	5-mar 12:00	0.94 ± 0.07	3.60 ± 0.02

is within 5% of a cubic fit in this attempt to correct for the self shielding. We note that this correction should also depend on the oxide density and the sample diameter.

Tables V & VI list the plutonium oxide measurement data and results, respectively. The 3 burnups are each fit with a straight line. This fit was not weighted least squares to all the data points. However this preliminary evaluation indicates that the measurements are consistent with a straight line calibration. We will continue this evaluation with a larger sample set and measurements with better precision.

For this presentation, we used the passive data from the plutonium samples to solve for the detection efficiency. The values for α are calculated for pure oxide and the multiplication values are from thermal counter measurements made a year earlier. Perhaps the assumptions used to derive the formula are not satisfied sufficiently.

8. SUGGESTED DATA ACQUISITION PROCEDURE

Our measurement experience with the PHONID 3b has led us to propose a different treatment of the data than that currently in use by inspectors. In addition to the analysis discussed above, we suggest the following for consideration. The normalization that has been traditionally used adds a component of uncertainty to the result. With a stable instrument, verification of the response without performing a correction will yield a result with a smaller measurement uncertainty. We propose that the normalization required when the source is

Table IV. Linear Calibration Coefficients for UO_2
Corrected Response = A + B(mass$_{235}$)

Enrichment	A ± ΔA	B ± ΔB	Reduced Chisquare	Fit range
92.42	7.52 ± 0.26	0.09669 ± 0.00039	2.7	> 300g
60.06	4.81 ± 0.32	0.10992 ± 0.00066	0.9	> 200g
34.98	1.26 ± 0.12	0.13880 ± 0.00050	0.7	> 49g
19.87	1.035 ± 0.099	0.14653 ± 0.00070	1.9	> 23g

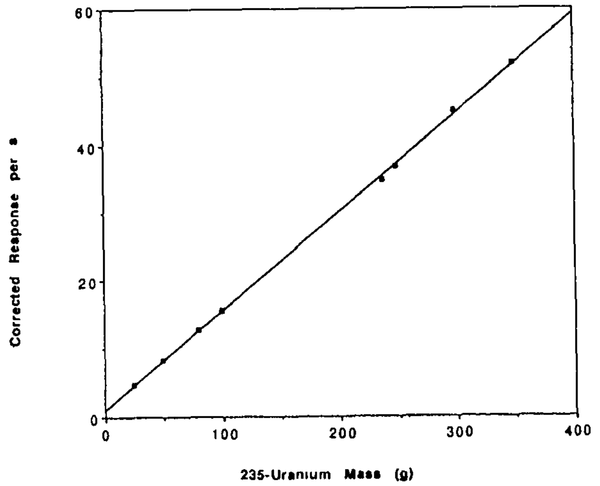

Figure 3 The corrected net active response per second as a function of the ^{235}U mass for the 20% enriched samples. The solid line is a least squares fit to the data.

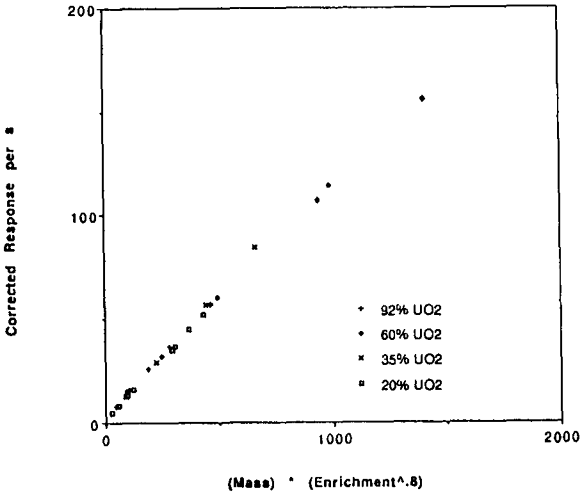

Figure 4 The corrected net active response per second as a function of the uranium mass times the enrichment raised to the 0.8 power. Data from four different enrichments fall on one line.

Table V. PHONID 3b Measurement Data for PuO_2 Samples

ID	tag mass 235$_{effective}$	tag mass 240$_{effective}$	time of measurement	passive rate (s^{-1})	active rate (s^{-1})
1	1431.0 g	958.4 g	12-mar 11:00	5301.8 ± 1.9	5414.1 ± 1.9
2	839.5	562.2	13-mar 10:22	2962.7 ± 1.2	3028.1 ± 1.2
3	582.2	389.8	14-mar 9:40	2076.9 ± 1.2	2123.8 ± 1.2
4	582.5	390.1	14-mar 10:50	1987.8 ± 1.2	2033.0 ± 1.0
5	1501.8	895.0	13-mar 14:33	4946.7 ± 1.8	5064.4 ± 1.6
6	604.7	360.4	13-mar 16:00	1919.6 ± 1.1	1970.7 ± 1.0
7	1524.5	671.4	11-mar 11:13	2766.2 ± 1.2	2866.6 ± 1.2
8	1220.5	537.5	11-mar 15:50	2170.9 ± 0.9	2254.0 ± 1.0
9	916.2	403.5	12-mar 9:57	1590.0 ± 0.9	1651.2 ± 0.9
10	613.7	270.28	12-mar 11:53	1026.0 ± 0.5	1067.6 ± 0.4
11	609.3	268.35	13-mar 12:00	1066.0 ± 0.7	1112.2 ± 0.4
12	305.9	134.73	11-mar 14:12	508.4 ± 0.2	531.7 ± 0.4
13	306.1	134.78	14-mar 10:00	489.9 ± 0.4	512.7 ± 0.3

changed be considered separately from the instrument stability check.

When a new ^{124}Sb source is installed, the instrument must either be recalibrated or a normalization measurement can be used to relate the new response to the previous calibration. A demonstration of the validity of the normalization measurement has yet to be performed.

After the instrument with a new source has been calibrated, we suggest the following procedures. Our proposal is an instrument control procedure that was evaluated previously/13/.

1. Perform a normalization check monthly or before and after each inspection. For example, if the average and standard deviation of fifteen 1000s measurements agree with the expected values, then

Table VI. PHONID 3b Results for PuO_2 Samples

ID	$1 + \alpha^a$	$M(\alpha,R,T)^b$	measured mass $235_{effective}$	measured to tag ratio: $235_{effective}$	efficiency
1	1.915	1.168	1431.0 ± 34. g	1.000	.00247
2	1.915	1.129	839.5 ± 22.	1.000	.00240
3	1.915	1.109	611.5 ± 22.	1.050	.00246
4	1.915	1.12	589.0 ± 22.	1.012	.00233
5	1.905	1.166	1501.8 ± 31.	1.000	.00248
6	1.905	1.106	604.7 ± 18.	1.000	.00248
7	1.470	1.182	1507.6 ± 25.	0.989	.00234
8	1.470	1.175	1240.6 ± 20.	1.016	.00230
9	1.470	1.165	912.5 ± 19.	0.996	.00225
10	1.470	1.136	607.8 ± 9.	0.990	.00222
11	1.470	1.129	688.0 ± 12.	1.129	.00233
12	1.470	1.102	323.7 ± 7.	1.058	.00226
13	1.470	1.099	326.7 ± 7.	1.067	.00219

a) calculated for pure oxide from isotopics of Mar-91
b) from Neutron Coincidence Counter measurements made 29-Nov-89

assume the instrument is functioning correctly. If they do not agree, repair the instrument.

2. Perform two daily background counts (perhaps 100s each) with the photoneutron source on and off. Compare with historical values.

3. Perform a daily bias check. Perhaps an assay of a "known" item for 300s. In this situation "known" means stable but not necessarily traceable to something else.

4. Tap the sample to be measured firmly on a hard surface to remove excess air, if it is a powder. Otherwise you can expect erratic error behaviour of the order of 0-5%.

5. Perform a passive count of the sample (for a 100s count, 1 sigma is ≈0.1c/s)

6. Perform an active count of the sample (choose a count time long enough for 1 sigma to be ≈0.1s)

A further proposal for consideration is that while the instrument is idle, perhaps the computer could monitor the background. If it exceeds a user specified value, print the result and the time.

9. CONCLUSIONS

Previous work/2/ reported on the short term stability, these results show long term stability. The stability of the PHONID 3b response is shown to vary <0.2% in the pre-PERLA reactor building. Two parameters have been identified as causing some of the past "stability" effects in the field. A temperature dependant response and density variations in the assay samples both could have caused >1% effects. These factors should be controlled in future in-field use.

The preliminary efforts to reduce the number of required calibration materials look very interesting. Straight line calibration functions for the mass ranges of interest provide a solid basis for the study of correction factors. Successful cross calibration of four instruments and an enrichment correction could have a significant impact in how this instrument would be utilized for inspections.

We plan to explore the possibility of combining the PHONID measurement results with NCC results from the same sample. Independent measurement of the fissile and fertile components of Pu samples would be a powerful inspection tool that might reduce the need for DA.

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SOME RESULTS IN THE MONTE CARLO SIMULATION OF PHONID-3B

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Abstract

The Monte Carlo Code MCNP from Los Alamos (Ref. 1) is used for the analysis of the physical performance of the PHONID-3B instrument. The main problems for this purpose were the normalization of calculated count-rates on measurements and the need for too long calculation times for statistically reliable results. We present solutions to these problems and describe eventually the result of a parameter study on PHONID.

Summary

1. Normalization Problem

The problem of normalizing MCNP-calculated count-rates on PHONID-measured count-rates is due to the fact that PHONID uses He-4 filled counting tubes and electronic pulse height discrimination. A neutron of energy E[MeV] leads to a count if in an elastic scattering with a 4He atom the energy T[MeV] transferred to the He is sufficient for the production of an ionization pulse of a height H[Volt] larger than the level D[Volt] set by the discriminator.

We construct a procedure for simulating this counting mechanism which provides us with a link (via "Least Square Fit" of some free parameters) between calculated count-rates (based on the energetics of elastic scatterings) and measured count-rates (as functions of the discriminator level).

2. Reduction of computer time

To obtain statistically reliable results for the calculated count-rates (which are eventually to be compared to the measured count-rates) we need a large number of (neutron) histories of the order of 10^5 to 10^6 (starting from the Sb-Be Source) which takes about 2 to 3 hours on our AMDAHL-mainframe computer.

To reduce this forbiddingly large calculation time to make parameter studies or routine calculations feasible we use the option of the "Surface Source" offered by the MCNP-Code. We surround the sample region in the irradiation chamber of PHONID by a large enough cylindrical surface F, start for example 300.000 Histories from the SB-Be source and write on a "Surface File" the positional data of all those neutrons crossing F from the outside. For all further calculations neutrons are then started from F by reading the "starting data" from the "Surface File". In this way the computation time comes down to 5-10 minutes and makes routine work feasible .

3. Applications

As applications of MCNP we describe

- the production of the Be-neutrons due to the Gamma-flux(in the Be-cylinder) generated by the Sb-capsules.
- normalization of calculated countrates on experimental data,and
- the results of a parameter study performed on PHONID giving the countrate as a function of sample-radius, -height, and -enrichment.

A) Evaluation of the (Sb,Be)-(γ,n) Neutron Source

The (Sb,Be)-neutron source Q of PHONID (see Fig. 1) is made of a Be-cylinder containing two small Sb-capsules.

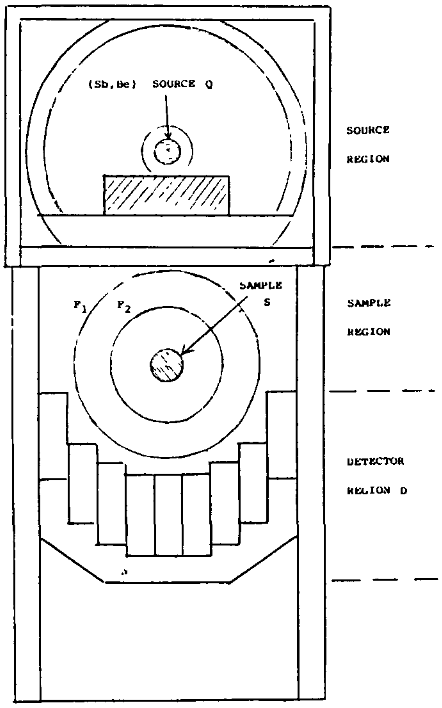

Fig. 1: Horizontal cross-section of PHONID-3B.

The Be-cylinder was divided by a set of horizontal planes in 10 equal (cylindrical) volume-elements $\Delta V_i(i=1,\dots,10)$. Gammas were then started from the two Sb-capsules (with equal probability). The number of neutrons ΔN_i (per decay of Sb-nucleus) produced in ΔV_i is then obtained by evaluating the integral (tally F4 modified by a dose-function):

$$\Delta N_i = \int_{V_i} dv \int dE \phi_\gamma(E) \Sigma_{\gamma,n}(E) \qquad (1)$$

where

$\Phi(E)$ is the energy dependent gamma flux in ΔV_i, and $\Sigma_{\gamma,n}(E)$ is the macroscopic (γ,n) cross section of Be.

Fig. 2 gives ΔN_i as a function of i. The total number of neutrons produced in the BE-cylinder per decay of one Sb-nucleus turned out to be $1.113*10^{-4}$ neutrons which means that an

-Sb-activity of 1 Curie corresponds to a neutron Source of $4.118*10^6$ (neutrons/sec).

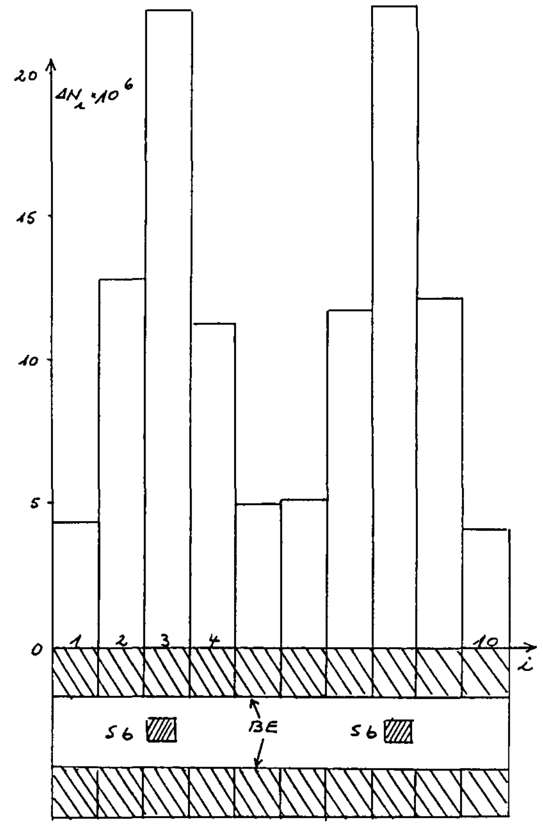

Fig. 2: ΔN_i is the number of (γ,n)-source neutrons produced by the Sb-gammas in volume element ΔV_i of the BE-cylinder (i=1,2....10).

B) Use of surface sources

a) Direct Monte Carlo run

The general situation is shown in Fig.1. Neutrons start from the Be-Source Q into the irradiation chamber, enter the sample S under investigation (to produce fissions) and eventually produce counts in the detector region D. It turned out that to avoid too large fluctuations in the quantities of interest (e.g. total number of fissions in the sample, counts produced in D etc.) a very large number of neutrons has to be started from the Be-source Q: one such run for 3*10E+05 source neutrons takes roughly 100 minutes (on the Amdahl main frame).
This computation time is obviously too long to do any parameter study or routine calculations.

b) Generation of first surface source

To reduce the long computation time we use the MCNP-option of the surface source:

- the sample region is enclosed within a cylinder F1 of a large enough radius (see Fig. 1)

- n (3.0*10E+05) neutrons are started from the Be-source Q. All source neutrons crossing F1 for the first time from the outside are stopped and their positional data (position, velocity, weight ...) are stored on a (surface-) file SF1.
This step needs about 80 minutes. Roughly 4.0*10E+04 histories cross F1 (only 13% of all the source neutrons enter the region within F1).

- this surface file SF1 is then used for each following calculation as a neutron source by scanning the complete file SF1 and starting the stored neutrons from F1 into the interior of F1.
Each MCNP-run using these 4.0*10E+04 surface neutrons as a neutron source takes about 15 to 20 minutes.
Note that each of these (surface source) calculations corresponds in reality to taking 3.0*10E+05 source neutrons from the Be-source.

c) Generation of the second surface source

The enclosing cylinder F1 is chosen such, that all possible container-configurations fit within F1. For small samples S this results again in too many neutrons from F1 missing S and only a fraction of all F1 source neutrons will enter the sample.

For this reason we introduce a second cylinder F2 surrounding S closely. Now we create a second surface file SF2 by starting the neutrons from SF1 and storing the positional data of all those neutrons crossing F2 for the first time from the outside.

Of the 4.0*10E+04 neutrons from F1 about 17000 cross F2. The generation of SF2 from SF1 takes about 10 minutes and the successive calculations using SF2 as a neutron source need about 7 minutes. Eventually we have achieved that a computer run for 300.000 source neutrons is done in 7 minutes.

We repeat the relevant data:

- less than 10% of all Be-source neutrons enter the sample

- the generation of the first surface file SF1 from Q takes 80 min. (300.000 source neutrons from Q give 40.000 neutrons crossing F1)

- the generation of the second surface file SF2 from SF1 takes 15 min. (40.000 neutrons from SF1 give 17.000 neutrons crossing F2)

- a MCNP-production run from SF2 takes 5 minutes and corresponds to a real case of 300.000 original source neutrons.

C) Simulation of the "Discriminator threshold" in MCNP-Calculations.

The discriminator level Do (measured in Volt) serves to suppress (count-)pulses with a pulse height H smaller than Do. All pulses with H > Do are counted.

The following argument transforms this mechanism into an MCNP recipe:

- the pulse height H of a (count-)pulse is proportional to the amount of ionization I produced by a recoil α-nucleus in an elastic neutron-scattering event

- this amount of ionization I is proportional to the recoil energy T given to the α-particle in the (elastic-)scattering.

- The pulse height H is therefore proportional to T; and to the pulse height level Do (discriminator threshold) corresponds a certain recoil energy threshold To

- Therefore: Counts are produced by only those elastic scatterings in which the recoil energy T is larger than To.

Consider a neutron of energy E making an elastic scattering with a He-nucleus. We denote by $P(E \rightarrow T)dT$ the probability that the recoil energy T (transferred to the α-particle in this elastic scattering) is in the interval dT around T.

The probability that this elastic scattering event will lead to a count is then given by (according to the above mentioned constraint):

$$P(E/To) = \int_{To}^{\infty} P(E \rightarrow T)dT \qquad (2)$$

The total countrate C is then obviously to be evaluated by

$$C(To) = \int dE\ \phi(E)\ \Sigma_s(E)\ P(E/To) \qquad (3)$$

$\Phi(E)$ is the total neutron flux at energy E averaged over the total counter volume

$\Sigma_s(E)$ is the macroscopic elastic scattering cross section of the total counter volume.

Note that the integral in C(To) can be directly evaluated in an MCNP-run if P(E/To) (as a dose function modifying tally F4) is given.

To construct $P(E \rightarrow T)$ we recall that the recoil energy T in the Laboratory-system is given by

$$T = E \cdot \frac{(1 - \alpha)}{2} \cdot (1 - \cos\varphi) \qquad (4)$$

where

ϕ is the scattering angle of the neutron in the CM-System and

$$\alpha = \left(\frac{M - m}{M + m}\right)^2 \qquad \begin{array}{l}\text{M:mass of target nucleus}\\ \text{m:mass of neutron}\end{array}$$

The distribution for $\cos\varphi = \mu$ of the scattered neutron (in the CM.-system) is usually given in the form:

$$\sigma_s(E, \mu) = \sigma_s(E)\sum_{l=0}^{\infty} \frac{2l + 1}{2} f_l(E)P_l(\mu) \qquad (5)$$

$$= \sigma_s(E)\ P(E \rightarrow \mu) \qquad (6)$$

Then we have:

$$P(E \rightarrow T)dT = P(E \rightarrow \mu)d\mu \qquad (7)$$

and, using (3):

$$P(E \rightarrow T) = P(E \rightarrow \mu)\ \frac{2}{E(1 - \alpha)} \qquad (8)$$

Combining (1) and (4) eventually gives us

$$P(E/To) = 0 \qquad \text{if } To > Tmax(E)$$

$$P(E/To) = \sum_{l=1}^{\infty} \frac{2l + 1}{E(1 - \alpha)} f_l(E) \int_{-1}^{\mu(To)} P_l(\mu)d\mu \qquad \text{if } To < Tmax(E) \qquad (9)$$

Tmax = Tmax (E) = E(1-α) is the maximal energy transfer (recoilenergy) in an elastic scattering.
This relation says that all neutrons with an energy E smaller than

$$E_0 = To/(1-\alpha) \qquad (10)$$

are not able to transfer at least the recoil energy To and, therefore, will never lead to a count.

Relation (8) therefore implies, that

- neutrons with an energy E below Eo do not produce any counts
- if $\Phi(E)\Sigma_s(E)\ \Delta E$ is the total number of elastic scatterings by neutrons with an energy in ΔE, then P(E/Eo) is that fraction of these scatterings which produce a count.

P(E/Eo) is tabulated as a function of E and Eo and used as the dose function modifying tally F4 in expression (2). (We use P(E/Eo) instead of P(E/To) in (2)).

For a practical application of (8) the relevant data for f_l (E) in (2) were taken from ENDFB/VI.

A simple method to normalize MCNP Calculations on measurements

To use MCNP Calculations for predictive purposes (e.g. generation of calibration curves) we have first to "normalize" the calculations on corresponding measurements.
We start from a fixed experimental configuration (given neutron source, high voltage etc.) and measure the counting rate C_{EX} as a function of the discriminator threshold D:

$$C_{EX} = C_{EX}\ (D) \qquad (11)$$

Fig. 3. shows an actually performed experiment using a CF-point source of unknown strength. Note that due to several reasons the measurements are meaningful only within a

certain interval $\{D_0, D_1\}$ and cannot be extrapolated into the range of very small thresholds $D \sim 0$.

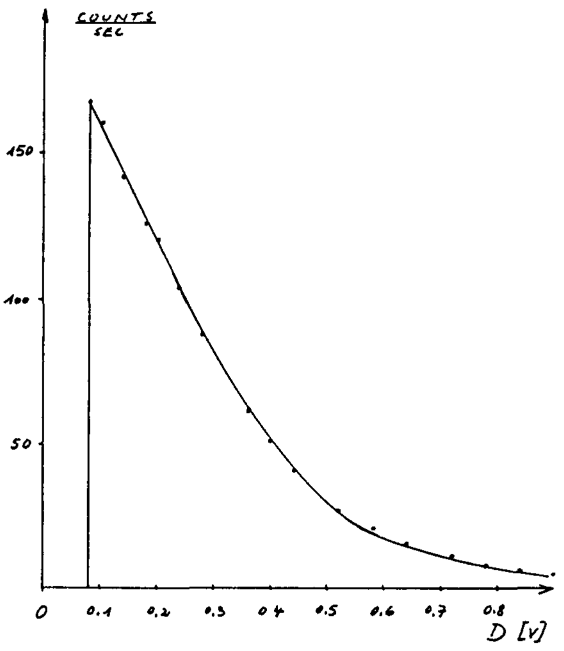

Fig. 3: Counting rate as a function of the discriminator threshold for a CF-point source in the center of the irradiation chamber.

MCNP calculations of the counting rate C_{CA} are then made for the same experimental configuration as a function of the energy-threshold E (for the same source-configuration but for a unit source strength: one source neutron emitted per sec.)

$$C_{CA} = C_{CA}(E) \qquad (12)$$

Fig. 4 shows the result of such a calculation for the experiment corresponding to Fig. 3. Note that the calculations may be performed down to zero-threshold and up to an upper level where all neutrons are cutoff.

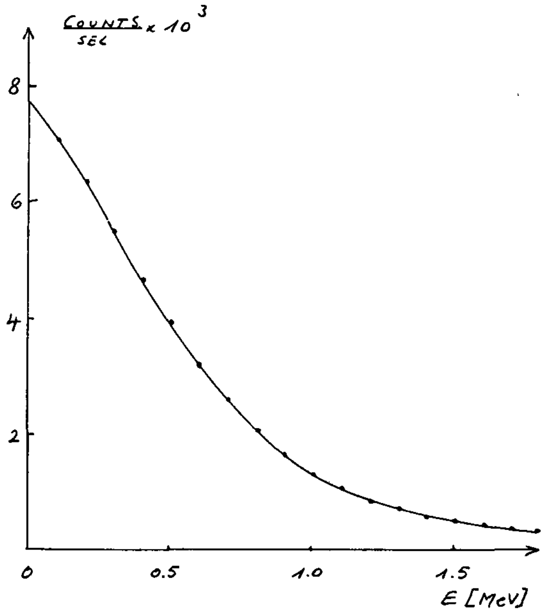

Fig. 4 : Calculated countrate as a function of the energy threshold E for the experimental setup of Fig. 3.

The "normalization" of C_{CA} on C_{EX} consists now in the problem to find

- an interval $\{E_0, E_1\}$ and
- parameters α, β such that (see Fig.5)

the following coincidences are "as good as possible":

$$D_0 = \alpha E_0 \qquad (13)$$

$$D_1 = \alpha E_1 \qquad (14)$$

and

$$X_0 = C_{EX}(D_0) = \beta C_{CA}(E_0) = \beta C_0 \qquad (15)$$

$$X_1 = C_{EX}(D_1) = \beta C_{CA}(E_1) = \beta C_1 \qquad (16)$$

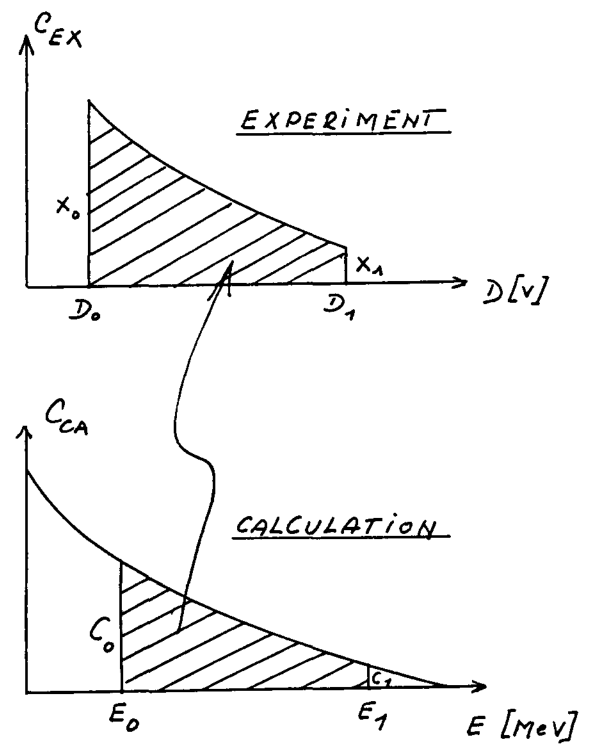

Fig. 5: Principle of normalizing MCNP-calculations on experimental values.

The condition "as good as possible" is formulated mathematically in terms of "Least Squares" and takes the form:

Find the minimum of

$$Q(\alpha, \beta) = (\alpha E_o - D_o)^2 + (\alpha E_1 - D_1)^2 + (\beta C_o - X_o)^2 + (\beta C_1 - X_1)^2 \qquad (17)$$

with respect to α and β.

As on the other hand we should have

$$\alpha = (E_0 D_0 + E_1 D_1)/(E_0 E_0 + E_1 E_1) = \alpha(E_0, E_1) \qquad (18)$$

$$\beta = (C_0 X_0 + C_1 X_1)/(C_0 C_0 + C_1 C_1) = \beta(E_0, E_1) \qquad (19)$$

we transform (7) into a minimum problem with respect to $\{E_o, E_1\}$:

$$Q(E_0, E_1) = (\alpha E_o - D_o)^2 + (\alpha E_1 - D_1)^2 + (\beta C_o - X_o)^2 + (\beta C_1 - X_1)^2 \qquad (20)$$

The actual "normalization " procedure therefore consists in checking all possible pairs $\{E_o, E_1\}$ and to find that pair $\{E_o^*, E_1^*\}$ which makes Q to a minimum.

α^* and β^* are then given by (8) and (9).

This procedure applied to the situation given in Fig.3 and 4 resulted in $E_o^* = 0.19$, $E_1^* = 2.38$ and gave eventually:

$\alpha^* = 0.41$, $\beta^* = 26.3$ corresponding to a source strength of $S_{EX} = 0.26.10^5$ (n/s).

Fig. 6 shows the agreement between the fitted calculated countrate and the experimentally measured countrate.

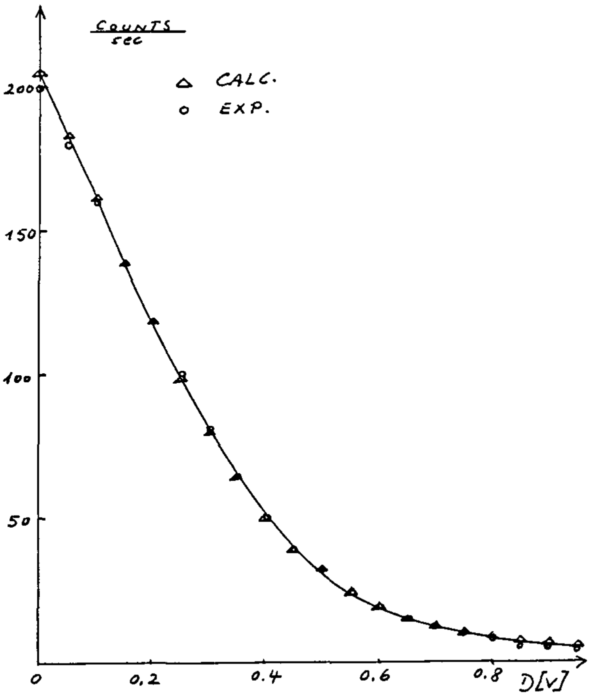

Fig. 6 : Result of normalizing the calculated values of Fig. 4 on the measurements of Fig. 3.

For a detailed investigation of the physical performance of PHONID-3B an extended data base was created by MCNP-calculations of various quantities in function of sample height H, sample-radius R and U235-enrichment E (for cylindrical samples containing UO_2 of density $\rho = 2.0$ [gr/cm^3]).

Some results are shown in the Figures (7) to (11).

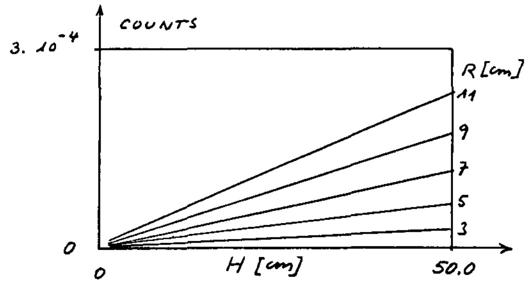

Fig. 7: gives the mean number of counts (per BE-source neutron) as a function of the sample height H.
(Sample radius R as parameter; U235-enrichment E=90%)

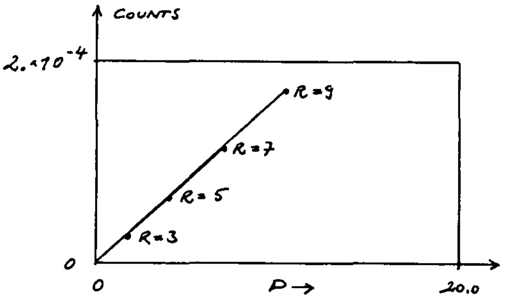

Fig. 8: gives the same data as in Fig.1 (mean number of counts per BE-source neutron) now as function of the quantity $P = P(R,H) = \text{const. } R^{1.6} H$.

Note the surprising effect, that all the curves fall on one line (for this one enrichment value E=90%).

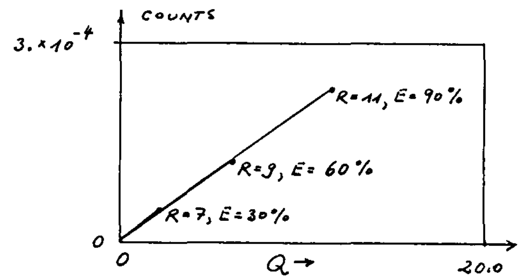

Fig. 9: shows again the mean number of counts (per BE-source neutron) as a function of the quantity
$Q = Q(R,H,E) = \text{const.}*R^{1.6}*H*E^{0.8}$ for different sample radii R and enrichment values E.

The very interesting effect is again the falling together of all the individual curves (almost) on one line.

In general we may try to fit the response in the form

$$C \sim R^{\alpha}\ E^{\beta}\ H^{\gamma} \quad (\alpha, \beta, \gamma = \text{const.})$$

There might be better values for the exponents of R, E and H to bring the individual curves closer together. The essential point however is just the fact that they can be brought together on one (calibration-) curve which then holds for a wide range of values for R,H and E.

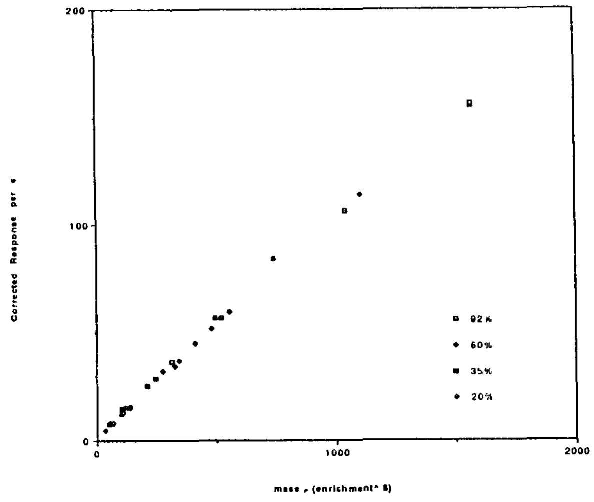

Fig.10: Experimental Counter response as function of $Q = \text{const.}*H*E^{0.8}$ for a fixed sample Radius R.

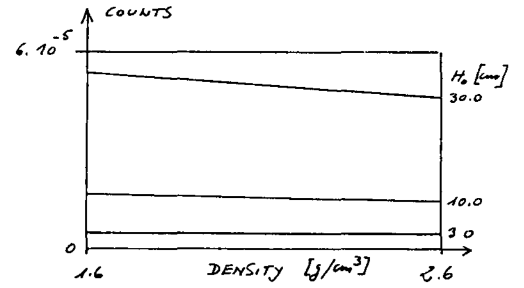

Fig.11: shows the change of the mean number of counts when compressing the sample from a density of 1.6 to 2.6 for various initial heights (at a fixed sample radius R=9.0 [cm]) at E=35%.

Note that for not too large sample heights (H < 20.0 cm) there is almost no change of the number of counts (NO density dependence of the counting rate).

A further interesting aspect of PHONID is the following.

As in general the discriminator threshold in set such, that no count is produced by Be-source neutrons, the actual counts are due only to fission neutrons.

So we may concentrate our considerations on fission neutrons only.

This suggest to separate the physical performance of PHONID into two steps:

Step A: production of (induced-)fission neutrons by the original Be-source neutrons entering the UO_2-sample, and

Step B: evaluation of the effect of these (virgin-) fission neutrons on the He-detectors(production of counts due to these fission neutrons).

The calculations for step B were done by starting fission neutrons homogeneously distributed over the sample material and isotropically and show the very surprising effect that g, the mean number of counts per starting fission neutron is almost constant (independent of R,H and E; $g = 0.0045$ in our case).

The response (of PHONID) separates therefore in

$$C = g*M \qquad (21)$$

where M is the mean number of (virgin) fission neutrons produced in the sample per BE-source neutron as evaluated in Step A (Source multiplication).

References:

1. MCNP - A general Monte Carlo Code for Neutron and Photon Transport. Vers. 3A - 1986, Rep. LA - 7396 - M, Rev-2. Los Alamos National Laboratory.

NEUTRON MULTIPLE CORRELATION ANALYSIS METHOD APPLIED TO THE ASSAY OF RADIOACTIVE WASTE

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Abstract

For the assay of Pu in radioactive waste with low beta and gamma contamination the spontaneous fission rate of the Pu isotopes with even mass numbers are used as fingerprints. For the elimination of neutron multiplication effects and the unknown positioning of the Pu debris in the waste the performance of pair and triple neutron correlation techniques are tested. For this purpose a set of waste drums was measured in the JRC waste barrel monitor together with well characterized Pu samples exposed in various radial and axial positions. The essential formulas required for the interpretation of the measured data are given along with the assay data.

1. Introduction

PuO_2 present in radioactive waste generates neutrons coming from spontaneous fission events, from (α,n) reactions and induced fissions events caused by primary neutrons.

Neutron pair correlation /1/ provides the necessary tools to determine the spontaneous fission rate F_S of the Pu. This is true if the isotopic composition of the Pu in the waste is known. Then the ratio formed by the (α,n) reaction rate S_α and the spontaneous fission neutron emission rate $\nu_{s(1)}F_S$ can be calculated. If neutron multiplication is neglected, then the detection probability ε can be eliminated, which depends on the global self shielding of the PuO_2 debris in the waste matrix. The experimental information required are the effective number of neutron singlets R_1 and the effective number of correlated doublets R_2. Both quantities can be obtained from the shift register /2,5/, the reduced variance meter /6/ and in approximation from the variable dead time counter /7/.

With the triple neutron correlation technique three quantities are available as experimental data the effective number of singlets, doublets and triplets. This permits to determine three unknowns of Pu contaminated waste i.e. either the spontaneous fission rate F_S the (α,n) reaction rate S_α and the detection probability ε of the unknown Pu distribution in the waste barrel or knowing the isotopic composition F_S, ε and the neutron multiplication factor M. Pair and triple correlation measurements can be performed contemporarily with the Euratom Time Correlation Analyzer (TCA) /8/ according to two different methods and analyzed with a special software based on the theory of /9,10,11/. The paper summarizes the necessary expressions for the data acquisition and interpretation. It gives a short description of the waste barrel monitor presents results confirming the used point model for the stochastic process and gives assay data for the most important waste barrels and matrices.

2. Theory

The Euratom Time Correlation Analyser measures the number of signals existing inside fixed time intervals τ. These intervals are opened either randomly or by each neutron signal of the pulse train. The quantities obtained are the frequency distributions $B_x(\tau)$ and $N_x(\tau)$ of the signals inside randomly and signal triggered observation intervals, respectively.

$B_x(\tau) \equiv$ number of events with x signals inside k randomly triggered intervals of duration τ.

$N_x(\tau) \equiv$ number of events with x signals inside intervals of duration τ in which each signal of the pulse train opens such an interval.

The frequencies $b_x^*(\tau)$ and $n_x^*(\tau)$ are obtained normalizing on the number of random interval triggers k and on the number of signals N_T collected during the measurement time T_M, respectively.

$$b_x^*(\tau) = \frac{B_x(\tau)}{k} \tag{1}$$

$$n_x^*(\tau) = \frac{N_x(\tau)}{N_T} \tag{2}$$

$b_x(\tau)$ and $n_x(\tau)$ are the probability counterparts to the measured frequencies $b_x^*(\tau)$ and $n_x^*(\tau)$. For the factorial moments of the probability distributions $b_x(\tau)$ and $n_x(\tau)$ of order μ

$$m_{b(\mu)} = \sum_{x=\mu}^{\infty} \binom{x}{\mu} b_x(\tau) \approx \sum_{x=\mu}^{\infty} \binom{x}{\mu} b_x^{*}(\tau) \qquad (3)$$

$$m_{n(\mu)} = \sum_{x=\mu}^{\infty} \binom{x}{\mu} n_x(\tau) \approx \sum_{x=\mu}^{\infty} \binom{x}{\mu} n_x^{*}(\tau) \qquad (4)$$

rather simple recurrence formulas are obtained /9,10,11/.

$$m_{b(\mu)} = \sum_{r=0}^{\mu-1} \tfrac{\mu-1}{\mu}\, m_{b(r)}\, m^{*}_{b(\mu-r)} \qquad (5)$$

$$m_{n(\mu)} = \sum_{q=0}^{\mu} m^{*}_{n(q)}\, m_{b(\mu-q)} \qquad (6)$$

with $m_{b(0)} = 1$ and $m_{n(0)} = m^{*}_{n(0)} = 1$

The quantities $m^{*}_{b(\mu)}$ and $m^{*}_{n(\mu)}$ are simple analytical expressions of the neutron physical characteristics of the spontaneous fission sample, the moderator detector assembly and the settings of the Time Correlation Analyser (TCA).
For the mathematical description of $m^{*}_{b(\mu)}$ and $m^{*}_{n(\mu)}$ following assumptions are introduced.

1. The test sample has point geometry.

2. There is only fast neutron multiplication in the test sample.

3. Primary neutron emission and the following neutron multiplication chain occur contemporarily.

4. The time response function of the neutron detection is a pure exponential with decay time $1/\lambda$.

5. Primary neutron energy has no influence on the neutron detection probability.

6. There are no dead time losses of signals.

It is then:

$$m^{*}_{b(\mu)} = \frac{\tau}{T_M}\, R_\mu\, w(\mu) \qquad (7)$$

$$m^{*}_{n(\mu)} = \frac{1}{N_T}\, R_{\mu+1}\, f^{\mu} \qquad (8)$$

with $w(\mu) = \sum_{k=0}^{\mu-1} \binom{\mu-1}{k} (-1)^k \frac{1-e^{-\lambda\tau k}}{\lambda\tau k}$

and $f = e^{-\lambda T}(1-e^{-\lambda\tau})$
and with the following expressions for R_1, R_2 and R_3

$$R_1 = \varepsilon F_S T_M M \nu_{s(1)}(1+\alpha) \qquad (9)$$

$$R_2 = \varepsilon^2 F_S T_M M^2 \left[\nu_{s(2)} + (M-1)\frac{\nu_{s(1)}\nu_{I(2)}}{\nu_{I(1)}-1}(1+\alpha) \right] \qquad (10)$$

$$R_3 = \varepsilon^3 F_S T_M M^3 \left\{ \nu_{s(3)} + 2(M-1)\frac{\nu_{s(2)}\nu_{I(2)}}{\nu_{I(1)}-1} + \right. \qquad (11)$$

$$\left. (1+\alpha)(M-1)\frac{\nu_{s(1)}}{\nu_{I(1)}-1}\left[\nu_{I(3)} + 2(M-1)\frac{\nu^2_{I(2)}}{\nu_{I(1)}-1} \right] \right\}$$

The symbols used are:

S_α = (α,n) neutron emission rate of test item.
F_S = Spontaneous fission rate of test item.
p = Probability that a neutron generates an induced fission event.
ε = Probability for detection of a neutron.
τ = Observation interval.
T = Delay between trigger signal and start of observation interval τ.
T_M = Measurement time.
λ = Fundamental mode decay constant of moderator-detector assembly.
$P_{j\nu}(p)$= Probability for the emission of ν fast neutrons per prompt fission caused by a primary neutron generated by reaction j (j=I when induced fission, j=s for spontaneous fission) /12,13/.
$\overline{\nu_{j(\mu)}(p)}$= μth factorial moment of the $P_{j\nu}(p)$ distribution.

$$\overline{\nu_{j(\mu)}(p)} = \sum_{\nu=\mu}^{\infty} \binom{\nu}{\mu} P_{j\nu}(p) \qquad (12)$$

The symbol M given by

$$M = \frac{1-p}{1-p\nu_{I(1)}} \qquad (13)$$

is used for the fast neutron multiplication factor.

In many practical applications the isotopic composition of the fissile material is known. From this information the so-called α-ratio is derived

$$\alpha = \frac{S_\alpha}{\nu_{s(1)} F_S} \qquad (14)$$

The experimentally determined factorial moments of the two signal frequency distributions (Eqs. 5 and 6) serve to obtain the expressions $m^{*}_{b(\mu)}$ and $m^{*}_{n(\mu)}$. These two quantities are proportional to the μ-tuplets of order μ and $\mu+1$ respectively. Having determined R_1, R_2 and R_3 the unknowns of a waste barrel assay can be determined. Following 4 analysis procedures can be applied:

Case 1. Unknown F, ε, M Code routine FEM
Used for fuel fabrication waste with well known isotopic composition of fuel debris but unknown matrix of the waste and debris distribution.

Case 2. Unknown S_α, F_S, M Code routine SAFM
Used for homogeneous waste or small waste

packages with calibrated neutron detection probability ε.

Case 3. Unknown F, ε, S_α Code routine FESA
Used for completely unknown waste.

Case 4. Unknown F,ε Code routine FE
Used in cases when triple correlation fails due to a too small detection probability ($\varepsilon < 0.035$).

3. The Waste Barrel Monitor and Samples

The Waste Barrel Monitor consists of three different elements the detector head, the electronic chains and the time correlation analyser for the data acquisition. The detector head has a cylindrical cavity housing a waste barrel up to 220 litre. The 60 He^3 neutron detectors are arranged in a 4π geometry inside 20 Polyethylene and Cadmium lined modules. A set of 4 He^3 neutron detectors is connected via an amplifier and discriminator directly to the TCA. The TCA is fed with 15 such signal inputs. The TCA processes the signal pulse train contemporarily for the two observation interval opening methods i.e. the randomly opened and the signal triggered intervals. The pulse train is automatically analysed in both methods for 16 observation intervals each of different duration in contrast to the present existing shift registers having only one observation interval. Thus each method gives 16 analysis results one for each interval size. This feature permits a check of the reliability of the obtained results. The adjustable interval range is such that the dead time and decay constant can be measured as well with high precision.

The stored frequency distributions of the two methods each with 16 time intervals are transferred after each measurement to a personal computer for analysis with the theory outlined above.

The statistical model of the neutron signal pulse train simplifies considerably as indicated in the equations listed above if the neutron response function of the detector head is a pure exponential with a decay constant λ. For this reason the detector modules were designed with a high neutron leakage and with an under moderation of the neutron spectrum optimised for the measurement of Pu in the g range. This leads for the same number of detector tubes to a lower neutron detection probability but gives a very good exponential decay of the neutron response function. It was found experimentally that the decay constant becomes this way independent of the waste size, its matrix and the position of the Pu debris in the matrix. Cosmic radiation neutrons and Pu fission neutrons give within the error limits the same values for λ with any type of matrix.

The present set of experiments explore the potential of the multiplet analysis applied to the assay of fuel fabrication waste incorporated in various matrices. The parameters of investigation are:

1. Waste barrel size (20 l, 32 l, 60 l, 220 l).
2. Waste matrices.
3. Source arrangements.
4. Performance of the two statistical methods.

The waste barrels have axial re-entrant tubes for the accommodation of the PuO_2 samples. These tubes are arranged in two perpendicular axes.

The PuO_2 samples have a diameter of about 10 mm. The weight and isotopic composition of the samples are collected in Table I /14/.

4. Assay Results

One of the major problems measuring Pu in a waste barrel is the unknown spatial distribution of Pu in an unknown heterogeneous waste matrix and the size of the individual Pu debris. For this reason it is necessary to consider the average detection probability of the neutrons originating from the Pu debris as an unknown in the used point model. Fig. 1 shows the variation of the neutron detection probability for various waste barrels ranging from 20 litre to 220 litre. The PuO_2 and Cf-252 neutron sources were placed for this purpose in the centre of the concrete waste barrels. The detection probability of the detector head decreases from 0.108 for a 20 litre and 0.0384 for a 220 litre concrete barrel. The detection probability is slightly smaller for a Cf-252 neutron source indicating that the detector modules of the detector head are under moderated. Fig. 2 shows the radial dependence of ε as function of the radius in a waste barrel of 220 litre with a concrete matrix. The lowest value is about 0.0304 and

Sample Number	Comp	Pu-wgt [g]	Pu-238 [% wt]	Pu-239 [% wt]	Pu-240 [% wt]	Pu-241 [% wt]	Pu-242 [% wt]	Am-241 [ppm]
2	PuO_2	1.997	0.072	85.151	13.157	1.353	0.267	4860
20	PuO_2	4.983	0.072	85.151	13.157	1.353	0.267	4830
21	PuO_2	9.967	0.072	85.151	13.157	1.353	0.267	4830
22	PuO_2	19.919	0.072	85.151	13.157	1.353	0.267	4830
30	metal	20.566	0.104	69.286	26.303	2.879	1.428	27510

Table I. Weight and Isotopic Composition of the Pu Samples Used.
* Refers to 30.07.1982

increases with increasing radius to 0.086.

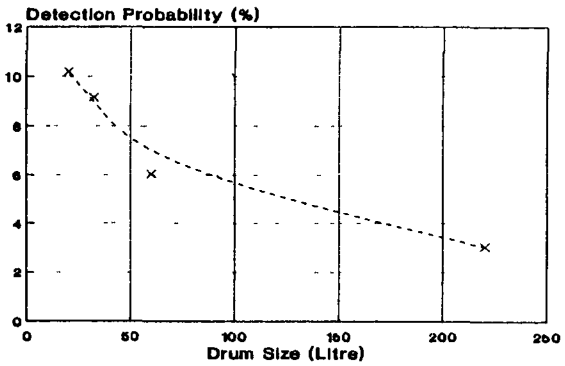

Fig. 1 Detection Probability as Function of Barrel Size. Concrete Matrix; Source in Central Position.

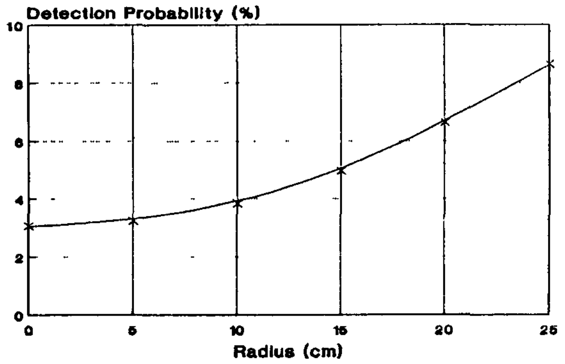

Fig. 2 Detection Probability as Function of Radial Source Position. 220 Litre Concrete Barrel.

The best assay results are obtained with a waste matrix of lowest hydrogen density. Table II summarizes the results of an empty 60 litre waste drum placing the PuO_2 sample at various radii. Best results are achieved with the triple correlation method opening the observation intervals randomly. The signal triggered version leads to slightly higher errors but all assay results are below a 4.5 % systematic error limit.

		Triple Correlation		Pair Correlation	
Radial Pos. [cm]	Trigger Method	$M240_{eq}$ [g]	Error [%]	$M240_{eq}$ [g]	Error [%]
0.0	Random	2.676	-2.5	2.249	-18.0
	Signal	2.638	-3.8	2.245	-18.2
5.0	Random	2.671	-2.7	2.255	-17.8
	Signal	2.626	-4.3	2.244	-18.2
10.0	Random	2.651	-3.4	2.278	-17.0
	Signal	2.642	-3.7	2.256	-17.8
15.0	Random	2.638	-3.9	2.275	-17.1
	Signal	2.627	-4.3	2.249	-18.0

Table II. **Assay Results of 60 litre Waste Barrel with Void Matrix for Various Radial Source Positions.**
Pu-240 Mass equivalent $M240_{eq}$= 2.744 g.

With both pair correlation methods the $M240_{eq}$ value is underestimated by about 17-18 %. A systematic trend of the assay data as function of radius does not exist.

The assay results of a 60 litre waste barrel with a concrete waste matrix shows that the triple and pair correlation lead to about the same systematic errors. Again the best results are achieved with the triple correlation method giving for the PuO_2 sample at the barrel centre a 4.5 % underestimation and at the outer periphery of the barrel a 5.3 % overestimation of the $M240_{eq}$. The triple correlation method with signal triggered observation intervals overestimates the mass with the PuO_2 sample at the centre by 8 % and decreases to 0.6 % with the sample at the periphery. The pair correlation method underestimates systematically the $M240_{eq}$ and gives the greatest errors with the sample at the periphery (Table III).

		Triple Correlation		Pair Correlation	
Radial Pos. [cm]	Trigger Method	$M240_{eq}$ [g]	Error [%]	$M240_{eq}$ [g]	Error [%]
0.0	Random	2.627	-4.3	2.663	-3.0
	Signal	2.963	8.0	2.572	-6.3
5.0	Random	2.766	0.8	2.664	-2.9
	Signal	3.003	9.4	2.554	-6.9
10.0	Random	2.822	2.8	2.557	-6.8
	Signal	2.914	6.2	2.443	-10.9
15.0	Random	2.890	5.3	2.440	-11.1
	Signal	2.768	0.6	2.353	-14.2

Table III. **Assay Results of 60 litre Waste Barrel with Concrete Matrix for Various Radial Source Positions.**
Pu-240 Mass equivalent $M240_{eq}$= 2.744 g.

Waste barrels of 220 litre with a concrete matrix lead with both triple correlation methods to rather large errors for a PuO_2 sample placed at its centre (Table IV). The errors decrease with increasing radial position of the PuO_2 sample and reach at the periphery a value of about 2-5 %. Nearly in all radial sample positions the $M240_{eq}$ is overestimated. Much better assay results are obtained with the pair correlation. Here the smallest systematic error of about 1 % occurs with the PuO_2 sample in the centre and increases with the radius of the sample position to about 10 %. In most cases the pair correlation leads here to an underestimation of the $M240_{eq}$. Assuming a homogeneous distribution of the PuO_2 debris the expected systematic error would be in the order of 5.7 % applying triple correlation and would be 10 % with pair correlation.

		Triple Correlation		Pair Correlation	
Radial Pos. [cm]	Trigger Method	$M240_{eq}$ [g]	Error [%]	$M240_{eq}$ [g]	Error [%]
0.0	Random	3.999	45.7	2.783	1.4
	Signal	3.769	37.4	2.708	-1.3
5.0	Random	3.595	31.0	2.778	1.2
	Signal	3.429	25.0	2.710	-1.2
10.0	Random	3.375	23.0	2.725	-0.6
	Signal	3.170	15.5	2.640	-3.8
15.0	Random	3.250	18.4	2.634	-4.2
	Signal	2.995	9.1	2.544	-7.3
20.0	Random	3.048	11.0	2.533	-7.7
	Signal	2.872	4.7	2.441	-11.0
25.0	Random	2.847	3.7	2.440	-11.0
	Signal	2.678	-2.4	2.354	-14.2

Table IV. Assay Result of 220 litre Waste Barrel with Concrete Matrix for Various Radial Source Positions

Table V gives the assay results using two PuO_2 samples, one sample of $M240_{eq}$ = 1.373 g remained during all experiments in the centre of the 220 litre concrete waste barrel. The other sample of $M240_{eq}$ = 2.744 g was moved after each experiment to a different radial position. With both samples at the centre the triple correlation method with signal triggered observation intervals τ gives an error of about 6 %. With the second sample in the outermost position the error increased to 19.7 %. The other triple correlation method gives worse results for both samples in the centre position and a smaller error with the second sample in the outer most position. With pair correlation the randomly opened observation interval method leads to a 16 % error with both samples in the centre and to 12 % with the large sample at the outer most position. A similar range of errors is observed for the signal triggered observation interval method only with 0.7 % error for both samples at the barrel centre.

		Triple Correlation		Pair Correlation	
Radial Pos. [cm]	Trigger Method	$M240_{eq}$ [g]	Error [%]	$M240_{eq}$ [g]	Error [%]
0.0	Random	5.937	44.2	4.789	16.3
	Signal	4.396	6.8	4.086	-0.7
5.0	Random	4.782	16.1	4.745	15.3
	Signal	3.715	-9.7	4.049	-1.6
10.0	Random	5.318	29.0	4.564	10.8
	Signal	4.211	2.3	3.903	-5.2
15.0	Random	4.857	18.0	4.284	4.1
	Signal	3.804	-7.6	3.695	-10.2
20.0	Random	4.503	9.4	3.915	-4.9
	Signal	3.551	-13.7	3.383	-17.8
25.0	Random	4.072	1.1	3.589	-12.8
	Signal	3.306	-19.7	3.124	-24.1

Table V. Assay Results of 220 litre Waste Barrel with Two PuO_2 Sources in Concrete Matrix.
Source 1: Fixed in centre; $M240_{eq}$= 1.373g
Source 2: Various Positions; $M240_{eq}$= 2.744g

Table VI summarizes results measuring PuO_2 samples of different $M240_{eq}$ in the centre of the 220 litre waste barrel. Triple correlation can only be applied with the present detector modules to samples with a $M240_{eq}$ larger than 2 g. This is mainly due to the low detection probability at the central position of the 220 litre concrete waste barrel and the cosmic neutron radiation background which is not a constant in time. Pair correlation gives still R_2 values which are well above the variation of the R_2 component of the cosmic radiation background.

		Triple Correlation		Pair Correlation	
$M240_{eq}$ [mg]	Trigger Method	$M240_{eq}$ [g]	Error [%]	$M240_{eq}$ [g]	Error [%]
275.1	Random	-		330.9	20.0
	Signal	-		295.6	7.5
686.5	Random	-		802.0	16.8
	Signal	749.0	-9.1	715.3	4.0
1373	Random	-		1402	2.1
	Signal	-		1379	0.4
2744	Random	3.999	45.7	2788	1.4
	Signal	3.769	37.3	2708	-1.3

Table VI. Assay Results of Various Sample Sizes with 220 litre concrete Waste Barrel.
Samples in Centre Position.

5. Conclusions

1. The measurement of low hydrogen density waste can easily be accomplished with triple neutron correlation.
2. Concrete waste barrels of 60 litre can be monitored with systematic errors in the limits of 5 to 10 %. Both pair and triple correlation can be applied.
3. Concrete waste barrels of 220 litres lead to average systematic errors of 6.7 % and 10 % for triple and pair correlation respectively. Both with rather large errors for Pu debris located close to the barrel centre.

4. The application limits for triple correlation in a 220 litre waste barrel is about 2 g of $M240_{eq}$ for the central position. For pair correlation it is at least ten times smaller. Shielding against cosmic radiation and a more refined treatment of the pulse train along with detector head modules designed specifically for low Pu mass detection reduces these limits considerably.

5. The detection of $M240_{eq}$ limit for a 'signal to background ratio' of one of a 220 litre concrete waste barrel using only single counts is 35 mg ± 17 mg. In case the cosmic radiation background is measured before and after any barrel measurement for one hour the absolute limit for the detection is $M240_{eq} = 1.8$ mg.

Acknowledgements

The authors acknowledge fruitful discussions with L. Bondar and R. Dierckx (JRC Ispra). Special thanks are due to prof. W. Ameling, K. Kleinekorte and A. Karavas (Rogovski Institut, RWTH Aachen) for building the Time Correlation Analyser and to J.F. Gueugnon and K. Richter (TUI, Karlsruhe) for providing the samples.

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ACTIVE NEUTRON COUNTING SOFTWARE FOR NUCLEAR MATERIAL VERIFICATION WITH THE NEUTRON COLLAR AND THE ACTIVE WELL COINCIDENCE COUNTER

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Abstract

The Uranium Neutron Coincidence Collar (UNCL) is used by the International Atomic Energy Agency (IAEA) to verify the Uranium-235 and Uranium-238 content in light-water reactor (LWR) fuel assemblies. The IAEA uses also the Active Neutron Coincidence Counter (AWCC) for the verification of the Uranium-235 content in uranium samples and MTR fuel assemblies. This paper describes the Active Neutron Counting System (ANCS). The application software is considered to be user-friendly and can lead inspectors through the measurement steps and provide final evaluation in the field and at the IAEA. The software is based on the functional specification prepared by the IAEA and reviewed by the Los Alamos National Laboratory (LANL), USA. The algorithms were provided by the IAEA in consultation with LANL.

1. Introduction

Earlier existing software for Active Neutron Non-Destructive Assay (NDA) with AWCC/UNCL was designed for the first generation of personal computers which had limited calculating power, smaller memory, no mass storage (hard disk), and limited software capability. Recently, advanced portable computers have become available which are suitable for field use by the inspectors. In addition the procedures of performing these kinds of measurements and calibration have been established /1,2,3/.

The IAEA's safeguards inspectors are required to efficiently verify Uranium content of nuclear material in various member states within strict time limits in an uniform and reliable way. All this has necessitated the development of ANCS.

The ANCS has been developed to assist IAEA safeguards inspectors to perform Active Neutron NDA with the AWCC/UNCL in the field. The system is used to collect, maintain, analyse and evaluate inspection data at facilities and the IAEA.

2. Material and Methods

Software Engineering

The development of ANCS is based on the experience gained with the High-level Neutron Coincidence Counting System for Plutonium assay, which was developed by Pham T.P., M.S. Krick and K. Javorzky in 1987. ANCS conforms to the IAEA's software standards. The software uses a modular library developed by the IAEA.

A structured method of software engineering is used through the whole software development cycle. An overview of ANCS within the inspection environment is summarized with the context diagram shown in Fig. 1.

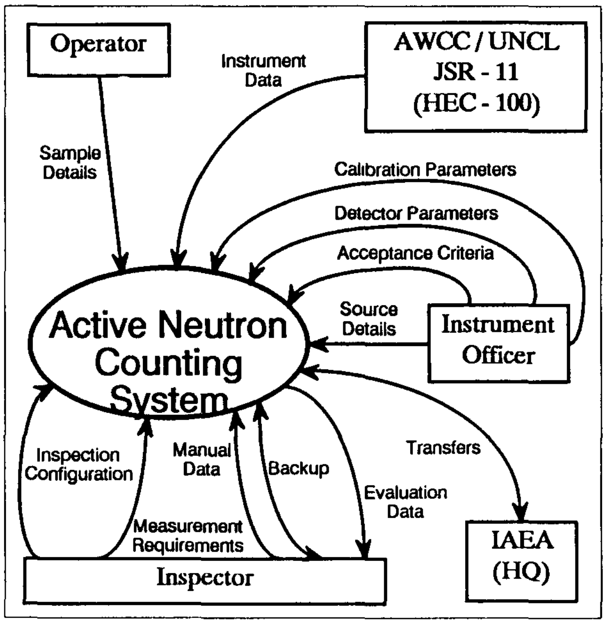

Fig. 1 - Context Diagram of ANCS

The system accepts Intrument Data from either the AWCC or UNCL through a shift-register electronics such as HEC-100 or JSR-11. These sets of measurement data are associated with **Samples Details** provided to the IAEA's inspector by the operator of the facility where the inspection is occuring. Sample Details for those samples to be measured with AWCC consist of:

1 - Sample Identification;
2 - Stratum Identification;
3 - Sample Length in centimeters;
4 - Uranium-235 content in grams.

In the case of the UNCL, the following additional information is required:

5 - Uranium-238 weight in grams;
6 - Total number of fuel rods;
7 - Number of burnable poison rods;
8 - Type of burnable poison;
9 - Weight percentage of burnable poison.

The software has to be setup before the inspection by the NDA specialist or the **Instrument Officer** at the IAEA. This involves specification of **Detectors Parameters, Source Details, Calibration Parameters, and Acceptance Criteria.**

The **Inspector** is the main user of the ANCS. The inspector specifies information specific to the actual inspection environment - **Inspection Configuration,** and **Measurement requirements.** The inspector performs **Evaluation of Data.** The inspector has the responsibility to make a **Backup** disk of the system's data and to extract measurement data - a **Transfer** disk to take back to the IAEA's headquarters. The system has the capability to accept instrument data through keyboard - **Manual Data** in case of communication failure.

ANCS is menu-driven. Its menu hierarchy is partly shown on Fig.2. The menu option "System Maintenance" is used mainly by active-NDA's specialists at the IAEA to setup the system as mentioned above. This option is not needed in the field during normal inspections. Normally, the option "Measurements" is used extensively by inspectors in the field, which allows them to make background, normalization and verification measurements.

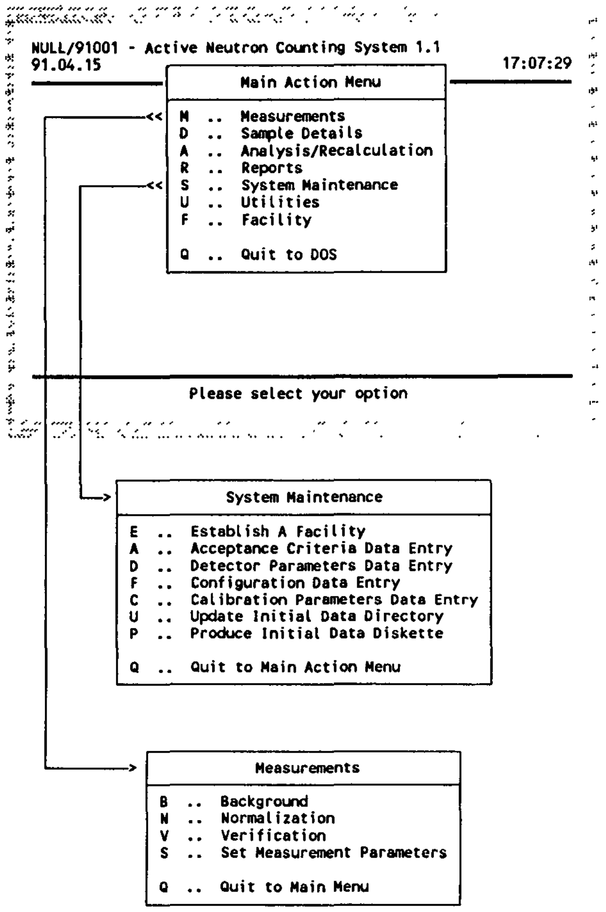

Fig. 2 - Menu Hierarchy of ANCS

Software's Algorithm

The IAEA's procedures for NDA measurements with the AWCC/UNCL using the latest calibration and evaluation methods as described in /2,3/ are fully implemented in ANCS.

Determination of Uranium-235 is based on active measurements. AWCC: The coincidence rate obtained with the AWCC in active mode is corrected for background and normalized with a constant

$$K=K_0K_1, \qquad (1)$$

where K_0 corrects for relative AmLi source strengths, K_1 corrects for electronic drift and geometric variations. UNCL: The normalized active-minus-passive coincidence rate in No-Cd mode is the basis for UNCL measurement of ^{235}U. ANCS takes into account 6 components of the normalization constant for UNCL assay:

$$K=K_0K_1K_2K_3K_4K_5, \qquad (2)$$

where K_0 and K_1 are identical to those of (1), K_2 corrects for relative detector efficiencies and source yield, K_3 - for presence of poisons, K_4 - for Uranium mass loading, K_5 - for other sample variations.

Calculation of Uranium mass can be done with one from 4 types of calibration curves:

1 - Cubic polynomial:

$$R=a+bm+cm^2+dm^3; \qquad (3)$$

2 - Power:

$$R=am^b; \qquad (4)$$

3 - Exponential:

$$R=a(1-e^{bm}); \qquad (5)$$

4 - Saturating:

$$R=\frac{am}{1+bm}, \qquad (6)$$

where R is the corrected coincidence net rate, m - ^{235}U weight, and a,b,c,d - curve's constants. Even though the system is designed to measure ^{235}U with UNCL in the aective thermal mode without Cadmium liners, fast active measurements of ^{235}U with Cadmium liners installed inthe UNCL can be made with ANCS.

Uranium-238 determination is based on passive measurements with UNCL. ANCS corrects the passive coincidence rate measured with UNCL for background, room-background-induced fissions, and ^{235}U fissions according to the equation:

$$R_{Corr}=[rR_{meas}-0.0014(\sqrt{r}T-a)]\frac{b+cE_{Cal}}{b+cE_{Meas}}, \quad (7)$$

where R_{Corr} is corrected net coincidence rate, R_{Meas} - measured coincidence rate, T - measured totals rate, E_{Cal} - enrichment of reference fuel assembly, E_{Meas} - enrichment of measured fuel assembly, and r,a,b,c - constants. Calculation of ^{238}U is performed with the power function type (4) of calibration curve.

Confirmation of Declared Poison. Since the ratio of net (active-passive) coincidence or total rates with and without Cadmium liners is proportional to the declared burnable-poison-correction factor K_3, ANCS uses four sets of measurements: passive and active without Cadmium liners and passice and active with Cadmium liners, to confirm the declared burnable poison in the fuel assembly. The empirical value of burnable-poinson-correction factor K_3 participating in equation (2) above is determined according to the following equation:

$$K_{3R}=a\frac{R_{Cd}}{R_{NoCd}}+b, \quad (8)$$

where R_{Cd} and R_{NoCd} are the net (active-passive) coincidence rates with and without Cadmium liners, and a,b - constants. Similiarly, the capability of deriving K_3 from the totals rates ratio is also provided by the software:

$$K_{3T}=c\sqrt{\frac{T_{Cd}}{T_{NoCd}}}+d, \quad (9)$$

where T_{Cd} and T_{NoCd} are the net (active-passive) total rates with and without Cadmium liners, and c,d - constants.

5. Results

Accepting measurement data from the AWCC, ANCS verifies the ^{235}U content of samples based on active measurements. In the UNCL mode the safeguards inspector measures the ^{235}U and ^{238}U content of LWR fuel assemblies and the burnable-poison-correction factor. The system reports immediately any significant difference between measured and declared values. The relative standard deviation for a group of samples identified with the same stratum identification and measured within a specified range of dates can be calculated by the software. All measured and evaluated data are available for transmitting from the field to the headquarters of the IAEA for archiving, analysing, and evaluating purposes.

With the capability to accept measurement data through keyboard entry, to review them and to re-calculate results, ANCS can also serve as an analytical tool to re-evaluate measurements and check measurement consistency.

6. Conclusion

Implementation of the cross-calibration concepts for the whole family of active NDA detectors (AWCC/ UNCL), and implementation of multiple correction terms, especially for the UNCL, required complicated designing and programming, but the result significantly simplifies the calibration and measurement procedures in the field. Evaluation feedback is achieved not only at the IAEA but on site to guarantee the reliability of the measurements. Measurements can be made in an automated, uniform and reliable way. Therefore, the overall and on-site time of an inspection is to be reduced.

ANCS has been fully developed since October, 1990, and is undergoing extensive testing. The results of field tests in Canada, Sweden and USA are satisfactory. ANCS is now available and approved for inspection use by the IAEA's inspectors.

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A PROGRAMME FOR EURATOM SAFEGUARDS INSPECTORS, USED IN THE ASSAY OF PLUTONIUM BEARING MATERIALS BY PASSIVE NEUTRON INTERROGATION.

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Abstract

The programme P.E.C.C. (Passive Euratom Coincidence Counters), /1/ has been developed at the Joint Research Center, Ispra by the Euratom Safeguards Directorate, Luxembourg and the Safety Technology Institute, Ispra for the acquisition, evaluation, management and storage of measurement data originating from passive neutron assay of plutonium bearing materials. The software accommodates the implementation of the NDA (Non Destructive Assay) procedures for all types of passive neutron coincidence deployed by the Euratom Safeguards Directorate, Luxembourg.

Overview of the system

The Programme was developed to satisfy Euratom's requirements for a software package capable of evaluating, managing and storing measurement data acquired by the High Level Neutron Coincidence Counter (HLNCC) family of detectors, /2,3,5/. The programme complies with Euratom's philosophy of standardisation for Non Destructive Analysis (NDA) techniques and optimisation of the calibration activity for a series of nominal identical detector systems.

PECC (Fig. 1) belongs to a family of NDA instruments, /4/ that has been designed according to a general framework for a Safeguards integrated data evaluation system. PECC is an example of the state of the art in the NDA workstation field. This generation of NDA instruments was designed and developed by the JRC Ispra in synergy with the EC Safeguards Directorate to cover the functions an inspector performs during the field activities. In the frame of this cooperation, the PECC workstation was constructed with the following design guidelines:

- **Inspection summary**
- **Inspection procedure support**
- **Data post-processing**
- **Intelligent assistance**
- **Minimize user input**

An Inspection (Fig. 2), from the user point of view, is the set of the declared and measured data collected during an inspection campaign. The system internally handles the files concerning with the background, the performance check and the sample measurements, and correlates them in order to achieve the final results. The internal software structure is hidden from the user, thus avoiding a need to remember names and parameters during the phases of data post processing or down loading to a headquarters station. The macro-operations the user is concerned with are related to the **inspection**; the system will expand them into more primitive functions that get advantage from the powerful software architecture. **All data that make up an inspection can be handled as one item. For instance to back-up or to restore an inspection from a diskette takes a single command, /6/.**

Inspection procedure support means that the user (i.e. the inspector) is guided along the activities being performed in an inspection: set up of the detector chain being used; selection of the proper normalization source and material type; performance of background and performance check measurements; and finally, the description and measurement of samples.

Data post-processing **(Head quarter analysis)** provides the tools to retrieve data from an old inspection and to process them. The user may verify the results or test the effect produced by changing an input parameter. The system allows the user to change any input parameter without affecting the primary data files.

Intelligent assistance means that the instrument provides powerful on line help that contains all the information the user may need during his inspection activity.

Minimize user input means that a great amount of input data can be provided to the measurement system with a minimum number of selections rather then be repeatedly entered via keyboard. The characterization parameters of the specific shift register device, detector head and normalization source used during an inspection are constant, and can be identified with names.

PECC provides a library containing the description of these objects, plus the material types and their associated calibration curves, /1/. Data selection is guided by the system that shows only objects compatible with the previously selected ones. In this way input data acquisition is simple, fast and safe, data are always consistent and clerical errors are avoided. Libraries are made up with standard ASCII files and therefore can be handled off line, i.e. adding an object to the library requires no changes to the program.

The programme is structured in such a way as to provide the user with a define **library of instrument types** such as the HLNCC, ISCC, UFBC and PNCC, /5/. **Calibration data** is available for various categories of plutonium bearing materials such as cans of PuO_2 powders or mixed oxide (MOX) fuel elements.

The operating characteristics for all detectors are stored as ASCII files in data libraries together with appropriate factors used to describe the relationship between detectors of the same instrument type. The programme can thus manage and evaluate measurement data acquired by registered Euratom detector using a **universal calibration** established for a given material category and instrument type.

The **operating performance** of any detector can be monitored using one from a library of registered Cf_{252} sources or a plutonium reference sample. The measurement data acquired during subsequent inspection activities can always be normalised if the operating performance or conditions of the detector vary significantly from the time of calibration. The programme incorporates several test procedures to assure that the instrument is functioning correctly and that no erroneous data is registered for subsequent evaluation. The criteria for the tests can be set by the superuser.

The programme manages and updates for decay operator declarations of plutonium mass and isotopic composition (Fig. 3). The isotopic composition can either be introduced via the keyboard or retrieved from magnetic storage devices using an appropriate fuel batch designation. The measurement data is evaluated before and after the use of the classical neutron self multiplication correction algorithms.

The programme provides a **printed report**, (Fig. 4) including the raw measurement results, the operators declaration, and the data evaluation. It can also store the information in data files for archive purposes.

To assure proper maintenance and consistency in its application two level of access to the programme are permitted. Apart from routine access and execution by the user (Inspector), the Supervisor (Head quarters) can also modify parameter and data files to update the contents of the libraries of calibrations or detectors and perform reevaluation of archive data if required.

PECC provides a powerful **on-line help** which the inspector can access from any point of the program. Help automatically shows the operation the inspector was performing. An on demand help index is shown, and the inspector can navigate on-line help himself. The on-line help function may be used either as a user procedure or as a guide to recovering from errors. The software has a series of built in **error trapping procedures** ensuring consistency in it's execution.

PECC provides the inspector with some **utilities** that simplify operations correlated with inspection activity. The most important are:

- View File. Allows the user to select and display any file belonging either to the library or to an inspection, the displayed file can be sent to the printer.
- Make Batch File / Delete Batch File. These two utilities allow a user to introduce (or delete) the isotopic composition of a new batch. During sample analysis the inspector needs only to select the name of the batch the sample belongs to.
- Copy Inspection. These utilities allow a user to copy from Hard Disk to Floppy Disk (and vice versa) all files related to an inspection. After a copy to Floppy Disk the inspection can be erased from Hard Disk to free disk space.

Hardware environment

The system is built for different types of elaboration and acquisition hardware (Jomar-11, SCRD) either connected through RS232 or AT/PS2 bus link. In fact the hardware drivers are loaded during system set up according to the user requirements. This approach allows a user to add new drivers and therefore to handle new data acquisition instruments with very few software modifications.

PECC and its family of NDA instrument has been designed to use the PC hardware technology. This PC hardware technology offers a modular and highly standardized platform of interfaces and peripherals, which can be easily tailored to user requirements. On the other hand, the PC high computing power at low cost and space requirements makes the past generation of dedicated systems completely obsolete.

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The software is written in the programming language "C" by Microsoft version 5.1. The Windows Development Tools version 2 has been implemented to provide an enhanced user-friendly interface, that is becoming world-wide a standard (Fig. 5,6). The porting to Windows 3 is scheduled for completion the end of this year. The programme runs under the operating system MS-DOS version 3.3 with the microprocessor INTEL 80286 or 80386 with math coprocessor. The user dialogue is controlled either via the keyboard or by a mouse pointing device or by trackball. The dialogue is menu driven allowing the user to easily select any required feature of the programme.

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An inspector using PECC during his inspection campaign has to deal with the following operations:

I) open and define the new inspection. The user is asked to insert the inspection parameters; the inspection name, the location and the inspector name. This new inspection is the working inspection until another one will be opened.

II) set up the environment. The user is asked to select the shift register device, the detector head and the normalization source he is going to use. Environment set up can change during the inspection. If it happens the user is required to perform a new background and a new efficiency check measurement. The system allows the user to handle different types of acquisition hardware either connected through RS232 or AT/PS2 bus link.

III) perform background measurements. The user is asked to introduce the counting time and the number of measurements to be performed. Computed background value is valid until a new background measurement is performed.

IV) perform performance measurements. The user supplies the counting time and the number of repetitions of the sample measurement. Data coming from the shift register are checked for consistency, outlier data are rejected. The computed efficiency correction factor value is used until a new performance check measurement is performed.

V) perform cycles of sample verification measurements. For each sample measurement the user is asked to introduce a few parameters describing the sample, to select its isotopic composition and to enter the counting time and the number of repetitions of the measurement. As for the performance check, measurement outlier and inconsistent data are automatically rejected and substituted with new repetitions. The verification results are displayed and stored on the hard disk, and an exhaustive report on the verification measurement is printed out.

Conclusions

We predict that further evolution in NDA instrumentation technology will take place especially in the directions of multitasking, networking and intelligent assistance. The planned porting to Windows 3 will give us the appropriate environment to meet all these requirements.

PECC and its family of NDA instrument has been designed :

- to use the PC hardware technology, because PC hardware technology offers a modular and highly standardized platform of interfaces and peripherals, which can be easily tailored to our requirements. On the other hand, the PC high computing power at low cost and space requirements makes the past generation of dedicated systems completely obsolete.

- to support a multi-instrument type, multi-detector and multi-material category environment.

- to comply with Euratom's philosophy of standardisation of NDA techniques and optimisation of the calibration activity for a series of nominally identical detector systems.

- to be open to use a new library for nuclear data, detector heads, analog electronic chains, shift registers, reference sources (Cf_{252}, Pu), nuclear material type, and statistical parameters.

- to be ported to Windows 3 and OS/2 to allow multi-tasking use. Multi-tasking is the possibility to create multiple virtual machines executing concurrently several jobs, for instance together with MGA for High Resolution Gamma Spectroscopy or Calorimetry or other applications that research will suggest. This is still possible in the Windows 3 environment if the appropriate hardware is used. An interesting improvement introduced by multitasking in the NDA field is the possibility to run different NDA programs at the same time. We plan a Gamma/Neutrons NDA workstation where PECC can use the isotopic composition files produced by PuMeter. PuMeter is an instrument for the plutonium isotopic composition evaluation based on gamma spectrometry. PECC can evaluate the Pu content of a sample, previously measured by PuMeter, while the PuMeter itself is measuring another one.

In the end it should be pointed-out that porting of the PECC software package to the much more powerful OS/2 operating system will involve very little software effort.

- to store measurement files on hard disk, which constitutes, a complete set of data useful for further analysis with a <u>Relational Database Management System</u>.

- to connect the NDA instrument to its headquarters in a <u>network</u> and to down load inspection data in a [illegible] and easy way. Headquarters will be used for data evaluation and statistical analysis of many inspections.

<u>References</u>

1 - V. Vocino, N. Farese, T. Maucq (JRC-Ispra), G.P.D. Verrecchia, B.G.R. Smith (CEC-luxembourg)
"PECC-Passive Euratom Coincidence Counter. Software Requirements"
T.N.I.90.48 June 1990

2 - W. Hage, D.M. Cifarelli
"On the Factorial Moments on the Neutron Multipicity Distribution of Fission Cascades"
Nuclear Instruments and Methods
A236 (1985) 165-177 July 1984

3 - M.S. Krick (Los Alamos National Laboratory)
"Algorithms and software for data acquisition and analysis using thermal neutron coincidence counters within the IAEA's IFSS/NCC system"
LA-UR-88-1301 April 1988

4 - R. Benoit, W. Hage, A. Prosdocimi, V. Vocino (JRC-Ispra), R. Haas (CEC- Luxembourg), B.L. Thaurel (CEA-Saclay)
"A computer controlled Shift Register system for the automatic assay of plutonium oxide by passive neutron interrogation"
ESARDA-EUR 11041 EN May 1987

5 - H.O. Menlove, J.E. Stewart (LANL)
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LA-11229-MS April 1988

6 - G. Rossi, M. Nebuloni, G. Milani (SYREA - Milan)
"PECC : Data and file system structure"
Private Communication

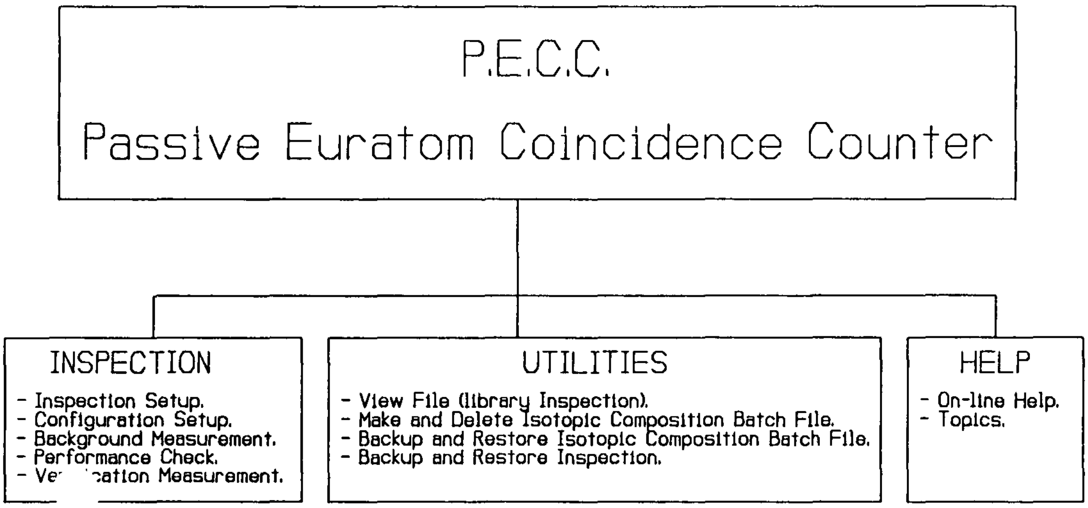


Figure 1 - Flow scheme of the programme

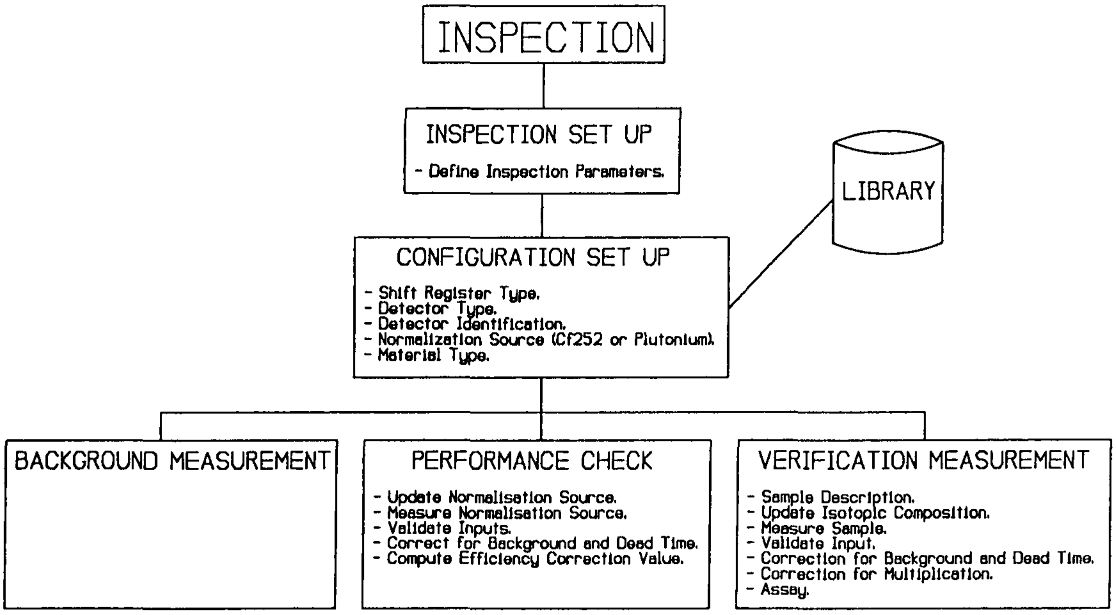


Figure 2 - Flow scheme of the "Inspection procedure"

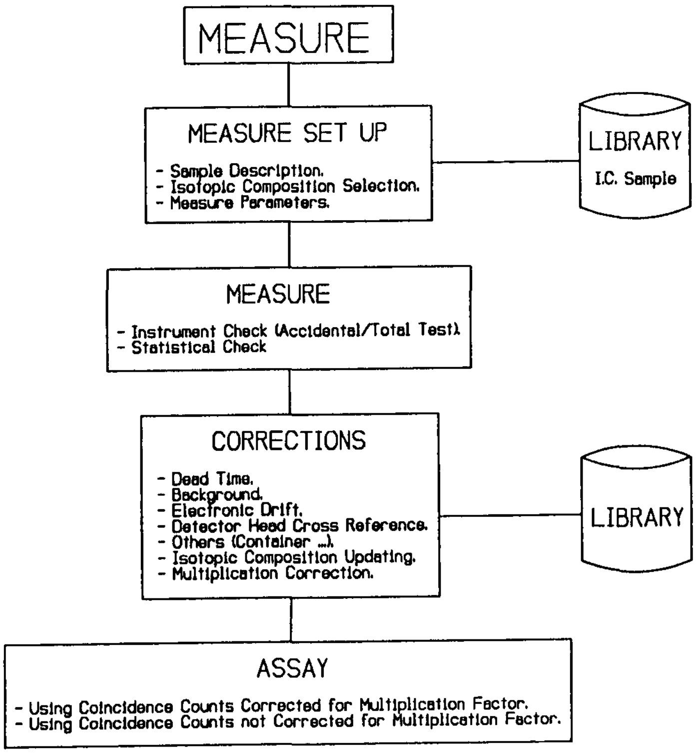
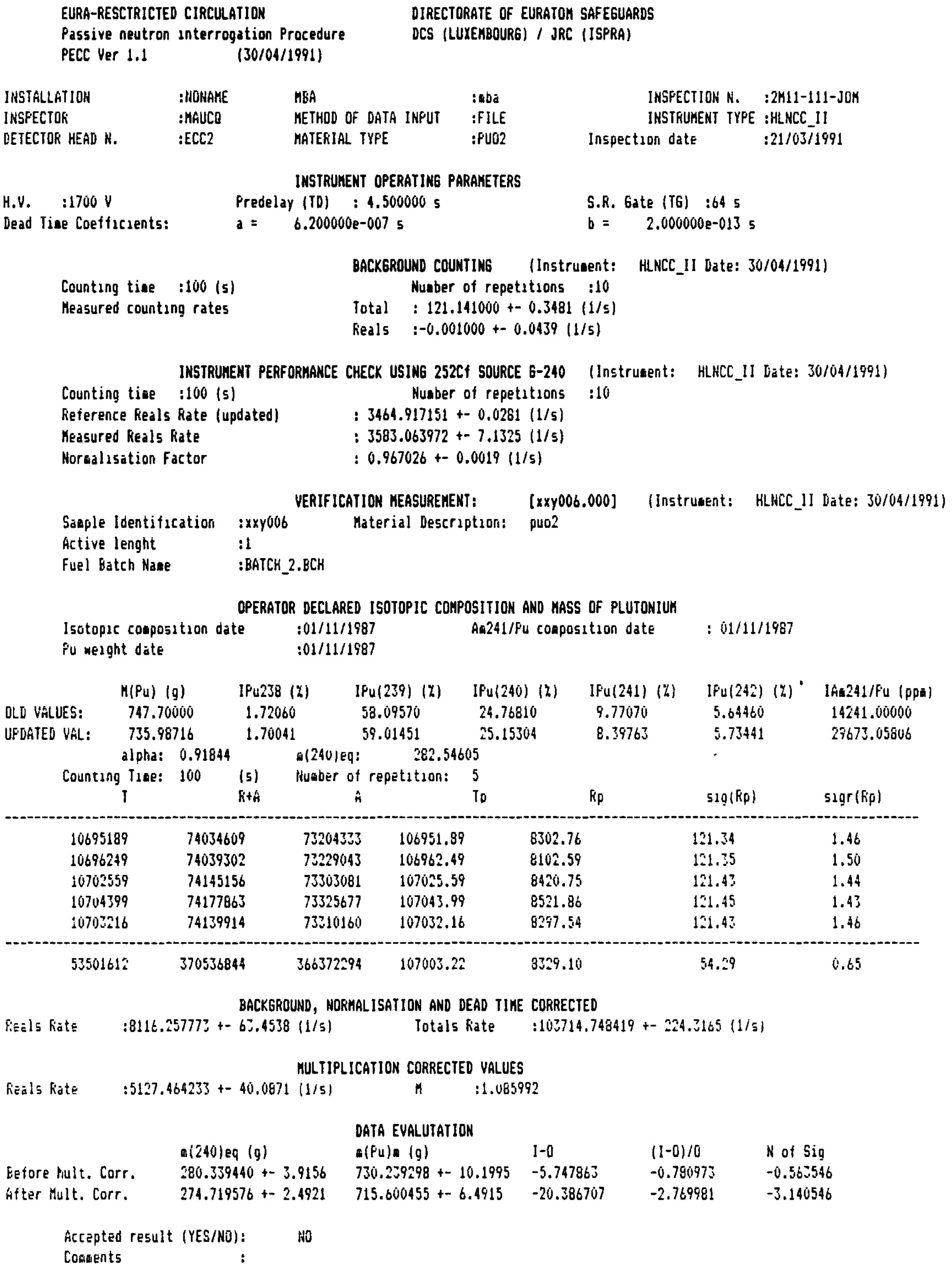


Figure 3 - Flow scheme of the "Verification procedure"



```
          EURA-RESCTRICTED CIRCULATION               DIRECTORATE OF EURATOM SAFEGUARDS
          Passive neutron interrogation Procedure    DCS (LUXEMBOURG) / JRC (ISPRA)
          PECC Ver 1.1           (30/04/1991)

INSTALLATION          :NONAME       MBA                    :mba                INSPECTION N.    :2M11-111-JOM
INSPECTOR             :MAUCQ        METHOD OF DATA INPUT   :FILE               INSTRUMENT TYPE :HLNCC_II
DETECTOR HEAD N.      :ECC2         MATERIAL TYPE          :PUO2           Inspection date      :21/03/1991

                                    INSTRUMENT OPERATING PARAMETERS
H.V.    :1700 V             Predelay (TD)   : 4.500000 s                   S.R. Gate (TG)  :64 s
Dead Time Coefficients:     a =     6.200000e-007 s                        b =     2.000000e-013 s

                                         BACKGROUND COUNTING     (Instrument:   HLNCC_II Date: 30/04/1991)
          Counting time   :100 (s)                 Number of repetitions   :10
          Measured counting rates                Total   : 121.141000 +- 0.3481 (1/s)
                                                 Reals   :-0.001000 +- 0.0439 (1/s)

                       INSTRUMENT PERFORMANCE CHECK USING 252Cf SOURCE G-240   (Instrument:   HLNCC_II Date: 30/04/1991)
          Counting time   :100 (s)                 Number of repetitions   :10
          Reference Reals Rate (updated)         : 3464.917151 +- 0.0281 (1/s)
          Measured Reals Rate                    : 3583.063972 +- 7.1325 (1/s)
          Normalisation Factor                   : 0.967026 +- 0.0019 (1/s)

                                    VERIFICATION MEASUREMENT:       [xxy006.000]    (Instrument:   HLNCC_II Date: 30/04/1991)
          Sample Identification   :xxy006         Material Description:   puo2
          Active lenght           :1
          Fuel Batch Name         :BATCH_2.BCH

                              OPERATOR DECLARED ISOTOPIC COMPOSITION AND MASS OF PLUTONIUM
          Isotopic composition date        :01/11/1987            Am241/Pu composition date         : 01/11/1987
          Pu weight date                   :01/11/1987

                M(Pu) (g)        IPu238 (%)       IPu(239) (%)    IPu(240) (%)     IPu(241) (%)     IPu(242) (%) '  IAm241/Pu (ppm)
OLD VALUES:      747.70000        1.72060          58.09570        24.76810         9.77070          5.64460         14241.00000
UPDATED VAL:     735.98716        1.70041          59.01451        25.15304         8.39763          5.73441         29673.05806
               alpha:  0.91844         m(240)eq:         282.54605                                    -
          Counting Time:  100     (s)     Number of repetition:   5
                  T                 R+A               A                  Tp               Rp               sig(Rp)         sigr(Rp)
-------------------------------------------------------------------------------------------------------------------------------
             10695189         74034609         73204333     106951.89           8302.76                 121.34            1.46
             10696249         74039302         73229043     106962.49           8102.59                 121.35            1.50
             10702559         74145156         73303081     107025.59           8420.75                 121.43            1.44
             10704399         74177863         73325677     107043.99           8521.86                 121.45            1.43
             10703216         74139914         73310160     107032.16           8297.54                 121.43            1.46
-------------------------------------------------------------------------------------------------------------------------------
             53501612        370536844        366372294     107003.22           8329.10                  54.29            0.65

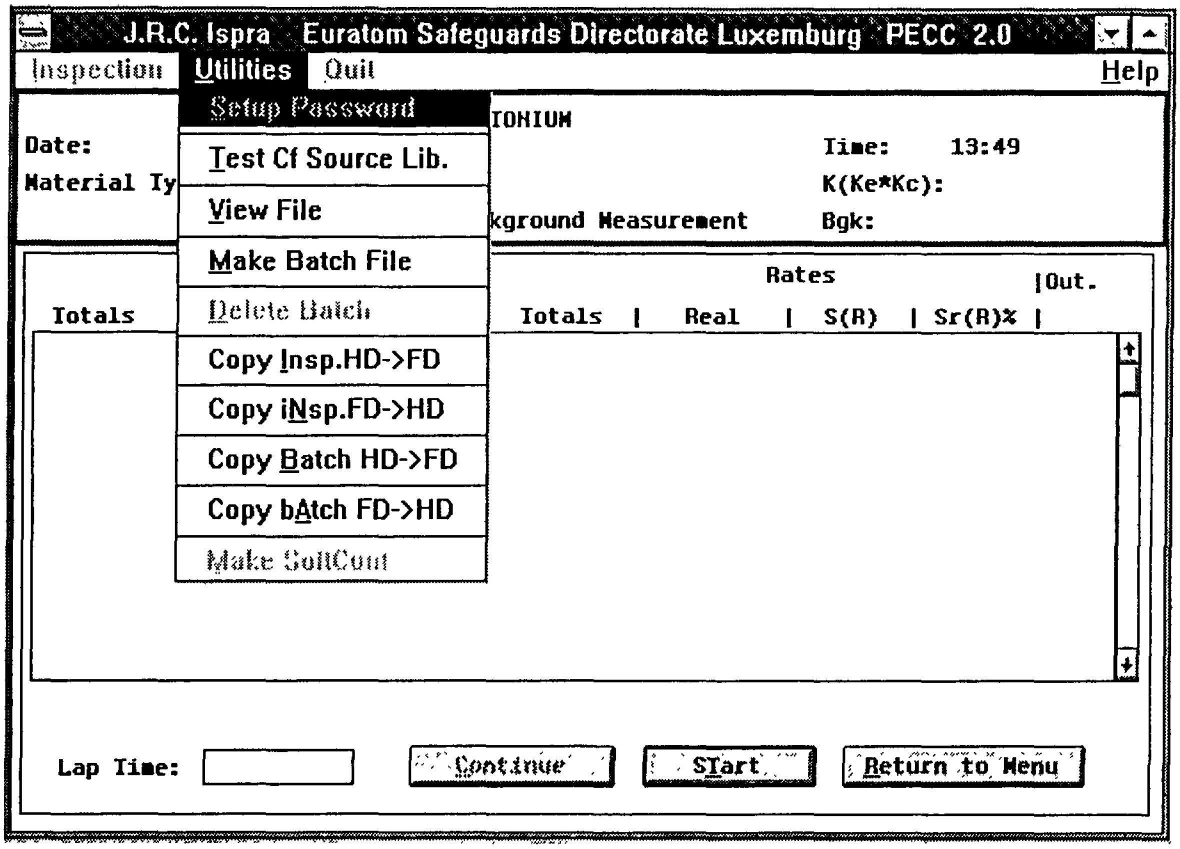
                                 BACKGROUND, NORMALISATION AND DEAD TIME CORRECTED
Reals Rate      :8116.257773 +- 63.4538 (1/s)          Totals Rate      :103714.748419 +- 224.3165 (1/s)

                                      MULTIPLICATION CORRECTED VALUES
Reals Rate      :5127.464233 +- 40.0871 (1/s)            M       :1.085992

                                              DATA EVALUTATION
                        m(240)eq (g)            m(Pu)m (g)            I-O               (I-O)/O          N of Sig
Before Mult. Corr.      280.339440 +- 3.9156    730.239298 +- 10.1995  -5.747863        -0.780973        -0.563546
After Mult. Corr.       274.719576 +- 2.4921    715.600455 +- 6.4915   -20.386707       -2.769981        -3.140546

          Accepted result (YES/NO):       NO
          Comments                :
```

Figure 4 - Printed report of the Verification



IONIUM

Date:

Material Ty

Time: 13:49

K(Ke*Kc):

kground Measurement

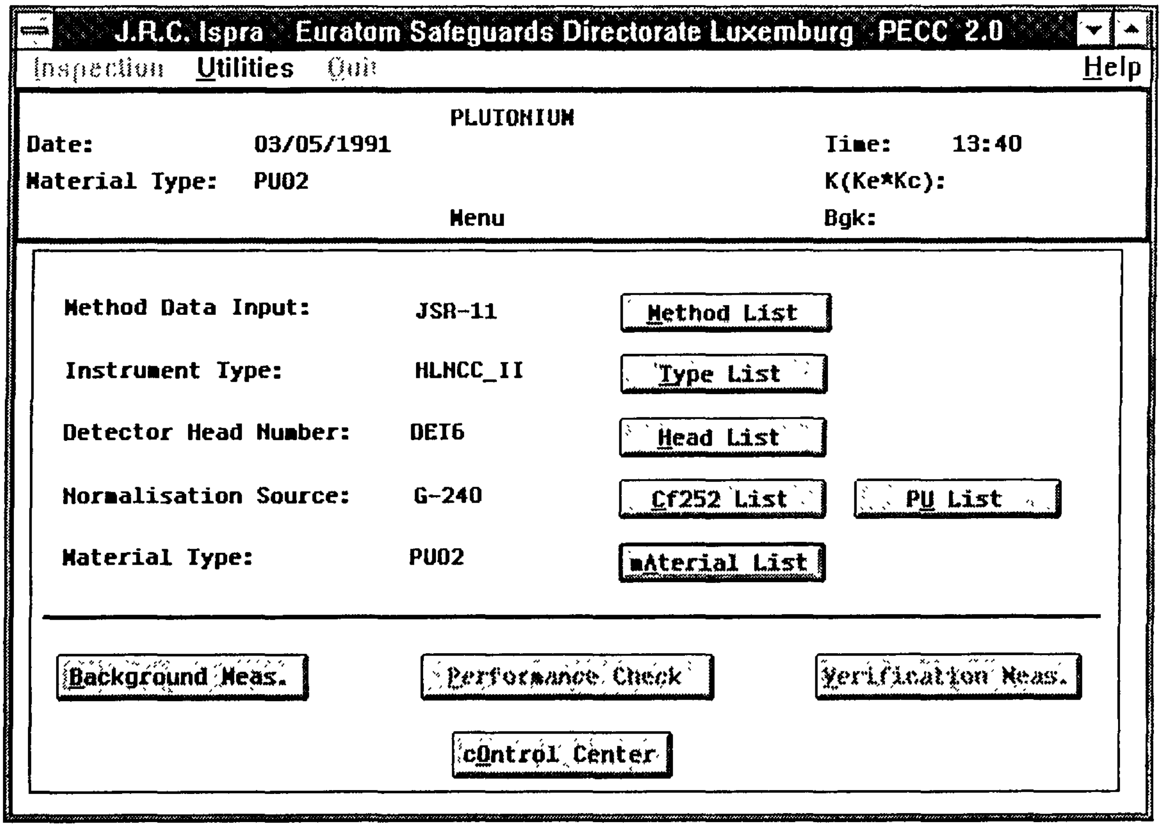
Bgk:

Rates

Totals | Totals | Real | S(R) | Sr(R)% | Out.

Lap Time:

Figure 5 - User interface : Utilities



PLUTONIUM

Date: 03/05/1991

Material Type: PU02

Menu

Time: 13:40

K(Ke*Kc):

Bgk:

Method Data Input: JSR-11

Instrument Type: HLNCC_II

Detector Head Number: DEI6

Normalisation Source: G-240

Material Type: PU02

Figure 6 - User interface : Menu

A PROGRAMME FOR EURATOM SAFEGUARDS INSPECTORS, USED IN THE ASSAY OF HIGH ENRICHED (H.E.U.) AND LOW ENRICHED (L.E.U.) URANIUM FUEL MATERIALS BY ACTIVE NEUTRON INTERROGATION.

G.P.D. Verrecchia[1], V. Vocino, N. Farese, T. Maucq, M. Nebuloni[2]

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Safety Technology Institute, I-21020 Ispra (Varese), Italy

[1]Euratom Safeguards Directorate, Luxembourg
[2]Syrea, Milan.

Abstract

The programme A.E.C.C. (Active Euratom Coincidence Counters) has been developed at the Joint Research Center, Ispra by the Euratom Safeguards Directorate, Luxembourg and the Safety Technology Institute, Ispra for the acquisition, evaluation, management and storage of measurement data originating from active neutron interrogation of H.E.U. and L.E.U. fuel materials. The software accommodates the implementation of the NDA (Non Destructive Assay) procedures for the Active Well Coincidence Counters and Active Neutron Coincidence Counters deployed by the Euratom Safeguards Directorate, Luxembourg.

Overview of the system

The Programme A.E.C.C., /1,2,3/, was developed to satisfy Euratom's requirements for a software package capable of evaluating, managing and storing measurement data acquired by the Neutron Coincidence Counter (NCC) family of detectors, /8/. The programme therefore complies with Euratom's philosophy of standardisation for Non Destructive Analysis (NDA) techniques and optimisation of the calibration activity for a series of nominal identical detector systems.

AECC (Fig. 1) belongs to a family of NDA instruments, /7/ that has been designed according to a general framework for a Safeguards integrated data evaluation system. AECC is an example of the state of the art in the NDA workstation field. This generation of NDA instruments was designed and developed by the JRC Ispra in synergy with the EC Safeguards Directorate to cover the functions an inspector performs during the field activities, /5/. In the frame of this cooperation, the AECC workstation was constructed with the following design guidelines:

- **Inspection summary**
- **Inspection procedure support**
- **Data post-processing**
- **Intelligent assistance**
- **Minimize user input**

An **Inspection**, from the user point of view, is the set of the declared and measured data collected during an inspection campaign. The system internally handles the files concerning with the background, the performance check and the sample measurements, active and passive configuration (with and without the interrogation source/s, respectively), fast and thermal configuration (with and without the Cd-liner) and correlates them to achieve the final results (Fig. 1), /6,9/. The internal software structure is hidden from the user, thus avoiding a need to remember names and parameters during the phases of data post processing or down loading to a headquarters station. The macro-operations the user is concerned with are related to the **inspection**; the system will expand them into more primitive functions that get advantage from the powerful software architecture. **All data that make up an inspection can be handled as one item. For instance to back-up or to restore an inspection from a diskette takes a single command, /10/.**

Inspection procedure support means that the user (i.e. the inspector) is guided along the activities being performed in an inspection: set up of the detector chain being used; selection of the proper normalization source and material type; performance of background and performance check measurements; and finally, the description and measurement of samples.

Data post-processing (Head quarter analysis) provides the tools to retrieve data from an old inspection and to process them. The user may verify the results or test the effect produced by changing an input parameter. The system allows the user to change any input parameter without affecting the primary data files.

Intelligent assistance means that the instrument provides powerful on line help that contains all the information the user may need during his inspection activity.

Minimize user input means that a great amount of input data can be provided to the measurement system with a minimum number of selections rather then be repeatedly entered via keyboard. The characterization parameters of the specific shift register device, detector head and normalization source used during an inspection are constant, and can be identified with names.

AECC provides a library containing the description of these objects, the material types and their associated calibration curves, /4,5,9/. Data selection is guided by the system that shows only objects compatible with the previously selected ones. In this way input data acquisition is simple, fast and safe, data are always consistent and clerical errors are avoided. Libraries are made up with standard ASCII files (Fig. 2) and therefore can be handled off line, i.e. adding an object to the library requires no changes to the program.

The programme is structured in such a way as to provide the user with a define **library of instrument types** such as the AWCC, and ANCC, /1,2/. **Calibration data** is available for various categories of high enriched uranium such as HEU metal, UO_2 powders or material test reactor (MTR), fuel elements and low enriched uranium Light Water Reactor (LWR) fuel assemblies, /4,5,8,9/.

The operating characteristics for all detectors are stored as ASCII files in data libraries together with appropriate factors used to describe the relationship between detectors of the same instrument type. The programme can thus manage and evaluate measurement data acquired by a registered Euratom detector using a **universal calibration** established for a given material category and instrument type.

The **operating performance** of any detector can be monitored using one from a library of registered Cf_{252} sources or associated Am-Li sources, /8/. The measurement data acquired during subsequent inspection activities can always be normalised if the operating performance or conditions of the detector vary significantly from the time of calibration. The programme incorporates several test procedures to assure that the instrument is functioning correctly and that no erroneous data is registered for subsequent evaluation. The criteria for the tests can be set by the superuser, /1,2/.

The programme also allows for the correction of measurement data on materials which contain thermal neutron poison following elaborate techniques established either as a part of the measurement procedure or using archive data.

The programme provides a **printed report**, (Fig. 3) including the raw measurement results, the operators declaration, and the data evaluation. It can also store the information in data files for archive purposes.

To assure proper maintenance and consistency in its application two level of access to the programme are permitted. Apart from routine access and execution by the user (Inspector), the Supervisor (Head quarters) can also modify parameter and data files to update the contents of the libraries of calibrations or detectors and perform reevaluation of archive data if required.

AECC provides a powerful **on-line help** which the inspector can access from any point of the program. Help automatically shows the operation the inspector was performing. An on demand help index is shown, and the inspector can navigate on-line help himself. The on-line help function may be used either as a user procedure or as a guide to recovering from errors. The software has a series of built in **error trapping procedures** (Fig. 4) ensuring consistency in it's execution.

AECC provides the inspector with some **utilities** that simplify operations correlated with inspection activity. The most important are:

- View File. Allows the user to select and display any file belonging either to the library or to an inspection, the displayed file can be sent to the printer.
- Make Batch File / Delete Batch File. These two utilities allow a user to introduce (or delete) the isotopic composition of a new batch. During sample analysis the inspector needs only to select the name of the batch the sample belongs to.
- Copy Inspection. These utilities allow a user to copy from Hard Disk to Floppy Disk (and vice versa) all files related to an inspection. After a copy to Floppy Disk the inspection can be erased from Hard Disk to free disk space.

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The system is built for different types of elaboration and acquisition hardware (Jomar-11, SCRD) either connected through RS232 or AT/PS2 bus link. In fact the hardware drivers are loaded during system set up according to the user requirements. This approach allows a user to add new drivers and therefore to handle new data acquisition instruments with very few software modifications.

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References

1 - V. Vocino, N. Farese, T. Maucq (CEC-JRC-Ispra), G.P.D. Verrecchia (CEC-DCS-Luxembourg) "AECC-Active Euratom Coincidence Counter. Software Requirements Part 1 : AWCC" T.N.I.90.77 September 1990

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6 - M.S. Krick (LANL) "Algorithms and software for data acquisition and analysis using thermal neutron coincidence counters within the IAEA's IFSS/NCC system" LA-UR-88-1301 April 1988

7 - R. Benoit, W. Hage, A. Prosdocimi, V. Vocino (CEC-JRC-Ispra), R. Haas (CEC-DCS Luxembourg), B.L. Thaurel (CEA-Saclay) "A computer controlled Shift Register system for the automatic assay of plutonium oxide by passive neutron interrogation" ESARDA-EUR 11041 EN may 1987

8 - H.O. Menlove, J.E. Stewart (LANL) "A new Method of Calibration and Normalization for Neutron Detector Families" LA-11229-MS April 1988

9 - H.O. Menlove, J.E. Stewart, S.Z. Qiao, T.R. Wenz (LANL), G.P.D. Verrecchia (CEC-DCS-Luxembourg) "Neutron Collar Calibration and Evaluation for Assay of LWR fuel assemblies containing burnable neutron absorbers" LA-11965-MS Final Draft

10 - G. Rossi, M. Nebuloni, G. Milani (SYREA - Milan) "AECC : Data and file system structure" Private Communication

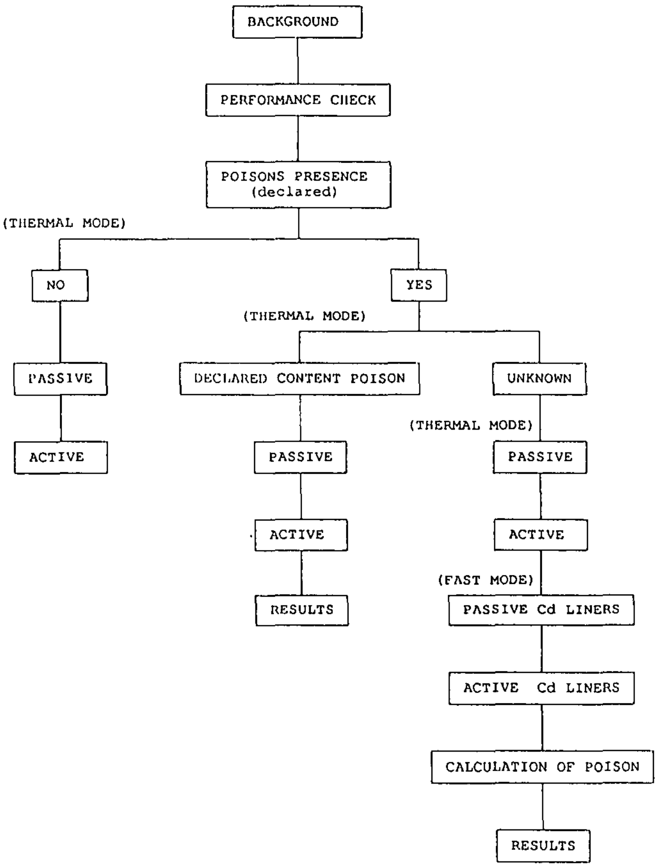


Figure 1 - Flow scheme of the programme

```
                   B A T C H     F I L E

BATCH FILE NAME:                batch_1.bch
IPU238:                         .1993
IPU239:                         70.9554
IPU240:                         24.5829
IPU241:                         3.2876
IPU242:                         .9752
IAM241:                         10207
IS_COMP_DATE:                   01/11/1987
IAM_241_DATE:                   01/11/1987
PU_DATE:                        01/11/1987
```

Figure 2 - Typical ASCII file of the programme

```
        EURA-RESCTRICTED CIRCULATION              DIRECTORATE OF EURATOM SAFEGUARDS
        Passive neutron interrogation Procedure   DCS (LUXEMBOURG) / JRC (ISPRA)
        AWCC Ver 1.00           (03/05/1991)

INSTALLATION          :prova            MBA                      :mba                INSPECTION N.   :5-11-111-esa
INSPECTOR             :nnnn        METHOD OF DATA INPUT    :KEYBOARD                 INSTRUMENT TYPE :THERMAL_2
DETECTOR HEAD N.      :EUR04       MATERIAL TYPE           :UO2 (35%)         Date            :03/05/1991

                                   INSTRUMENT OPERATING PARAMETERS
H.V.    :1700 V               Predelay (TD)   : 4.500000 s                 S.R. Gate (TG)  :64 s
Dead Time Coefficients:       a =     : 6.800000e-007 s                    b =     :2.200000e-013 s

                                          BACKGROUND COUNTING     (Instrument:  THERMAL_2 Date: 03/05/1991)
        Counting time   :100 (s)                  Number of repetitions   :1
        Measured counting rates           Total   : 35.580000 +- 0.5965 (1/s)
                                          Reals   : 0.020000 +- 0.0424 (1/s)

                              INSTRUMENT PERFORMANCE CHECK USING Am-Li SOURCE
        (Instrument:  THERMAL_2 Date: 03/05/1991)
        Counting time   :100 (s)                  Number of repetitions   :1
        Reference Totals Rate (updated)       : 17166.705703 +- 0.0000 (1/s)
        Measured Totals Rate                  : 17315.659807 +- 13.2446 (1/s)
        Ke ( electronic)                      : 0.982869 +- 0.0015 (1/s)

                                   VERIFICATION MEASUREMENT:       [SAMPLE5.000]
        (Instrument:  THERMAL_2 Date: 03/05/1991)
        Sample Identification          :SAMPLE5                      Material Description:  UO2
        Declared Enrichment (235U)     :35.0000 %           Declared 235U (g):      100.0000
        Declared Poison []       :0.0000 ppm           Declared Concentration (ppm of 235U):   0.0000

        Active lenght            :0 cm
        Counting Time:  1000     (s)     Number of repetition:   1
                T               R+A              A               Tp              Rp              sig(Rp)         sigr(Rp)
----------------------------------------------------------------------------------------------------------------------------
          2980556         983958          568558        2980.56        415.40                1.25              0.30



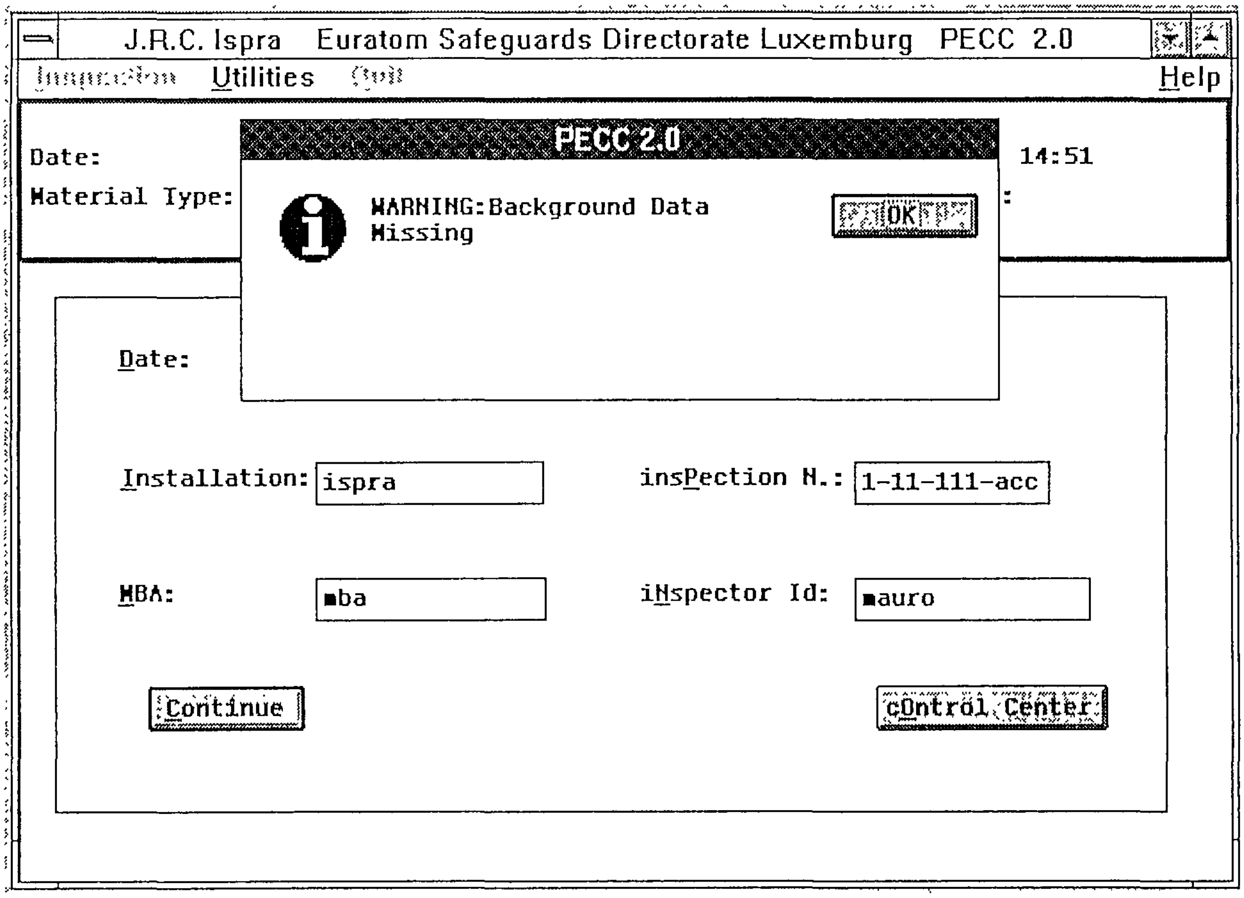
----------------------------------------------------------------------------------------------------------------------------
          2980556         983958          568558        2980.56        415.40                1.25              0.30

                              BACKGROUND, NORMALISATION AND DEAD TIME CORRECTED
        Totals Rate     :2921.141564 +- 2.8781 (1/s)            Reals Rate      :409.093450 +- 1.3782 (1/s)

        Normalisation Factor (K = Ke * Kc * Kcc * Kp )  : 0.982869 +- 0.0015 (1/s)
        Kc ( cross reference )  : 1.000000 +- 0.0000 (1/s)
        Kcc ( container cor.)   : 1.000000 +- 0.0000 (1/s)
        Kp ( poison cor.)       : 1.000000 +- 0.0000 (1/s)
                                          DATA EVALUTATION
        Declared mass 235U      (O) :100.000000
        Measured mass 235U      (I) :97.953523 +- 0.0000 (1)    Error on meas. mass 235U        (%) :0.000000
        ( I - O )       (g) :-2.046477(2)
        ( I - O ) / O   (%) :-0.000205
        N. of Sigmas (2) / (1)  :-0.020892

        Accepted result (YES/NO):       YES
        Comments                :
```

Figure 3 - Printed report of the Verification



Date:

Material Type:

14:51

WARNING:Background Data Missing

Date:

Installation: ispra

insPection N.: 1-11-111-acc

MBA: ■ba

iNspector Id: ■auro

Figure 4 - User interface : error trapping

METHODOLOGY OF STANDARD CERTIFICATION FOR CALIBRATION OF NEUTRON COINCIDENCE COUNTERS

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Abstract

The paper presents the methodology of standart certification for calibration of neutron coincidence counters. The developed approach assumes the certification of standards by the magnitude of quantity of neutron flux and spontaneous fission activity ^{240}Pu. In this case the number of needed physical standards is significantly reduced, Metrologic characteristics are given for metal specimens SP-1 and SP-2 used as primary standards of a neutron flux and spontaneous fission activity ^{240}Pu that belong to the standard complexes SRS 10-5-88 and SRS 6-18-88.

Introduction

Non-destructive methods of detection of special nuclear materials containing plutonium by the passive neutron assay (especially neutron coincidence methods) are widely used in nuclear technology both to control the processes of spent nuclear fuel regeneration and with analytical purposes as well as to provide safeguards in the inspection activities of IAEA. Large families of instruments of different types and uniformity of neutron measurements are not possible without metrological support which is based on the procurement and verification of physical standards and calibration procedures.

The widely employed method of instrument calibration using physical standards certified by different methods for the nuclear material content, isotopic and chemical compositions has some significant disadvantages.

First, with a growing variety of samples and items analyzed by neutron methods the system of physical standards needed will be highly cumbersome and expensive. Second, calibration curves that establish the relationship between the instrument response and the nuclear materials content in an item analyzed are substantially non-linear. And, third, even insignificant variations between the isotopic and chemical compositions, shape, mass, density and structure of an item analyzed and a standard can significantly influence the response of an instrument and it is quite difficult to correct in this approach unaccounting for the physical processes of a response formation.

In this connection from our viewpoint the system of a metrologic support of passive neutron methods must be based on standards certified for the magnitude of a physical quantity directly measured in an experiment i.e. a neutron flux and activity of a spontaneous fission of one of most characteristic nuclides /1.2/. Data on the isotopic and chemical compositions, mass, shape, density and structure of those standards must be considered to be reference ones.

Then, the uniformity of neutron measurements based on a procurement scheme. The magnitude of a unit of a standardized physical quantity istransferred from primary standards to secondary and working ones by the established methods.

To certify primary standards, high accuracy ones, methods (both destructive and non-destructive) must be used that ensure the minimal error in the results of measurements. The methods of certification of secondary and working standards or instruments are specified by the procurement scheme along with the guarantee errors at the selected confidence level for each level of the scheme.

This paper describes the methodology of certification of standards of a neutron flux and activity of transactinium nuclide spontaneous fission as well as the results of the implementation of such an approach using, as an example, specimens of plutonium metal containing ~90% ^{240}Pu and minimum impurities certified as primary standards of a neutron flux and ^{240}Pu spontaneous fission activity and included in the complexes of standards of a neutron flux SRS 10-5-88 /3/ and activity of radionuclide spontaneous fission SRS 6-16-88 /4/. It should be pointed out that the primary standards of a spontaneous fission activity ^{240}Pu were developed and certified for the first time in the measuring practice.

Methodology of Procurement and Certification of Standards of Neutron Flux and Spontaneous Fission Activity

^{240}Pu, ^{244}Cm and ^{252}Cf are most suitable nuclides for the production of standards. Pu is most preferable since its neutron energy spectrum and multiplicity distribution are characteristic of actually analyzed items. Besides, as compared to ^{244}Cm and ^{252}Cf, ^{240}Pu has a much larger half-life /5/, which makes it unnecessary to introduce corrections for the radioactive decay of the major nuclide in a standard specimen.

Plutonium metal specimens SP-1 and SP-2 were prepared containing ~90% mass ^{240}Pu and less than 0,3% mass impurities. After longterm investigations the specimens SP-1 and SP-2 were certified by us and D. I. Mendeleev's Institute of Metrology for the first time in the measuring practice as a primary standard of ^{240}Pu spontaneous fission activity.

The primary Standard of a Unit of Radionuclide Spontaneous Fission Activity SRS 6-16-88 is a complex of primary standards that serve to preserve and transfer a size of a radionuclide activity unit GET 6-84 to secondary and working reference standards to increase the accuracy and provide the uniformity of radionuclide spontaneous fission activity measurements. The following means of measurements form the complex:

- primary standards of radionuclide spontaneous fission activity ^{240}Pu base - SP-1 and SP-2 sources of the known isotopic composition,
- comparator - neutron coincidence counter UNS-1 /4/ serves to transfer

the size of a spontaneous fission activity unit to the secondary and working standards of a radionuclide spontaneous fission activity.

The primary standards of a spontaneous fission activity (SP-1 and SP-2 sources) serve to preserve and transfer the size of a spontaneous fission activity unit from the primary standard with the neutron coincidence counter UNS-1 to the secondary and working standards of a spontaneous fission activity in the range from 20 to $2*10^6$ Bg and to the secondary neutron coincidence counters to measure the spontaneous fission activity within 20 - $2*10^6$ Bg by direct measurements.

The method error in the neutron coincidence counter UNS-1 transfer of the activity unit size varies within 0.5-1.5% at the confidence level of 0.95 depending on the nuclide composition, shape and mass of sources verified. The relative error in the certification of the secondary and working standards of a spontaneous fission activity varies in the range of 4.0-8.0% at the confidence level of 0.95 and that for the neutron coincidence counters varies from 0.5 to 2.0%.

The secondary standards of a spontaneous fission activity are used to verify the working standards as well as to certify commercial products with high accuracy. The working standards and instruments are used for a technological control as well as for an analysis of the content of spontaneous fission nuclides in items and commercial products.

The neutron coincidence counter UNS-1 used as a comparator comprises:

- a well type neutron detector incorporating a three-circle assembly of slow neutron counters with six, twelve and eighteen counters in the first second and third circles, respectively, that are located in a polyethylene moderator around the central well as well as preamplifiers,

- units of high voltage power supply, pulse amplification, shaping, discrimination, selection and registration that provide both the integral counting of all and individual groups of counters and selection of time correlated spontaneous fission neutron in the CAMAC standard on-line with the LSI-11 computer. The unit of selection uses shift-register logic and allow the analyses of double, triple and more coincidence.

The rates of neutron counting and coincidence with the source in the central well are proportional to the spontaneous fission activity in the specified range taking account of the nuclide composition, corrections for dead time, radiation background and self-multiplication effect which determines the comparision properties of the neutron coincidence counter UNS-1 when comparing between the activities of a verified source and the primary standard.

The investigation of the metrologic characteristics of the primary standard consisted in the investigation of the properties and the certification of a primary set of ^{240}Pu base spontaneous fission sources SP-1 and SP-2 on a calorimeter UEA-5M and in the determination of standardized characteristics of the neutron coincidence counter UNS-1 used as a comparator.

The radionuclide activity value of the source is determined from the results of measurements of the energy of the radiation emitted by the source and the value of the energy emitted by a radionuclide per one event of a radioactive transformation.

To reproduce the spontaneous fission activity quantity a differential double calorimeter was used having the high sensitivity.

The procedure is based on measuring the activity of ^{240}Pu radionuclide in sources mainly through the -decay and determination of the mass fraction of ^{240}Pu radionuclide on the basis of the results of those measurements. Knowing the mass fraction of ^{240}Pu and the ratio of the half-lifes through the spontaneous fission and α-decay one finds the value of the activity A_{SF} of ^{240}Pu spontaneous fission in the source from the formula

$$A_{SF} = A_{\alpha}\frac{T_{\alpha}}{T_{SF}}, \qquad (1)$$

where A_{α} is the α-activity of a radionuclide in a source, T_{α} is a half-life through α- particle emission, T_{SF} is a spontaneous fission half-life.

For ^{240}Pu $T_{\alpha} = 6563 \pm 7$ years /5/, $T_{SF} = (1.15 + 0.03)*10^{11}$ years /6/ were chosen.

For ^{240}Pu the ratio of the specific heat fluxes W_{α} - through the α-decay and W_{SF} - through the spontaneous fission is

$$\frac{W_{\alpha}}{W_{SF}} = \frac{A_{\alpha}E_{\alpha}}{A_{SF}E_{SF}} = \frac{T_{SF}E_{\alpha}}{T_{\alpha}E_{SF}} \approx 5*10^{5}, \qquad (2)$$

where E_{α} and E_{SF} are average energies released on the α-decay and spontaneous fission, respectively.

Thus the heat flux of the ^{240}Pu spontaneous fission can be neglected.

Taking account of the fact that SP-1 and SP-2 sources have other radionuclide impurities the corresponding corrections are introduced into the results of ^{240}Pu spontaneous fission activity measurements. The content of ^{241}Am accumulated as a result of the α-decay of ^{241}Pu ($T_{1/2} = 14.64$ years (/5/) was taken into account by calculation assuming the absence of Am from plutonium at the moment of a source preparation and agrees with the results of the γ-spectrometric determination (table 1).

Table 1.

Metrologic Characteristics of Primary Standards of ^{240}Pu Spontaneous Fission Activity (SP-1 and SP-2 sources)

Source	SP-1	SP-2
Mass, g	82.93	41.46
Density, g/cm	17.10	17.10
Mass fraction of ^{241}Am, %	1.3	1.3
$^{240}A_{SF}$, Bq, (calorimetric method), P=0.99	3.640*E4	1.828*E4
Neutron flux, s^{-1}, P=0.95	8.775*E4 (1.4%)	4.290*E4 (1.5%)
$^{240}A_{SF}$, Bq, (neutron method), P=0.95	3.64*E4 (2.0%)	1.83*E4 (2.0%)
Multiplication coefficient	1.124 (0.3%)	1.095 (0.2%)

In June 1987 and May 1984 the ^{240}Pu spontaneous fission activity in Sp-1 and SP-2 sources was measured, the results of the measurement random error that does not exceed 0.2% (table 1); this points to the stability of the fission activity values of the primary standards under study and the absence of the unaccounted for radionuclide impurities from the sources. The systematic error is 4.0% and makes a decisive contribution to the total error of the certification of the primary standards through the summing up the errors in the determination of E_α - 0.1% that of T_α - 1.0% and T_{SF} - 3.0%.

The results of the calorimetric measurements of the ^{240}Pu spontaneous fission activity in sources are in good agreement with the data determined by the method based on measuring a neutron flux from sources /6/ taking account of the average neutron multiplicity $\nu = 2.156$ /5/, the contribution made by neutrons of spontaneous fission of other transactinium nuclides taking place in source, neutrons of the (α,n)-reaction on light impurity elements and the effect of neutron self-multiplication (table 1).

Along with the primary standards to certify means of measurements of the radionuclide spontaneous fission activity we use secondary standards of ^{252}Cf spontaneous fission activity based on californium sources of a SNC-type /7/. Their metrologic characteristics are listed in table 2. The systematic error of the certification is not more than 2.0% while the random one is 0.6%.

The systematic component of the method error in the neutron coincidence counter UNS-1 transfer of the magnitude of the spontaneous fission activity quantity was calculated according to GOST 8.207-76 taking account of the following:

Θ_1= 0.5% - the systematic error due to the uncertain values of the self-multiplication coefficients of neutrons in the sources /6/.

Θ_2= 0.1% - the systematic error due to the uncertainty in the dead time of neutron coincidences.

Table 2 (a, b)

Metrologic Characteristics of Secondary Standards of ^{252}Cf Spontaneous Fission Activity (as of 01.04.87)

a)

Source number	Calorimetric method A, Bq (P = 0.95)
3-6/07	1.162*E6
1-06/04	1.394*E5

b)

Source number	Neutron method Neutron flux, s^{-1}, (P=0.95)	A, Bq, (P=0.95)
3-6/07	4.40*E6 (1.4%)	1.17*E6 (1.7%)
1-06/04	5.12*E5 (2.0%)	1.36*E5 (2.2%)

Θ_3= 0.2% - the systematic error due to the nonlinearity of the neutron coincidence counter UNS-1 in the verification range.

The instability of the UNS-1 operation does not exceed 0.13% for 8 hours.

Thus, the method error in the neutron coincidence counter UNS-1 transfer of the magnitude of the spontaneous fission activity quantity is 0.5 - 1.5% at confidence level 0.95 depending on the nuclide composition, shape and mass of sources certified.

Conclusions

The metrologic characteristics of the available primary standards of a neutron flux and ^{240}Pu spontaneous fission activity show that in principle it is posssible to use them for the high accuracy calibration of neutron coincidence counters.

Besides it is anticipated that calibration procedures may be developed for neutron coincidence counters using the standards of the Pu spontaneous fission activity as well as the certification method widely used be the IAEA for measuring californium sources (of CR-5 type) by the effictive spontaneous fission activity of ^{240}Pu. Neutron detectors of the efficiency more than 45% and neutron coincidence selectors are to be used for this purpose.

To increase the accuracy of the californium source certification using the primary standards of ^{240}Pu spontaneous fission activity the real neutron leakage multiplicity distribution calculations are to be performed for the latter by the Monte-Carlo method in the multigroup approximation.

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DETERMINATION OF ^{240}PU SPONTANEOUS FISSION HALF-LIFE

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Abstract

The improvement of accuracy of nuclear physics data of transactinium nuclides, namely half-lives determination, is of vital importance for the development of nuclear physics assay and monitoring methods and ensuring their reliability. The half-life of ^{240}Pu due to α-radiation has been determined to be 6563+7 years.

The value of ^{240}Pu half-life due to spontaneous fission has not been precisely determined until recently. It varied from $1.15*10^{11}$ to $1.47*10^{11}$ years.

The value of ^{240}Pu spontaneous fission half-life was obtained using results of precision neutron flux measurements from two spherical metal plutonium samples with ^{240}Pu content of about 90% and impurities less then 0.3%.

The effect of self-multiplication, specific neutron fluxes of (α,n)-reactions and spontaneous fission of ^{238}Pu and ^{242}Pu were taken into account.

The obtained value of ^{240}Pu spontaneous fission half-life consists $(1.15+0.02)*10^{11}$ years.

1. Introduction

The improvement of accuracy of nuclear physics data of transactinium nuclides, namely half-lives determination, is of vital importance for the development of nuclear physics assay and monitoring methods and ensuring their reliability. The half-life of ^{240}Pu due to α-radiation has been determined to be 6563+7 years /1/.

The value of ^{240}Pu half-life due to spontaneous fission has not been precisely determined until recently. It varied from $1.15*10^{11}$/11-12/ to $1.47*10^{11}$ years /8/.

Integrated development of reference standard base carried out in A. A. Bochvar All-Union Scientific Research Institute of Inorganic Materials jointly with the NPO D. I. Mendeleev All-Union Scientific Research Institute of Metrology, introduction of working reference standards of the neutron flux unit SRS 10-5-88 and radionuclide spontaneous fission activity SRS 6-16-88 besides ensuring the accuracy and unity of neutron flux and radionuclide activity measurements, contributed to a great extend to the refinement of the value of ^{240}Pu spontaneous fission half-life. The value of T_{SF}^{240} = (1.15 + 0.03)$*10^{11}$ years was obtained earlier /12/ with account of the data of precision measurements of the neutron flux of Pu specimens having high ^{240}Pu content and the multiplication effect in the specimen neutron intrinsic field, the contribution of neutrons of the (α,n)-reaction on light impurity elements and spontaneous fission of other transactinium nuclides present in the specimens, as well as the value of multiplicity of neutrons, emitted during a single event of ^{240}Pu spontaneous fission, that equals (2.154 + 0.025) /14/. The value of T_{SF}^{240} = (1.160 + 0.025)$*10^{11}$ years is currently recommended elsewhere in literature /1,2/.

The determination of the ratio of Pu half-lives due to α-decay and spontaneous fission, i.e. the indirect verification of T_{SF}^{240} obtained earlier, is within the scope of this paper.

2. Materials and method

SP-1 and SP-2 specimens are (used included in SRS 6-16-88) as ^{240}Pu spontaneous fission activity standards. These specimens having a spherical shape are manufactured from metal plutonium containing ~90% of ^{240}Pu and 0.3 mass % of impurities.

Plutonium isotopic composition of SP-1 and SP-2 specimens is shown in Table 1.

Table 1.
Plutonium isotopic composition

Isotope	mass-fraction, %
Pu-238	10*E-4
Pu-239	7.71+0.01
Pu-240	89.33+0.01
Pu-241	2.79+0.01
Pu-242	0.17+0.01

The specimens contained notable amounts of ^{241}Am accumulated due to β-decay of ^{241}Pu ($T_{1/2}$ = 14.35 + 0.10) years /1/. Mass fraction of ^{241}Am determined by the γ-spectrometric method was 1.1%. This value is well matched with the data calculated with account of the assumption that the specimens contained no ^{241}Am as manufactured.

Certification of SP-1 and SP-2 activity standards was carried out on the UEA-5M unit included in the radionuclide activity state primary standard. This unit implements the calorimetric method of activity measurements that proved to be the most universal one. The reference standard certification technique is based on ^{240}Pu activity measurements in the sources, mainly due to α-decay and subsequent determination of ^{240}Pu mass fraction. The value of spontaneous fission activity A_{SF} was determined with account of ^{240}Pu mass fraction and the ratio of half-lives due to spontaneous fission and α-decay, according to the following formula:

$$A_{SF} = A_{\alpha}\frac{T_{\alpha}}{T_{SF}}, \qquad (1)$$

where

A_α - -activity of ^{240}Pu in a source,
T_α - half-life due to α -decay,
T_{SF} - half-life due to spontaneous fission.

The ratio of specific neutron fluxes W_α - due to α -decay and W_{SF}- due to spontaneous fission for ^{240} Pu is calculated as follows

$$\frac{W_\alpha}{W_{SF}} = \frac{A_\alpha E_\alpha}{A_{SF} E_{SF}} = \frac{T_{SF} E_\alpha}{T_\alpha E_{SF}} \approx 5{*}10,^{5} \tag{2}$$

where

E_α and E_{SF} - mean values of energy, released due to α -decay and spontaneous fission, respectively. Thus, the value of ^{240}Pu spontaneous fission thermal flux may be considered negligible. As the material used for SP-1 and SP-2 sources contained other radionuclide impurities, certain corrections were introduced to the results of spontaneous fission activity measurements.

Simultaneously, SP-1 and SP-2 sources were certified as neutron flux reference 1st grade standards included in the neutron flux working standard SRS 10-5-88, permission of the custodian of the radionuclide activity working standard provided /14/. The results of measurements of SP-1 and SP -2 metrologic characteristics are shown in Table 2.

The results of ^{240} Pu spontaneous fission activity measurements in the sources /15/ obtained by the calorimetric method, are in accordance with the data, obtained using the method, based on measurements of neutron fluxes from the sources /12/ with account of the mean multiplicity of spontaneous fission neutrons $\nu^{240} = 2.156 + 0.012$, the contribution of neutrons of the (α ,n)-reaction on light impurity elements and spontaneous fission of other transactinium nuclides as well as the neutron multiplication effect in the source intrinsic field (see Table 2). The measured neutron flux from the sources equals

$$Y = M_{SF} \cdot \nu^{240} \cdot N_{SF} + M_{\alpha n} N_{\alpha n} \tag{3}$$

where

N_{SF}- spontaneous fission activity,
$N_{\alpha n}$- neutron flux due to (α ,n)-reaction,
- spontaneous neutron multiplicity,
M_{SF} and $M_{\alpha n}$- multiplication factors in neutron intrinsic field due to spontaneous fission and the (α ,n)-reaction, respectively.

3. Results

Table 2.
Metrologic characterictics of the SP-1 and SP-2 reference standards at the time of measurements

Source characteristics	SP-1	SP-2
mass, g	82.93+0.01	41.46+0.01
density, g/cm	17.10+0.03	17.10+0.03
mass-fraction of Am-241, %	1.1	1.1
alfa-activity of Pu-240 A_α,Bq	6.41*E+11	3.22*E+11
relative error for A_α ,% (P=0.99)	0.1	0.2

SF-activity of Pu-240 A_{SF}, Bq (calorimetric method)	3.640*E+4	1.828*E+4
random error for A_{SF}, % (P=0.99)	0.1	0.2
systematic error for A_{SF},% (P=0.99)	4.0	4.0
SF-activity of Pu-240 A'_{SF}, Bq (neutron meth.)	3.648*E+4	1.830*E+4
random error for A'_{SF}, % (P=0.95)	0.3	0.3
systematic error for A'_{SF},% (P=0.95)	1.7	1,9
neutron flux Y, s^{-1}	8.655*E+4	4.230*E+4
random error for Y, % (P=0.95)	0.3	0.3
systematic error for Y, % (P=0.95)	1.2	1.4
multiplication factor M	1.124	1.095
random error for M, % (P=0.95)	0.3	0.2
systematic error for M, % (P=0.95)	0.2	0.2

The value of $N_{\alpha n}$ equalled 10.0+1.0 /12/. As $N_{\alpha n}$ was less than 1% from N_{SF} and the total spectrum of neutrons of the (α,n)-reaction on light impurity elements differed insignificantly from the fission spectrum, it was assumed that

$$M = M_{SF} = M_{\alpha} \qquad (4)$$

The value of ^{240}Pu spontaneous fission activity in the sources was calculated from Eg.(3) using the neutron method (see Table 2).

The ratio of spontaneous fission and α-decay half-lives was derived from Eg.(1)

$$R_B = \frac{T_{\alpha}}{T_{SF}} = \frac{A'_{SF}}{A_{\alpha}} \qquad (5)$$

The ratio equalled (5.68ö0.11)*10^{-8} that corresponds to the value of T_{SF} = (1.16+0.02)*10^{11} years at T_{α} = 6563+7 years /1/.

4. Conclusion

The obtained value of R_B is in accordance with the value of (5.7+0.1) *10^{-8}, recommended in /1/.

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LASCO: A PERFORMANCE ASSESSMENT AND TRAINING LABORATORY FOR CONTAINMENT AND SURVEILLANCE TECHNIQUES

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Abstract

The use of Containment and Surveillance (C/S) techniques for the implementation of nuclear safeguards has increased substantially in recent years.

Sealing and identification systems are used on a large scale and video surveillance is replacing gradually film camera systems. Furthermore many C/S systems are operated in an unattended mode.

In order to evaluate the performances and assurance which can be obtained from C/S systems, JRC has installed a laboratory, called LaSCo, for testing C/S components and systems, simulating, where possible, field conditions.

This laboratory includes facilities for training inspectors, for instance for reviewing aid of video pictures, use of integrated multisensor systems, evaluation of ultrasonic sealing methods.

Several years ago, JRC installed a performance laboratory for NDA, called PERLA, which is now extensively used. LaSCo is expected to play the same role as PERLA but in the field of Containment and Surveillance and by extension to non nuclear measurements (e.g. weighing, volume determinations).

The newly built facilities and the first experimental lay-out of LaSCo are described.

It is expected that industry and inspectors will make extensive use of LaSCo.

1. Introduction

Physical verification activities performed by safeguards inspectors are presently largely based on the assay of nuclear materials by measurements. These measurements are traditionally subdivided in non destructive assay techniques (NDA) and on sampling of material followed by destructive assay techniques (DA) for the determination of the chemical and isotopic composition of the nuclear materials.

Sealing and surveillance techniques are also applied extensively with the objective to assure the continuity of knowledge, after measurements have been performed, and to reduce in this way the re-verification frequency by assay methods.

Depending on the part of the fuel cycle considered, the frequency of the application of material accountancy measurements and containment and surveillance (C/S) may be very different. As an example, C/S is used extensively in reactors and limited application is made of NDA. In bulk handling facilities (e.g. where the fabrication or reprocessing of nuclear fuel is performed) accountancy (assay) measures are largely applied and the frequency of the use of C/S is much more reduced, except in the storage areas.

Important changes are taken place at present in the EC nuclear fuel cycle, in particular at the back end of the fuel cycle.

They may be summarised as follows:

a) The annual discharge of spent fuel assemblies is steadily increasing as new reactors are being installed.

b) Two major reprocessing plants will be put into operation in the 1991-94 period, increasing the potential throughput by a factor of 7 to 8 in respect to the present situation.

c) The throughput of MOX fuel fabrication for LWR is expected to increase by a factor 3, when the three or four new facilities will become operational.

The direct consequences of these changes in the fuel cycle will be that not only the flow of nuclear materials but also the spent fuel and separated Pu on store will increase in an important manner.

Furthermore, the reprocessing and MOX fuel fabrication plants are passing from pilot and prototype size to large industrial scale bulk handling facilities. This change in throughput brings with it an increased level of automation in the handling and processing of nuclear materials and, consequently, a greater difficulty of accessing this material.

This new industrial development calls for techniques integrated in the process/storage areas and more specifically a more intensive use of C/S often to be operated in an unattended way. It is, for this reason, urgent to draw more attention to the efficient and effective use of C/S systems.

The safeguards technical features of the changes in the fuel cycle have been analysed by ESARDA during the Karlsruhe specialist meeting in 1988 (1) (2).

2. Performance Assessment of Safeguards Measures

DA and NDA techniques have been applied for more than two decades for nuclear materials management and verification activities. A large effort has and continues to be dedicated worldwide to establish the performances of these techniques for the different applications in the nuclear fuel cycle.

Furthermore, many training courses are organised to familiarise and instruct inspectors in the use of NDA techniques. One may state that by enlarge well established procedures exist for the application of DA and NDA techniques and that for the existing fuel cycle a high standard of performance has been reached for applying these techniques satisfactorily. It is important to maintain and continuously improve this standard of quality and this requires important resources from R and D and support organisations. Many papers have been presented in past years on the subject of performance assessment and more particularly on the methodologies applied to define them and on target or performance values (3) (4).

If one considers the area of C/S the situation is very different. Seals and surveillance cameras have been used also for more than two decades. The performance of these techniques have not been evaluated in a systematic way.

In fact, no clear approach or methodology has been designed in the field of safeguards to express the performance/assurance of a C/S device or a C/S system.

Several attempts have been made to elaborate such a methodology (5) (6) and the IAEA has called for an expert meeting in 1989 to better formulate the problem of characterising the performance of C/S devices and systems (7).

From the existing studies, one can extract some elements which are directly related to the definition of the objectives of a performance assessment laboratory for C/S.

- Several technical characteristics define the performances of a C/S device (performance profile). This performance profile has to be compared or matched with the tasks the C/S device is expected to fulfill in a particular situation (task profile).
- Some of the technical performance characteristics can be expressed in numerical terms, even in some cases on an arbitrary scale (e.g. image resolution, illumination range and reliability). Other technical performance characteristics are very difficult to quantify numerically, such as tamper resistance, ease of application or verification of seal, and a figure of merit has to be assigned.
- The combination of the two types of performance characteristics is still an open question which should be addressed.

This has as a consequence that there is no objective way of comparing the assay techniques with C/S and one has difficulties to define clear guidelines for applying in the most efficient way these different but powerful verification techniques.

3. Objectives of LaSCo

The JRC has taken an initiative to contribute to the difficult problem of characterising the performance and the proper use of C/S. A special laboratory called "LaSCo" (laboratory for Surveillance and Containment) has been established and the following objectives have been defined:

a) performance assessment of C/S devices/systems

b) demonstration and development of effective procedures for proper use of C/S in specific applications

c) training of users for the application of different C/S techniques

Figure 1 presents a general schema for these objectives.

a) The assessment of performances will be made on C/S devices/systems or certain components, developed by JRC, other R/D laboratories and industry. Based on some experience gained at JRC with tests on fiber optic and ultrasonic sealing systems and at the EURATOM Safeguards Directorate on film camera systems, an analysis is now being made to develop a well defined schema and standard procedure for the systematic examination and testing of C/S devices under conditions representative of its final application. In other terms, one tries to define the technical performance characteristics and to establish performance profiles.

b) Demonstration of proper use of C/S. One has to define the tasks or functions C/S is supposed to perform in a particular situation and select a device which has the appropriate performance profile (match performance profile and task profile). This will be done, for instance, by preparing mock-ups of containers or some storage areas and demonstrating the specific applications of different types of C/S devices. An important additional step is to describe in well defined procedures the proper use of the C/S in the particular application considered.

c) Training of inspectors is obviously oriented to the practical applications of specific C/S techniques, including installation of sensors, signal processing, data evaluation and management.

One may expect that after a proper assessment of performances has been made and training for the use of C/S devices/systems has been delivered with appropriate procedures, test in nuclear facilities is to be performed before final implementation is decided by the inspectors.

As mentioned earlier, LaSCo, intends to contribute to a more efficient and effective use of C/S and in this sense to bridge the gap between laboratory developments and their application in an industrial environment.

This same approach is applied since four years in PERLA for the implementation of NDA techniques and EURATOM and IAEA inspectorates are making extensive use of this facility.

4. Lay-out of LaSCo

The general lay-out of the LaSCo facility, constituted of a large hall, is presented in Figure 2.

The hall (260 m^2) is subdivided in different parts:

- training room which can accommodate up to 12 persons for general presentations or exercises;
- advanced storage area. A mock-up is assembled corresponding to horizontal and vertical storage cells of, for instance, PuO_2 cans or assemblies. This mock-up is being equipped with different C/S devices and remote handling equipment;
- control room for multisensor C/S systems of LaSCo and for the advanced storage area;
- general purpose laboratory;
- demonstration area for different sealing/identification and optical surveillance systems now being developed or applied, with display of instruments and explanatory posters.

The laboratory, as illustrated above, has been completed in January 1991 and the experimental set-up are now being installed.

5. Planned Activities

The activities planned for LaSCo have been defined in cooperation with C/S developers and Safeguards Inspectorates and are now briefly illustrated.

a) In the advanced storage area, eight real size storage "cells" are being mounted and equipped with different types of seals (fiber optic, ultrasonic) and means of identification (bar codes.....).

The containers in the cells will also be uniquely identifiable and sealed.

Remote handling of cans will be tested using a computer aided teleoperation vehicle equipped with cameras for navigation and visual items identification. An arm is mounted on the vehicle for the extraction of samples (e.g. cans) out of a cell and simulating its positioning in front of a detector. A description of the characteristics (hard and software) of the teleoperated vehicle and its potential use has been made earlier (8) (9).

Another subject in this context is the continuous and automatic verification both of the storage area and of the specific items. Two devices are being installed for this purpose. They are the multi ultrasonic sealing system (called VALSEUR) applied on each cell and/or container and a multi video camera system. They will be both integrated in a modular multisensor system still to be developed.

The experimental lay-out of the advanced storage area will provide in general the opportunity to study C/S systems based on different combinations of new and traditional C/S devices and to perform long term test of these single devices.

b) A multi sensor system is being assembled within the LaSCo hall for the surveillance of areas defined as "strategic". This system is similar to CAVIS (computer aided video system), now being used by EURATOM Safeguards Inspectors. It will serve the purpose of demonstrating its capabilities, to perform long term testing, to provide training to inspectors and to test some components when upgrading is required.

The recently developed laser beam surveillance system will also be installed for long term testing.

c) A study is being made to compare different general purpose fiber optic seals under development or already in use. The performances will be tested systematically under different environmental conditions defined in a standard procedure. Furthermore, the procedures for the application of the different seals, including the data evaluation, will also be compared.

d) Long term tests of the TITUS ultrasonic general purpose seal are being started and the identification/re-identification procedures are under preparation.

e) The bulk measurement laboratory operated by IAEA at Vienna Headquarters for several years, has now been transferred to JRC, Ispra. This laboratory is intended to test measurement techniques for the determination of masses of liquids in tanks. The measurement techniques considered are scales and meters for the determination of flow, levels and density, humidity and temperature. Training of inspectors and test of measurements procedures is the main purpose of this laboratory. The experimental set-up is temporarily installed in LaSCo and not directly related to C/S, but it offers a good opportunity to reflect on authentication of this type of measurement techniques, using for instance a surveillance system.

Other initiatives are being studied for LaSCo:

- Reviewing of surveillance tapes is becoming of great interest, particularly in view of its potential to reduce manpower to perform this type of activity. An attempt is made to collect tapes with representative scenes of industrial operations which are of safeguards relevance. These tapes will be used as the basic material for the training of inspectors.

6. Conclusions

The use of C/S for verification activities is steadily increasing and the important changes at the back end of the nuclear fuel cycle will influence, to a large extent, future development and application of identification and surveillance techniques.

It is important to characterise the performance and the assurance which can be gained from C/S systems and from the combination of assay methods (DA/NDA) with C/S.

The JRC, Ispra has established a laboratory, operational since January 1991, which has the intention to contribute to the assessment of the performances of C/S, to demonstrate the effective and efficient use of these techniques and to provide training to inspectors.

The present lay-out of the laboratory permits to study specifically the application of C/S in storage areas and to perform tests on a variety of general purpose seals.

A second group of experiments are being made on volume/weight measurements in tanks and are provisionally installed in LaSCo.

Contacts are being established with safeguards inspectorates and potential users of industry to develop a comprehensive programme for future years.

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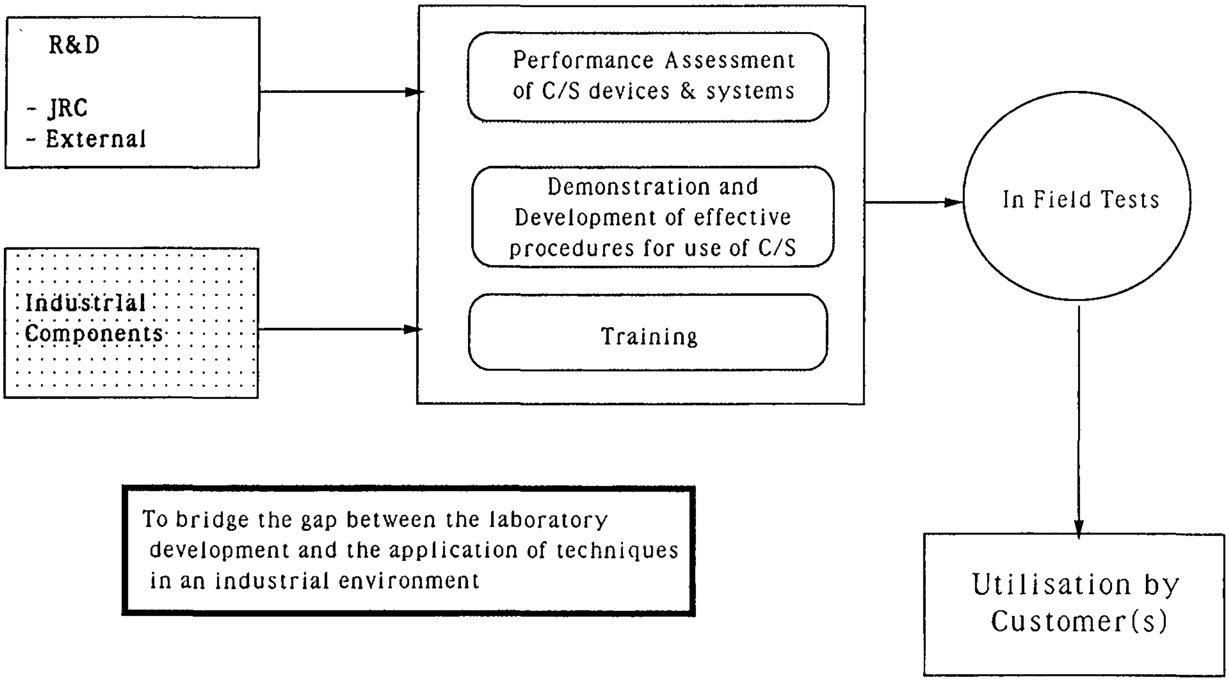

Fig. 1 - Objectives of LaSCo

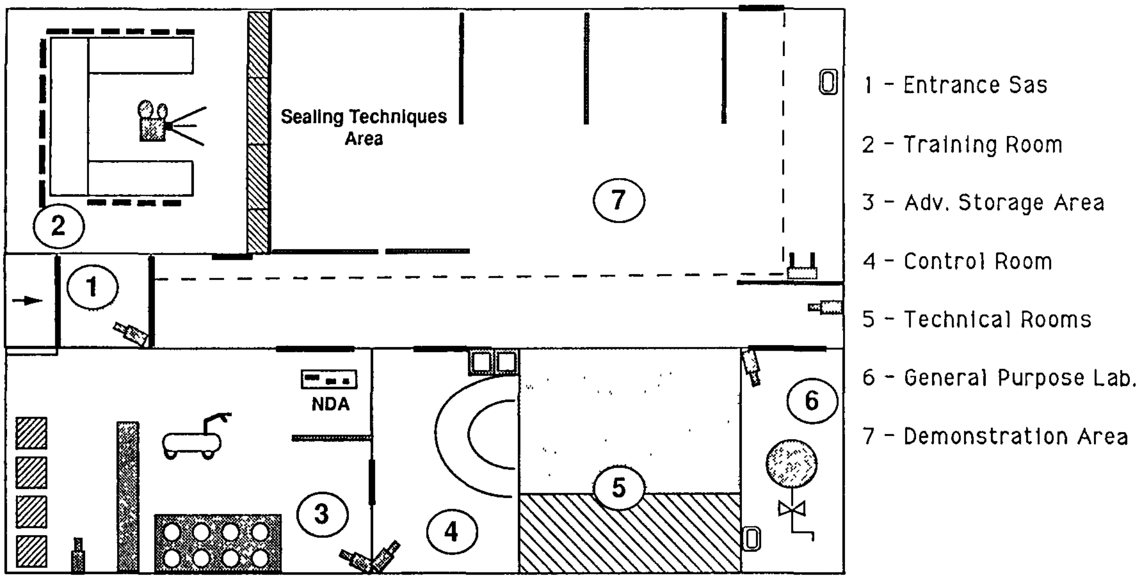

Fig. 2 - Lay-out of LaSCo

THE MULTI-CAMERA OPTICAL SURVEILLANCE SYSTEM (MOS)

Design and Reliability

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ABSTRACT

The transition from film camera to video surveillance systems, in particular the implementation of high-capacity multi-camera video systems, results in a large increase in the amount of recorded scenes. Consequently, there is a substantial increase in the manpower requirements for review. Moreover, modern microprocessor controlled equipment facilitates the collection of additional data associated with each scene. Both the scene and the annotated information have to be evaluated by the inspector. The design of video surveillance systems for safeguards necessarily has to account for both appropriate recording and reviewing techniques. An aspect of principal importance is that the video information is stored on tape. Under the German Support Programme to the Agency a technical concept has been developed which aims at optimizing the capabilities of a multi-camera optical surveillance (MOS) system including the reviewing technique. This concept is presented in the following paper including a discussion of reviewing and reliability.

1. Introduction

The development and implementation of video systems has led the safeguards authorities to the revision of their requirements for optical surveillance systems. They can be briefly described as follows:

- Many applications suggest the use of more than one video camera in one nuclear facility, i.e. the video surveillance system should have many cameras.
- Secondly, the video tape should have a high capacity to enable the recording of many scenes per camera in order to facilitate long surveillance periods.
- The recording unit for the video scenes from all cameras should be located in one safeguards control room.
- The multi-camera system should have a very high reliability taking into account its unattended operation.
- The video signals should be authenticated.
- The operation of the system should be standardized and user-friendly including the retrieval of the video data and other safeguards relevant information.

Under the German Support Programme to the Agency a technical concept has been developed which aims at optimizing the capabilities of a multi-camera optical surveillance system including the reviewing technique. This concept is presented in the following paper including a discussion of reviewing and reliability.

2. System Description

2.1 General

The Multi-Camera Optical Surveillance (MOS) System excluding the cameras is designed to be installed in a safeguards control room of a nuclear facility with access restricted to inspectors, whereas the cameras are mounted in different locations of the plant and connected to the MOS System by standard transmission lines, e.g. coaxial or fibre optic cables which can be accessible to anyone. The MOS System shall provide long term unattended video surveillance. The number of camera channels is 8 but can be expanded to 16. The basic features are summarized as follows :

- Authentication of video signals of each camera channel using the Tamper Resistant TV-Link /1/
- Individual timing of recording of each camera channel
- External triggering e.g. using video motion detection
- Immediate storage of a detected i.e. externally triggered scene or of a programmable sequence of scenes at a different timing interval
- Interfacing of the VACOSS-S electronic safeguards seal /2/
- User definable text insertion for externally triggered scenes
- Electronic notebook for relevant inspection-period data
- Testing capability for a video motion detector
- Review station.

2.2 The Modules of MOS

MOS has a modular design which enables flexibility in configuration and maintenance and allows for the introduction of improved technology as it appears on the market. MOS complies with the European monochrome TV-standard CCIR and is adaptable to the US monochrome TV-standard EIA. The mechanical standard for the console meets the international 19" rack-mount requirements. Electrical and safety standards are met according to the requirements of the Federal Republic of Germany. The basic components of MOS are

- Tamper Resistant TV-Link (TRTL)
- Buffer memory for video scenes
- Recording Unit, remotely controllable Super-VHS video recorder
- Review Station
- System Application Architecture software platform.

For immediate checking of the MOS performance the built-in recording unit can be used at the expense of loss of surveillance during review, i.e. for inspection purposes a separate review station is necessary.

In spite of its modular design the MOS System appears to be a well integrated, complete video surveillance system.

2.3 The Authentication of Video Images

The basis of the MOS System is the Tamper Resistant TV-Link (TRTL) which is designed to authenticate video signals. In order to authenticate electronic data, in particular video signals, two conditions have to be fulfilled:

- Time authenticity, i.e. data recorded in the past are not valid.
- Authenticity of the image, i.e. the data recorded must be an exact mate of the camera data (TV-signal).

To the end of ensuring the time correctness of a video signal, there has to be a timer and a transmitter unit (MOS-TRU) on the camera side, i.e. the camera and the MOS-TRU including the timer must be housed together in one sealed enclosure. The MOS-TRU feeds the 'camera time' together with the video signal into the transmission line. At the receiving end of the transmission line there is a timer in a receiver unit (MOS-RU). The MOS-RU compares the 'camera time' with its own generated time, the 'receiver time'. The timers of MOS-TRU and MOS-RU are synchronized.

To ensure that the recorded video signal was originally generated by the video camera, the MOS-TRU measures the amplitude of the camera video signal within a defined

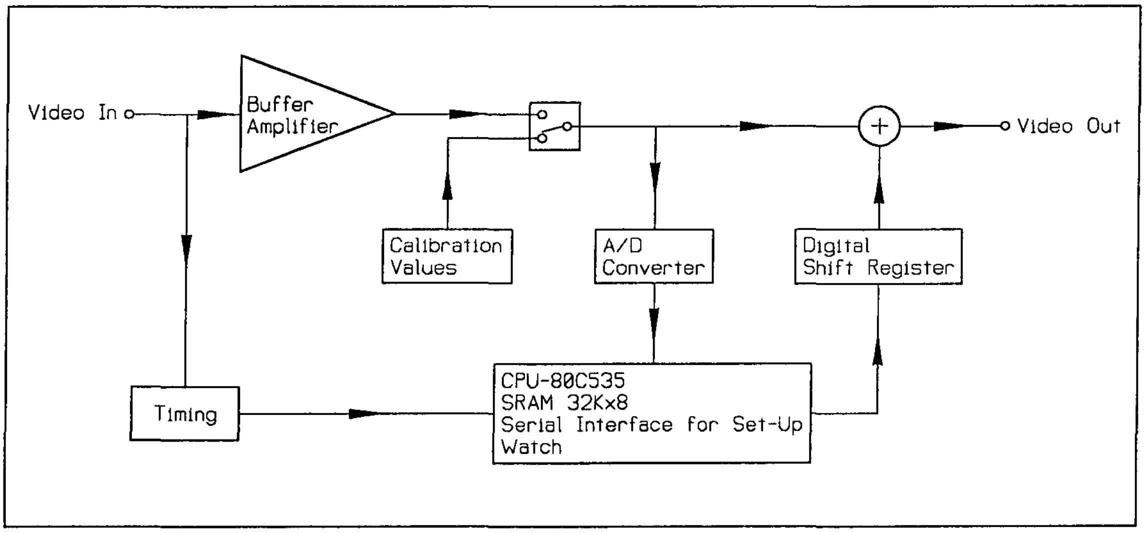


Fig. 1: **Block Diagram of the Transmitter Unit**

segment of the picture, encrypts this information and transmits it to the MOS-RU together with the video signal itself. For decryption the MOS-RU applies the same algorithm to the image data and compares the two data sets. Of course, the processed picture segment varies in position on a random basis. For encryption the Data Encryption Standard (DES) Algorithm is applied. A more detailed description of the authentication method can be found in references /1/.

3. Description of the Components

3.1 Transmitter Unit (MOS-TRU)

Fig. 1 shows a schematic block diagram of the MOS-TRU. The camera video signal is fed through the buffer amplifier and to the input of a timing device, which controls the electronic switch and the digital circuitry (CPU-80C535, digital shift register, etc.). Depending upon the position of the electronic switch, the MOS-TRU feeds either the video signal or constant grey levels into the video cable, which is connected to the video output. The constant grey levels are necessary to calibrate all analog amplifiers in the system, i.e. to compensate for varying transmission parameters of the analog circuitry. This automatically includes the compensation of different cable lengths between MOS-TRU and MOS-RU pairs. Like the authentication signal itself, these grey levels are part of the video image but are normally invisible. With the analog-to-digital converter the micro processor measures the amplitude of small segments of the video image, encodes this information with the time, the x-y position of the segment etc., and stores the results in the digital shift register.

The present MOS-TRU is designed as a package of three printed circuit boards in surface mount technology. These boards are enclosed in a small aluminium box, which can be inserted into a sealed camera housing with a mounting bracket. A complete MOS-TRU (dimension of 64.7mm (width) x 42.3mm (height) x 99mm (depth)) with the shockproof shipping container is displayed in Fig. 2.

Apart from the authentication electronics, a power supply for a CCD-camera (12 VDC nominal) is integrated into the MOS-TRU module. The module itself is powered with 15 to 40 Volt DC.

In addition to the video-in, video-out, power-in and power-out connectors, there is also a connection for a 'tamper switch'. The MOS-RU monitors the position of a connected micro switch inside the camera housing (open/closed).

The DES Algorithm requires an encryption key to be available both in the MOS-TRU and MOS-RU. For reasons of tamper resistance the key is changed after 24 hours. A Transmitter-Receiver pair is loaded with identical keysets for the encryption and decryption of the transmitted authenticating information. The downloading procedure of the keyset into MOS-TRU and MOS-RU is performed by direct interconnection of both units.

The most important principle of the authentication electronics is that the reliability of the MOS-TRU is not

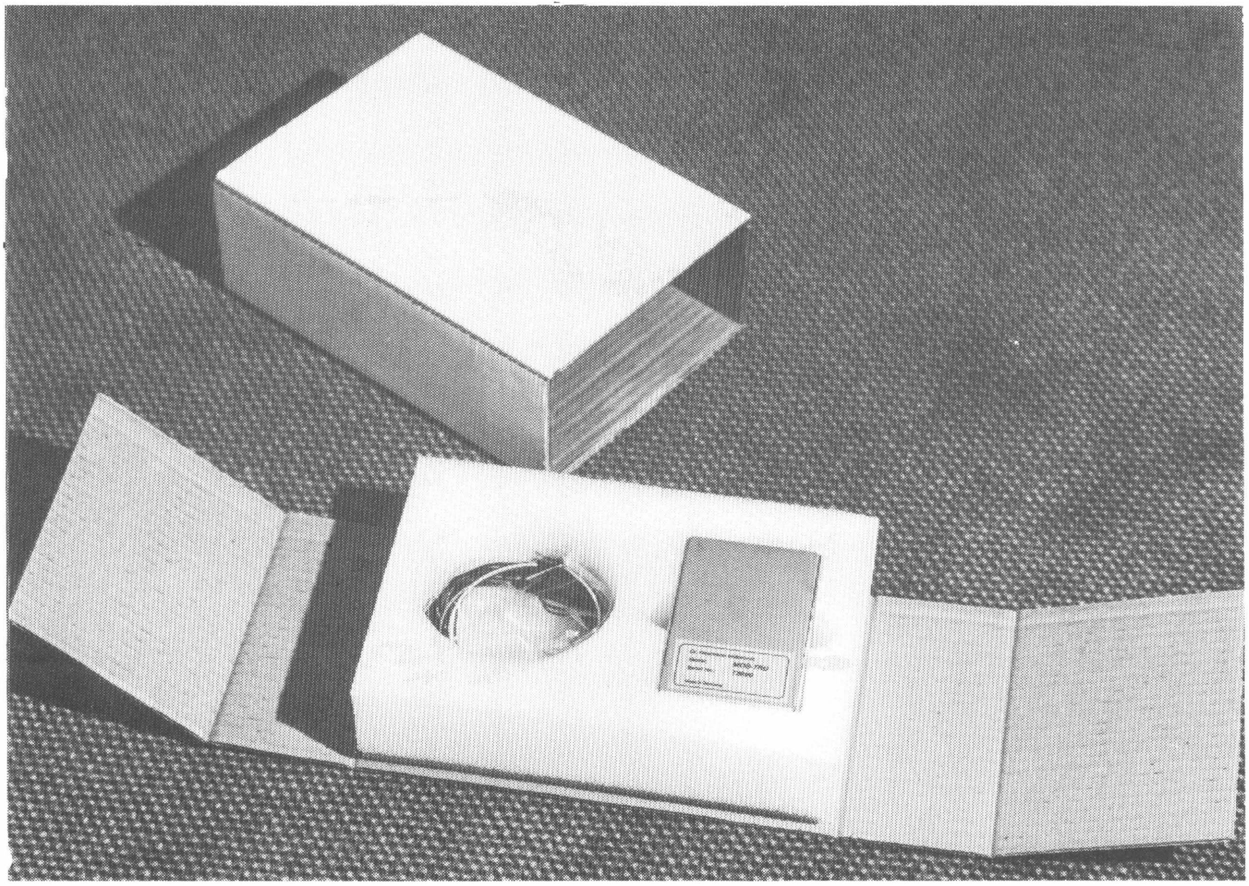


Fig. 2: **Photo of the Transmitter Unit in its Shipping Box**

affected at any time. For this purpose an extensive use of electronic monitoring is an essential part of the system. If the authentication electronics fails or even the power supply for the MOS-TRU, the video signal is transmitted to the MOS-RU without authentication, assuming the CCD camera is powered separately. The same principle applies to the MOS-RU.

3.2 Receiver Unit (MOS-RU)

The MOS-RU is the functional counterpart of the MOS-TRU and hence has a very similar authentication circuitry.

The MOS-RU printed circuit board provides space for a plug-in cable correction amplifier, if the MOS-TRU is installed at distances longer than 50 m from the MOS-RU.

The inspector is interested in events such as missing data or a tamper attack, detected during an inspection period of typically 1 to 3 months. For this purpose the MOS-RU inserts relevant text into the video image for evaluation during review, i.e. the video monitor is a sufficient output device.

The MOS-RU is a bus-oriented device operating on two video buses. One video bus can be connected to the recording device (image memory MOS-IVS, VCR) and the other one can be connected to a monitor for easy inspection of picture quality (one button operation). In a multi-channel system no video switcher is required. Up to 16 MOS-RUs can share one video bus. This means that a multi-channel authentication system is a multi-channel surveillance system at the same time. If the multiplexing capabilities of the system are not required, it is possible to use an additional constantly active video output of each MOS-RU module.

3.3 Network Controller (MOS-NET)

A multi-channel system is a network of 'intelligent' devices. They communicate with each other via a party-line. The master device is the Network Controller (MOS-NET).

The MOS-NET is designed to distribute information to a communication network (party-line) and to interface a PC to the system. Furthermore, the Network Controller reports all event data of one inspection period using an electronic notebook with a battery backed-up RAM. For reasons of redundancy the electronic notebook records the same event data which are inserted into the video images and recorded on tape.

The Network Controller also supports the MOS-RUs with real random numbers (based on white noise) for the key download procedures. The system time is also generated on the MOS-NET board and communicated through the party-line to all units. Also the power supply for the complete unit is integrated into the Network Controller.

3.4 MOS Recording Modules

Contrary to other multi-channel video systems the MOS System employs a 'normal' video recorder rather than a time lapse recorder. The Panasonic AG7330 Super-VHS VCR has been chosen for reasons of high resolution. In addition, this VCR has the advantage of being fully remotely controllable which implies that the user of the MOS System will only touch the tape cassettes but not the function keys of the VCR.

In conclusion, the user does not work with single components or modules but rather with a system as a whole. This has also an impact on future design changes or improvements of the system. For instance, the VCR can be replaced by digital recording media as they become available without the user being affected at all. There will be no need for the inspector to go through a new training programme.

The combination of S-VHS recorder and digital buffer image memory MOS-IVS32 allows individual timing of each camera channel; i.e. scenes from camera #1 can be recorded at one-minute intervals and scenes from camera #2 at 30-minute intervals. In addition, there may be individual changes of the recording sequences due to external conditions such as high gamma counting rates. The interaction between the VCR and the MOS-IVS32 can be described as follows:

The MOS-IVS32 memory stores up to 32 scenes. A MOS-RU transmits a storing command and a corresponding video signal via the video bus to the MOS-IVS32 memory. Associated with each scene the MOS-IVS32 stores the camera-channel and a scene number. When the MOS-IVS32 has stored 32 scenes it starts the VCR and dumps all the scenes onto video tape including channel numbers and scene numbers. In other words, the scenes are encoded and stored on tape. The code is displayed in the first unused lines of each video image.

Without special measures such a procedure results in a great waste of tape, because gaps come up between the dumps. A tape full of gaps could hardly be reviewed and evaluated, since the noisy gaps would affect the

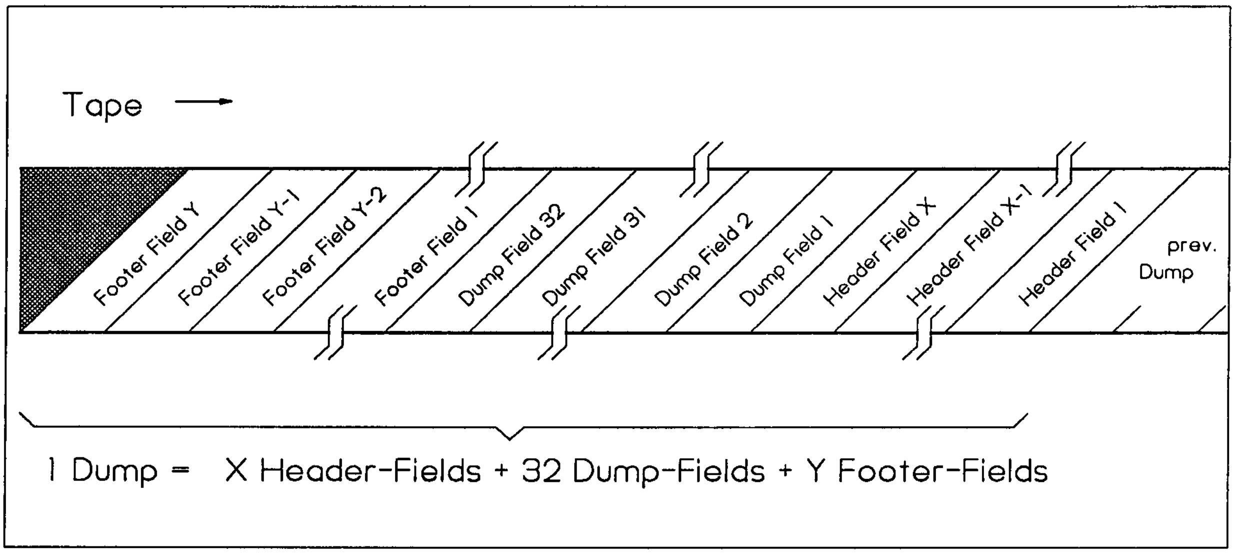


Fig. 3: **Schematic of a Dump**

synchronization of the monitor. Therefore, a so-called backspace editing has been introduced, in order to efficiently utilize the tape and to avoid gaps.

Each dump, i.e. sequence of 32 scenes stored in the MOS-IVS32, is supplemented with footers and headers, i.e. the first and the last scenes of a dump are copied several times and encoded (see Fig. 3).

The header and footer fields carry a special code and a dump number. Before a sequence of 32 scenes is stored resp. dumped on tape, the MOS-IVS32 traces the end of the last dump.

In the reliability test, this method of indicated dumps turned out to be very safe. No dumps are lost due to overwriting. There are no gaps on the tape, and the waste of tape can be controlled by the number of header and footer scenes as well as the storage capacity of the MOS-IVS32.

4. System Operation

The MOS System is largely computer-oriented. Installation and set-up of the system are completely realized by means of a PC. The inspector can use his own laptop computer. During the surveillance period of the MOS System there is no need for a PC, i.e. the PC is not hooked on.

4.1 Set-up of the MOS-System

To set up a complete system, a PC has to be connected to the Network Controller (MOS-NET). With a menu-driven installation software running on the PC, all required data can be transferred to the communication network via the Network Controller (MOS-NET). The system data consist of:

- Cable correction amplifier set-up (length and type of cable)
- Dedicated text for insertion into each MOS-RU
- Position, width and height of text within the video image
- Position of inserted time and date within the video image
- Multiplex time for each individual channel.

Although this data set may be different for each MOS-RU, each MOS-RU stores the system data of the whole system, i.e. also the system data of its neighbouring MOS-RU. The set-up data of each device is stored in a dedicated nonvolatile memory and in the RAM with battery back-up of the MOS-NET.

If a device has to be replaced due to failure, the new device will automatically receive system data for its own set-up once it has been plugged in. In case of a system breakdown, only one MOS-RU needs to survive to guarantee system data communication to the new modules.

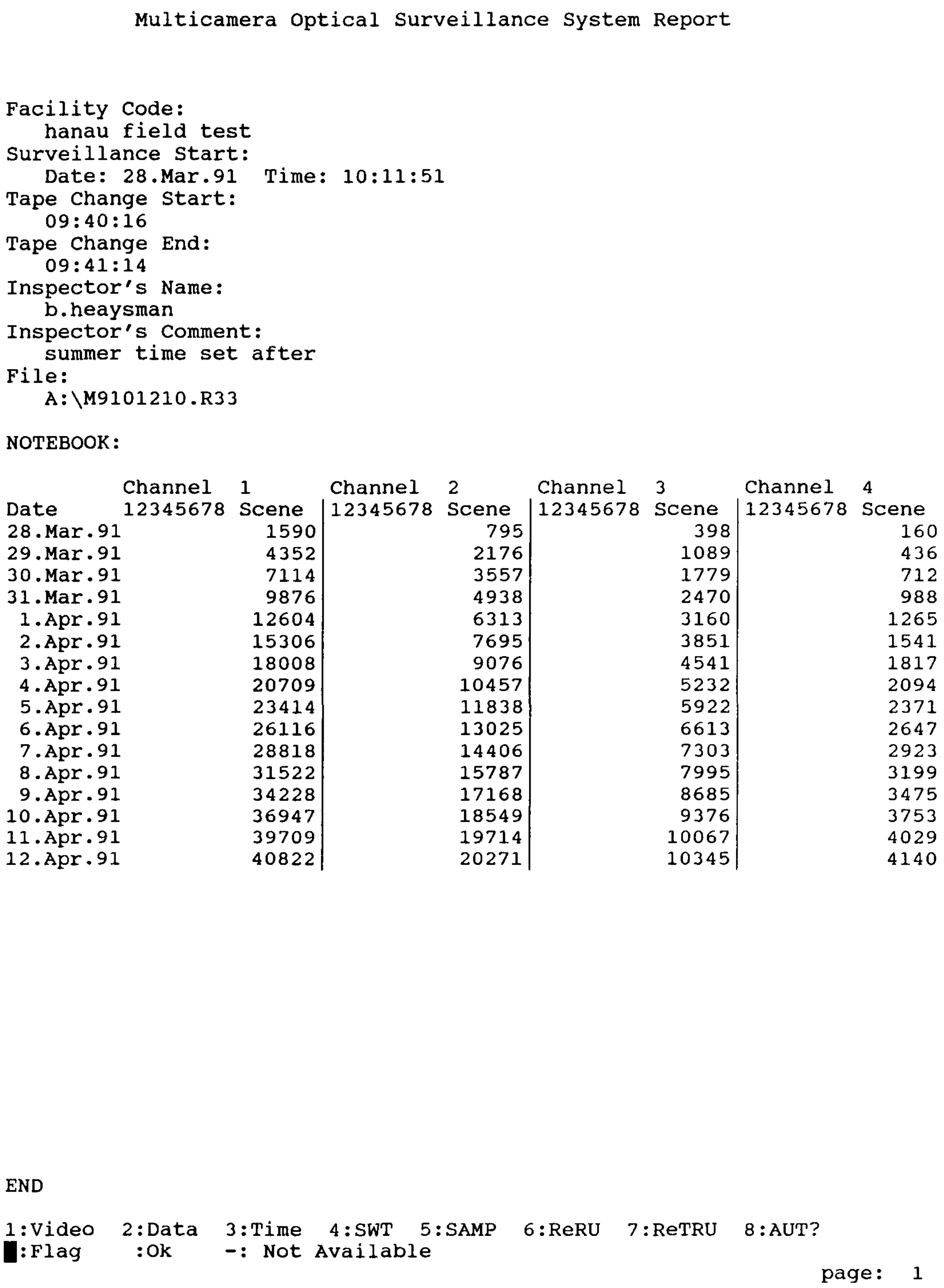
The authentication signal is encrypted in the camera's MOS-TRU and decrypted in the MOS-RU using the Data Encryption Standard (DES), patented by IBM. This algorithm requires a 64 bit key which is changed every 24 hours in both the MOS-TRU and MOS-RU. Upon set-up keys are loaded into a MOS-TRU/MOS-RU pair for an operating period of at least 5 years. These keys are generated by the Network Controller (MOS-NET) with a real random generator (noise generator). To download a 5 year keyset, a MOS-TRU/MOS-RU pair is interconnected. This direct connection must be performed before startup of a surveillance period. This can be done at Agency Headquarters, but then the MOS-TRU/MOS-RU pair has to be clearly labelled and sealed.

A complete installation of the system data can be done within one minute. The downloading of the keyset takes one minute per MOS-TRU/MOS-RU pair.

4.2 Start of a New Surveillance Period

The inspector has a great deal of information at his disposal by connecting a PC to the Network Controller of the system. Within seconds he can retrieve all information stored in the electronic notebook giving him an overview of all events which have occurred during the unattended surveillance period. He furthermore has the information that a camera or a power supply may have failed. In addition, these data are automatically stored on a standard floppy disk for further evaluation at Agency Headquarters or in a regional office. A typical printout of a notebook is shown in Fig. 4.

Furthermore, the system application programme asks the inspector if a tape change should be done. On request the system automatically ejects the tape(s) and prints out if a proper tape change was performed.



```
          Multicamera Optical Surveillance System Report


Facility Code:
   hanau field test
Surveillance Start:
   Date: 28.Mar.91  Time: 10:11:51
Tape Change Start:
   09:40:16
Tape Change End:
   09:41:14
Inspector's Name:
   b.heaysman
Inspector's Comment:
   summer time set after
File:
   A:\M9101210.R33

NOTEBOOK:

         Channel  1      Channel  2      Channel  3      Channel  4
Date     12345678 Scene |12345678 Scene |12345678 Scene |12345678 Scene
28.Mar.91          1590|             795|             398|             160
29.Mar.91          4352|            2176|            1089|             436
30.Mar.91          7114|            3557|            1779|             712
31.Mar.91          9876|            4938|            2470|             988
 1.Apr.91         12604|            6313|            3160|            1265
 2.Apr.91         15306|            7695|            3851|            1541
 3.Apr.91         18008|            9076|            4541|            1817
 4.Apr.91         20709|           10457|            5232|            2094
 5.Apr.91         23414|           11838|            5922|            2371
 6.Apr.91         26116|           13025|            6613|            2647
 7.Apr.91         28818|           14406|            7303|            2923
 8.Apr.91         31522|           15787|            7995|            3199
 9.Apr.91         34228|           17168|            8685|            3475
10.Apr.91         36947|           18549|            9376|            3753
11.Apr.91         39709|           19714|           10067|            4029
12.Apr.91         40822|           20271|           10345|            4140







END

1:Video  2:Data  3:Time  4:SWT  5:SAMP  6:ReRU  7:ReTRU  8:AUT?
█:Flag    :Ok    -: Not Available
                                                         page:  1
```

Fig. 4: **Printout of the Electronic Notebook**

5. Review of MOS Surveillance Data

5.1 Review with the Recording System

The MOS System stores the scenes from all camera-channels on one video tape as they occur. To the end of facilitating the review of the tape, measures for channel sorting have to be taken.

A crucial component is the MOS-IVS32 digital memory. It has an operating mode which switches the VCR automatically to playing while only scenes of a preselected camera-channel will be stored in the MOS-IVS32 buffer memory and displayed on the monitor. The user has the possibility to operate the MOS-IVS32 in the following way: If he detects an interesting scene, he can stop the recorder by pushing a button. Then he can retrieve the 31 scenes immediately preceding the scene of interest which are stored in the MOS-IVS32 buffer. Pushing the button once more, the interrupt is cancelled and the review of the preselected camera-channel resumed.

In conclusion, the MOS-IVS32 functions as a buffer store both during recording and reviewing. This possibility of review exists in each configuration of a MOS System. However, it should be mentioned that if the recording unit is used for reviewing, the MOS System will not record new surveillance data.

5.2 MOS Enhanced Review Station

The review of tapes of a multi-channel surveillance system is most time consuming and requires a large inspection effort. To the end of facilitating the review, an Enhanced Review Station has been constructed which has the following components:

- 1 digital buffer memory MOS-IVS32
- 1 screen splitter
- 1 video recorder
- 2 TV-monitors
- 1 PC including a dedicated system application software.

The inspector is able to select scenes according to time, date, and channel number by a simple mouse-click. The functions of the digital buffer memory MOS-IVS32 as described in section 5.1, are realized in the PC's dedicated system application software.

By means of the screen splitter scenes from 4 different camera channels can be simultaneously displayed on the monitor. The use of this capability depends on the set-up resp. views of the different cameras.

Moreover, the Enhanced Review Station can be used to perform an automatic analysis of a tape. The analysis consists of comparing the actually recorded scene id-number with the expected scene id-number, performing a tape statistics and counting the number of dumps.

6. Reliability

A test programme was agreed upon which focused on both the reliability of the authentication electronics of the MOS System and failure-free recording of scenes. According to the MOS specifications a failure is defined as

> the missing of two consecutive scenes on any channel of the MOS System. A recorded scene is accepted only if it is authenticated. This implicates MOS as a whole, i.e. the cause of a missing scene is immaterial.

According to the agreed functional and technical specifications of the MOS System the reliability statement prescribes a reliability of 96.8% for 200,000 scenes recorded by one MOS unit with eight cameras. In this statement the time span is expressed in terms of the number of scenes. Based on statistical considerations, the plan for the reliability test was designed in a way to confirm 96.8% reliability with 70% confidence. According to this plan, 252,000 dumps equivalent to 8,064,000 scenes were to be recorded without a failure.

6.1 Hardware Layout

The hardware layout depended on the total number of scenes to be recorded as well as the available testing time. The test period was agreed to be not more than 100 days. Six MOS Systems with four camera-channels each were installed in two cabinets, i.e. altogether 24 camera-channels were used (see Fig. 5). Four cameras were focused on a clock and their video signals led independently via four distribution amplifiers to three MOS Systems, covering 12 camera-channels. Camera #1

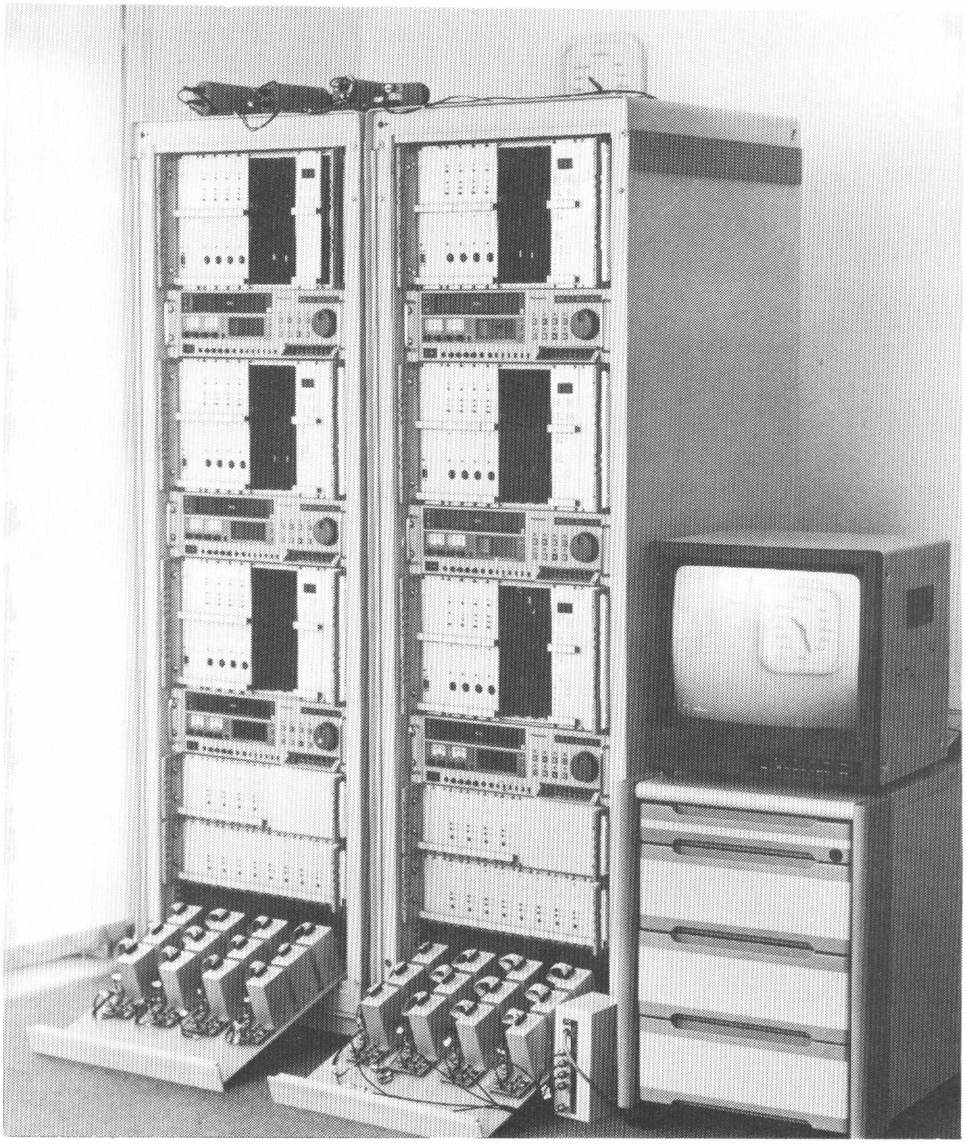


Fig. 5: View of the Testing Set-up

served channel #1 of all three MOS Systems, camera #2 served channel #2 of all three MOS Systems, etc. . The video signals of a fifth camera were branched through all 12 channels of MOS Systems # 4 through 6.

6.2 Test Results

The programme started on September 10, 1990. It took us nearly two complete runs of 8 million scenes with a lot of software changes and enhancements in the MOS-IVS32, to come up with a dump procedure (see section 3.4) which fulfilled the reliability requirements. In the first test phase, we introduced a total of 100 tamper attacks into one channel of one system at different times of the day during 10 days altogether. The success rate for the detection of all types of tamper attacks was 100%.

During the whole test period of September 10, 1990, and January 16, 1991, no false alarm was recorded in any channel.

The last phase of the test yielded 42 video tapes (S-VHS 180min), i.e. seven tapes for each MOS System, within 23 days. This was an accelerated test which put more stress on the VCR's than under usual surveillance conditions in a facility. The dumping rate was about 1 minute per dump which currently is the limit of the MOS System.

Altogether 262,420 dumps were recorded during this last phase, exceeding the required number by 10,420 dumps equivalent to 333,440 scenes. Careful evaluation of the tapes yielded no MOS failure.

Another result is that the capacity of the used 180-minute video tape is at least 200,000 scenes.

Finally, attention is drawn to the fact that the authentication function of the MOS System, known as the Tamper Resistant TV-Link, was successfully field-tested in a nuclear facility. The results of this test are presented in reference /3/. At present, a field test of a four-channel MOS System with redundant recording is being carried out in a German facility.

7. Summary

The Multi-camera Optical Surveillance System (MOS) has a lot of features that enable the Inspectorates to reduce inspection efforts without loss of credibility. The system has been designed to collect a maximum of information on a high reliability level and to facilitate retrieval as well as evaluation of the data.

- The recorded video information is credible because it is authenticated.
- MOS has a number of self-diagnostic features. The results are retrievable for evaluation.
- Front end data reduction can be implemented by the use of external triggering, e.g. video motion detection (VMD). The proper functioning of the VMD is checked by a built-in self-testing device.
- Video signals are coded in the invisible part of the frame. The code has binary format and precedes the video information. The channel number is assigned to each frame at the highest level of reliability. The coding also facilitates image processing.
- Both MOS and its review station are PC-controlled using a System Application Architecture software surface making operation very user-friendly.
- The review station is capable of channel sorting as well as detection of missing scenes.
- MOS complies with the relevant technical standards.
- MOS passed a reliability test successfully.

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EURATOM MULTI-CAMERA OPTICAL SURVEILLANCE SYSTEM (EMOSS)
A DIGITAL SOLUTION

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Abstract

In 1989 the Euratom Safeguards Directorate of the Commission of the European Communities drew up functional and draft technical specifications for a new fully digital multi-camera optical surveillance system. HYMATOM of Castries designed and built a prototype unit for laboratory and field tests. This paper reports and system design and first test results.

1. Introduction

Over the past five year there has been a marked increase in the number of video systems installed for safeguards surveillance. During that time attention was focussed mainly on improving the reliability and functionality of the video recording system, and less importance was attached to the fact that optical information stored by any safeguards video system must ultimately be reviewed by inspectors and that the reliability and ergonomy of this review procedure is crucial. An analysis of reviewing problems reported by inspectors and tests by staff from the Euratom Safeguards video group showed that the main problem was the difficulty of accessing individual scenes in single or slow-step mode without noise bars, decoding instabilities and lengthy searches involving repeated forward and rewind scans of the video tape. It was decided that this problem could best be resolved by changing to digital image storage given that most commercial video multiplexers already digitise images internally. Following requests from inspectors for video systems which allow easier review with frame-accurate access to events, and based on several years experience with different video surveillance systems, the Euratom Safeguards Directorate (DCS) drew up functional and draft technical specifications for a fully digital multi-camera video system in 1989. The French company HYMATOM, selected after a call for tender, designed and built a prototype unit which has been subjected to laboratory tests. The design includes digital image recording on erasable optical disks, three different storage modes involving change detection and a mouse-driven graphical user interface.

2. User Requirements - Functional Specifications

The user requirements were translated into functional specifications as follows. Images from one to four cameras are to be multiplexed and subsequently digitised to at most 512 by 512 pixels with up to 8 bit grey level resolution. A string of images should be buffered in video RAM, reduced and compressed and then recorded on optical disk (in digital form). The frequency at which images are recorded on the optical disk should be selectable between 10 seconds and 30 minutes for each camera. The system should give the choice between simple time lapse recording (images are written to disk at a fixed preselected frequency which may be different for each camera), and recording on change detection only (images are written to disk only if they represent a change in grey levels exceeding a preset threshold over more than a certain preset sub-area of a window within the full image. The system should recognize mains power failure and automatically switch to power-down mode where cameras are switched on in single shot mode and the disk is switched on and written to only when the video RAM is full. The image storage capacity should be sufficient for at least 25 000 images. Critical components such as the optical disk and possibly the video RAM should be duplicated for back-up in case of malfunction. The system should have a number of alarm contact and sensor switches which indicate possible tampering, opening of the cabinet door and environmental sensors (shock, temperature, humidity, etc). The date and time of sensor events should be superimposed on the recorded image together with a flag indicating the nature of the event. If the video cable is broken or the camera is changed this must also be indicated and recorded. An option for encrypted status and/or image transmission via modem and telephone lines should be included. The complete system should fit into a cabinet suitable for wall mounting or a suitcase and system weight and size should be as small as possible. The external surfaces of the housing should be tamper evident. The system should be capable of operating in a temperature range of -5°C to 45°C and a humidity range of 10%-90%.

A detailed and amended set of functional specifications based on the above draft specifications was drawn up by HYMATOM and agreed by DCS in March 1990.

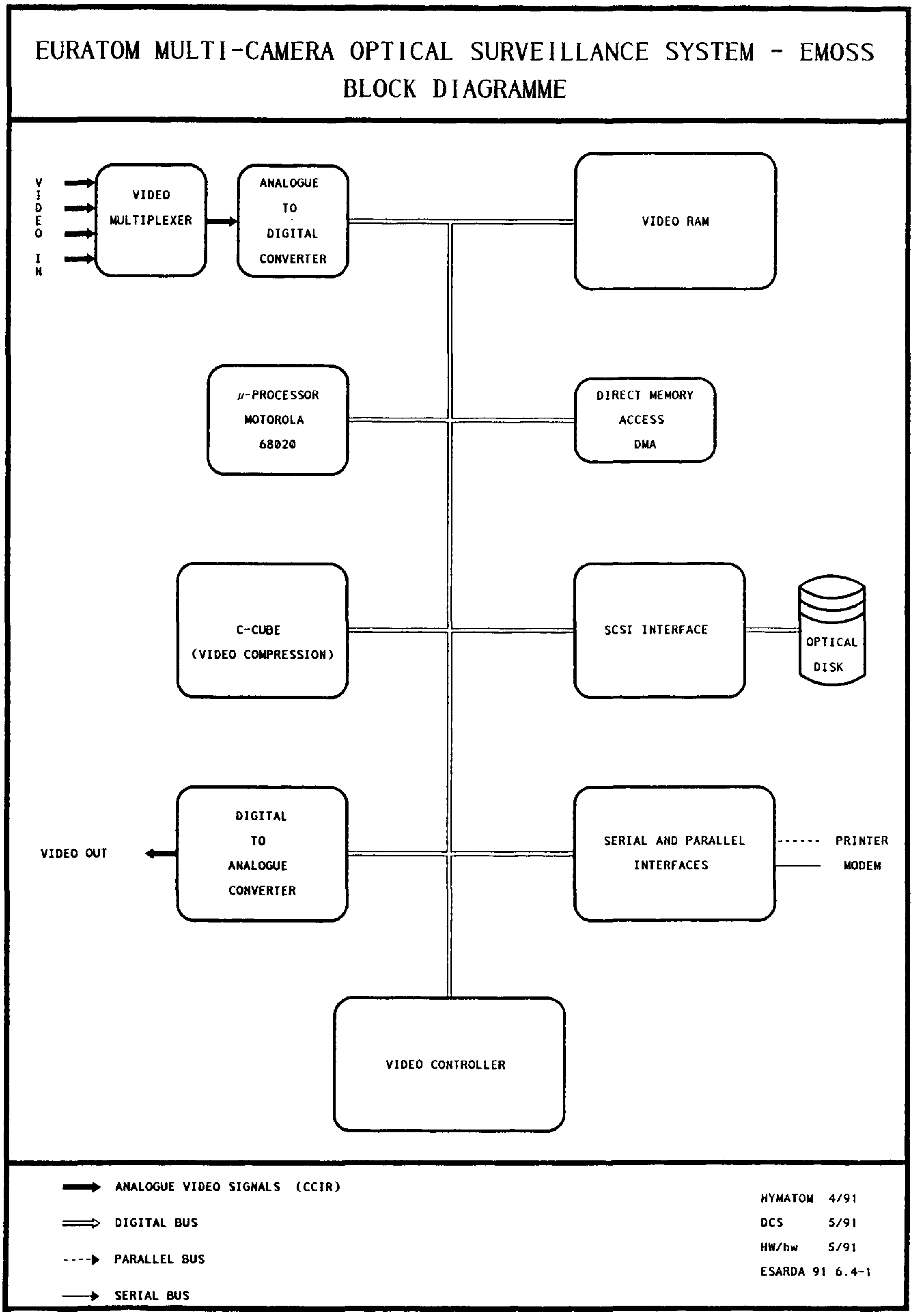

Figure 1: EMOSS Block Diagram

3. Technical Specifications

General system description: HYMATOM designed hardware and software for the EMOSS during 1990 utilising the simultaneous development of a commercial product called "MEMOBANK". The main printed circuit board is the same for both devices. Figure 1 shows a simplified block diagram of the EMOSS. The system is controlled by a Motorola 68020 microprocessor, a second processor also present (Motorola 68000) is not shown. Analogue CCIR images from up to four cameras are multiplexed, converted to digital images in a dedicated chip and stored in 1 to 8 MB Video RAM which is organised as FIFO (first in first out) cyclic storage for each camera. The amount of RAM and hence the number of images to be held in FIFO for each camera can be selected during system set-up. Images are reduced by comparing subsequent frames and calculating differences and by hardware compression in a dedicated unit known as "C Cube" which implements the latest JPEG standards. Images are sent for recording to one erasable optical disk (Sony 2x300 MB) through a SCSI interface. Thanks to the standard interface, additional or alternative storage devices can be connected. The optical disk cartridges can be removed and inserted into a separate review station yet to be built.
Access to Video RAM is through a DMA controller saving valuable processor time. Display of video images is effected through a video controller unit and they can also be output as CCIR analogue video signals through the digital to analogue convertor. In view of the price and the reliability of optical disk drives it was decided not to duplicate the disk drive and the video RAM but to install a second EMOSS in parallel where deemed necessary. A printer and a modem can be connected to the system to produce listings of tamper, environmental and change events and to transfer system status, listings and also images (encrypted) through telephone lines to an appropriate receiving station. The system also has an alarm interface for up to 8 external sensors.

Principal software functions: The main functions are:
- acquisition of video images of 1 to 4 cameras
- visualisation of images on monitor
- digitisation and storage of images
- review of (event annotated) images
- review or print-out of event lists and further processing of images

Graphical user interface: The user interacts with the system through a mouse driven graphical user interface by clicking on icons and clicking and dragging windows.

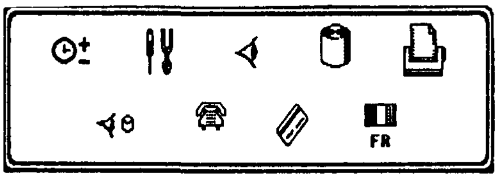

Figure 2a: Main Menu

Main menu: This menu consists of nine icons of which two are reserved for future applications (modem comms/ card access). The seven active icons correspond to the following sub-menus:
- system timer for automatic activation/de-activation of EMOSS
- system parameters; this is only accessible if an external contact is closed
- display mode; controls the way in which the camera images are displayed on the system monitor
- system activation; switches recording to disk on or off
- review submenu; gives access to images archived on disk
- language choice; currently the language of the user interface can be selected from French, English, Spanish, Italian, Dutch and German
- Print menu; starts print-out of event listings

The most important submenus are described in more detail below.

Display mode: This menu is accessible irrespective of whether the EMOSS is in recording mode or not. The submenu allows the following options each represented by an icon or a group of icons:
- choice between full screen image for the camera selected and a four-way split where images from all four cameras are displayed in the four screen quadrants
- Electronic zoom of regions of the full image or quad screen. The area to be zoomed is selected by drawing a box around it with the mouse
- Freezing of images in quad screen mode
- Non-linear grey level correction for images with too much contrast, or with strong light sources making areas of interest in the shade appear too dark to recognize details, or for objects which appear dark and featureless against bright light(s) shining directly into the camera. This is a very important feature because neither camera positions nor lighting conditions are necessarily always satisfactory for safeguards cameras. The grey level correction can either be carried out automatically by a special algorithm (not yet implemented) or manually by clicking and dragging the source grey level vs display grey level line

System timer: Storage of images to the disk can be started and stopped in three ways,
- by clicking on disk icon in main menu
- by closing an external contact
- by setting a system timer

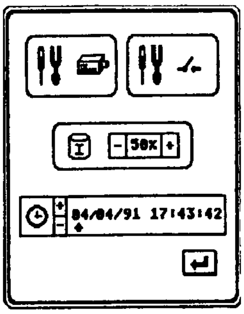

Figure 2b: Parameter Menu

System parameters: Before system set up through parameter tables can be explained it is necessary to comment on the different recording modes possible. Image storage proceeds in two steps. The images from each camera are read into a cyclic FIFO storage in Video RAM. From there they are transferred to optical disk when a trigger is received (mode 1) or when the change detector senses a relative grey level change greater than a predefined threshold in a sufficiently large sub-area of the total image or of a window (modes 2&3). After an alarm has been detected the system continues acquisition into Video RAM until the FIFO buffer for the camera for which the alarm was triggered contains 2 images before and the maximum number of images after the trigger within the size of the buffer. Then the contents of the FIFO buffer are transferred to the disk in the correct order. This is an important feature which permits the reviewer to check the events leading up to the change detection alarm.

In mode 1 storage to disk is triggered by external contacts C1 to C4 and G1 to G4. In modes 2 and 3 images are stored only if gray level changes are detected between subsequent time lapsed images as already described above. Mode 2 is normally selected for cameras trained on high activity areas while mode 3 is chosen for low activity areas. The difference between mode 2 and 3 is the way in which the event history is written to a separate event logging file on the disk.

The parameter submenu consists of four icons not counting the "return to main menu" icon which is of course available in all submenus (Figure 2).

- **camera recording parameters**, see tables 1a & 1b.
  acq(uisition) selects a camera for storage to RAM and disk
  frequency defines the frequency at which images are acquired for each camera
  grey level switches between 64 and 256 grey levels
  res(olution) toggles 256 or 512 pixel resolution
  grey (level) *change* sets threshold for relative changes in grey level between consecutive camera images in percent.
  number (of) *frames* fixes the consecutive number of frames to be held in the FIFO buffer in Video RAM
  total/partial describes whether the full image or only a window is stored
  % memory lists the proportion of total Video RAM required for the buffer storage for each camera with the parameters chosen
  store mode selects way in which images are recorded on disk as described previously.
- **alarm contact parameters** define which contacts are activated for which camera and to what part of the optical disk the corresponding event messages are written

  There are three types ot external contacts available, C1 to C4 can be activated for cameras 1 to 4 respectively, G1 and G2 can trigger all cameras but only if they appear simultaneously with the camera specific triggers C1-C4, and G3 and G4 which can trigger all cameras independent of the status of the camera specific triggers.
- **disk parameters** selects the relative distribution between the two logical disk areas
- **system time** sets system time and date

camera	acq	frequency						grey levels			res			grey change		
1	0	-	0	+	-	0	+	-	256	+	-	512	+	-	2	+
2	0	-	0	+	-	0	+	-	256	+	-	512	+	-	2	+
3	0	-	0	+	-	0	+	-	256	+	-	512	+	-	2	+
4	0	-	0	+	-	0	+	-	256	+	-	512	+	-	2	+

Table 1a: acquisition and comparison parameters

camera	number frames			total partial	window	% memory	% surface			record mode
1	-	10	+	P		4	-	30	+	1
2	-	10	+	P		1	-	6	+	2
3	-	20	+	P		1	-	10	+	3
4	-	2	+	T		33	-	1	+	1

Table 1b: storage parameters

Image review: This submenu gives access to images stored on the optical disk. The different icons (see Figure 3) represent the choice of internal or external disk drive, the disk area chosen, the camera number(s) to be reviewed, a starting date and time for the review, a routine to copy the disk files to an external disk and the actual start of the reviewing procedure after the parameters listed previously have been set. During image review a horizontal submenu bar appears at the bottom of the monitor (Figure 4) which allows forward, backward, and fast forward review of images, zoom of images and non-linear grey level correction.

Alarms: There are three types of alarms which can partially or completely stop the system if not acknowledged and corrected for. The first is a "disk full" alarm which is first raised when 90% of disk capacity of area II has been exceeded. The others are "camera masked" or "camera fault". When disk area II is completely full the system is stopped until a message icon is clicked after which new images are recorded over the first images stored on area II of the disk. Cameras for which the system detects a "camera fault" condition are ignored until a user re-activates the camera. The first time a "camera masked" condition appears for a particular camera the message is recorded in the alarm log. No further images of this camera are recorded until the "camera masked" condition is no longer valid.

Event log: Significant changes detected in grey levels of images and/or windows and camera fault messages are stored as event records in the event logging file. Alarms caused by closure of external contacts are stored as a different type of event in the same log. Events may be listed on screen or printed out. The event log review submenu allows the selection by type of event, camera number and date and time.

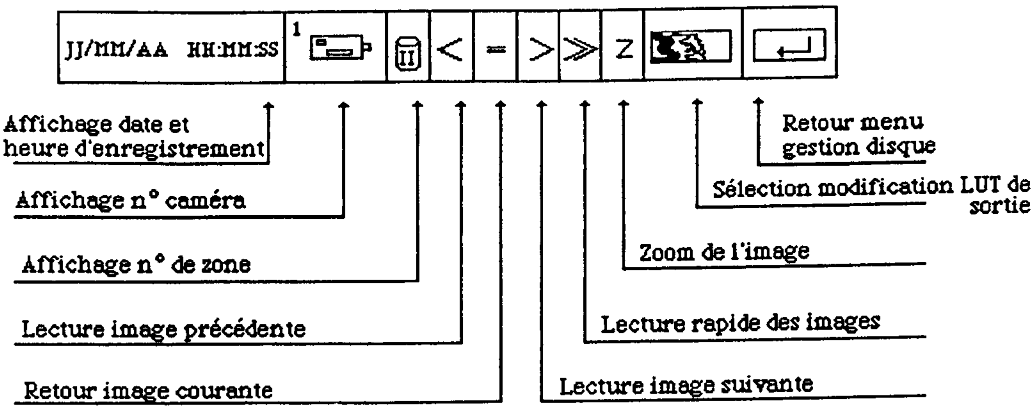

Figure 4: Review Control Menu

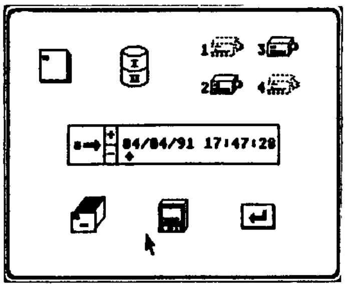

Figure 3: Image Archive Menu

4. First test results

The EMOSS prototype was delivered to DCS in December 1990. Laboratory tests started in January 1991. The test showed that the system worked as specified but that especially at the high resolution setting reduction and compression of images by software was insufficient for high activity areas. Some minor faults were identified in the system software. Since then new versions of system software and hardware were installed including the C Cube image compression board which can compress images by a factor of 20. It was found that the upgraded EMOSS stored as many as 270 000 images for low activity areas. The upgraded EMOSS will enter field testing in June 1991.

5. Further developments

Following successful conclusion of the field tests of the upgraded protoptype, three production units and a matching review unit will be used for system reliability tests. It is expected that EMOSS can enter routine inspection use in 1992. In parallel new options will be implemented such as the superposition on the images of radiation sensor data (joint project CEA-DCS-HYMATOM), alternative image compression hardware, mixed storage modes (front-end video motion detection) and larger storage media.

6. Conclusions:

Under a contract and on the basis of specifications defined by the Euratom Safeguards Directorate the first fully digital video system for safeguards applications has been designed and built by HYMATOM. The system has substantial advantages over analogue recording devices such as superior image quality, easy and fast frame accurate access to images by date and time or by event number, image related event logging and event listing, and easier post processing of images.

PROGRESS SINCE 1990 WITH THE JRC ULTRASONIC SEALING-BOLTS TECHNOLOGY

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Abstract

The last Sellafield demonstration in June 1990 on a dummy MultiElement Bottle in a non-active pond and the first two exercises of a "50 MEBs field-test" in a transfer bay , in November 1990 and April 1991, are presented.

The demonstration in the non-active pond led to the selection for implementation of the JRC Ispra MK3r Sealing Bolt, to be sealed in the New Transfer Bay and reverified later on, in THORP. The first sealing exercise was organized in November 1990, very soon after the decision to start implementing the technique. It has been possible to complete a first step in this field-test programme, using transitional tools and reading/processing equipment. Operations were globally successful, indicating progress still to be accomplished before providing site and inspectorate with reliable sets of equipment and good understanding and support from the local staff.

1. Introduction

From beginning 1986 to end 1987, a long term experiment has been carried out by JRC-Ispra in the open air storage pond B27, at the BNFL plant in Sellafield. It was aimed at the evaluation of an Ultrasonic Sealing System (derived from the VAK-III technique) for the sealing of MultiElement Bottles (MEB) transport/storage nuclear containers. This technique and its progress have been reported in several publications [1,2,3].

In 1989, a discussion took place among Euratom, BNFL, and IAEA on the potential interest of that technique and Ispra was asked to prepare for a field-test on 50 MEBs to be sealed in the New Transfer Bay, prior to shipping to THORP, and reverified some months later in THORP Inlet Bay or Main Pond. It was agreed upon to organize beforehand a "real scale" demonstration on a dummy MEB in an inactive pond.

That demonstration led to the selection by the interested parties of a "ready-to-use" MK3r Sealing-Bolt (SB) as a candidate for the 50 MEBs test. The first sealing exercise was organized in November 1990, very soon after the decision to start implementing the technique and a very short time was allowed to manufacture a first series of MK4r (modified MK3r) SBs and to provide BNFL with the documentation on their fabrication. Transitional tools and electronic equipments were used.

We report on that demonstration and on the first two exercises, in November 1990 and April 1991, during which 4, then 6, MEBs could be sealed in real conditions.

2. Inactive Pond Demonstration

This was a two-days test aimed at presenting the technique to BNFL, Euratom and IAEA representatives. In particular, to BNFL staff, it was essential to show our ability to remove normal BNFL bolts from the MEB without any mechanical problem.

An empty 1166 MEB had been prepared by BNFL in the inactive pond, for the 19th of June. Its lid was closed but 7 bolts out of 19 had been removed in advance in order to free sites for JRC Sealing-Bolts.

During the first day, a BNFL normal bolt was removed, reinstalled and retightened. A second BNFL bolt was installed in a free location as well. Then, one MK3r, one MK3 and three MK2r Sealing-Bolts were installed according to the scheme in **Table I**. The location for one MK3 (D119) appeared to be too close to a lid's rib, causing difficulty in screwing operations. Also, some resistance was found to screw in MK3 nb D151 and apply the correct torque. In order to simplify shipping, a unique series of extension tubes was sent and used to constitute: either a "Grasping Tool" used for installing or removing both BNFL and JRC bolts (without applying more than hand torque); either a "Reading Tool", with a "Reading Head", to read the integrity and the identity at once (in the case of MK3r) or in two sequences (in the cases of MK2r and MK3) but without taking the tool out of water. The third tool was the normal BNFL spanner tube which allows tightening the bolts to the nominal torque of 74 Nxm with a dynamometric wrench.

After the tool transformation, we could read and verify the installed seals with our reading equipment consisting at that time (Generation II) of a SONIC ultrasonic instrument, connected to the transducer which "reads" the seal signature, and providing an analog output to a BMC/SHARP set which A/D converts the signal and processes the data (the BMC digital voltmeter being monitored by the SHARP 1500 micro-computer). For the first time we also used a TOSHIBA 1600 laptop computer to visualize the results given by the SHARP computer and perform in parallel the correlation calculations **(fig.1)**. A portable DICONIX printer was used as well. The coefficients of correlation obtained between phase 0 (Ispra) and phase 4 (Sellafield) on SBs 112, 113, 115 and 039 are given in **Table I** under the SBs numbers.

	x	x	x	o	o	o	X	o	o	x	o	o	x
nb:				112	113	115		119	151		039	041	
corr:				.942	.976	.957		(.912)			.987		
type:				MK2r				MK3			MK3r		
use:				(ready)				(transformable)			(ready)		
trsd:				two				two			one		

TABLE I

The second day, we have tried unsuccessfully to restore our SONIC instrument after a network breakdown the previous evening. An attempt to use a PANAMETRICS instrument found in Sellafield just allowed to remeasure the MK3 119 sealing-head out of the pond. The correlation Ispra(SONIC)/Sellafield(PANAMETRICS) obtained was 0.912. We also could break and check the broken integrity of MK2r nb 115. Then we installed MK3r nb 041 after removing a BNFL bolt and we transformed the MK3 nb 151 into a seal by clamping its sealing-head. Since we could not perform a verification in the presence of observers, we agreed with them to break and remove MK3r nb 039 and to remeasure it later on in La Hague, with another set of instruments. This was done within two days and a correlation factor of 0.925 was obtained.

After the demonstration in June, the inactive pond was emptied and we could come back in July to check and remove the SBs installed previously. Confirmation was given that on this kind of MEB, some locations near the lid's ribs or weldings are improper for putting SBs. Therefore MK2r nb 112 and MK3 nb 151 were found in bad conditions. The presence of a gasket 7mm thick was confirmed as well. We also checked the depth of all threaded holes in the MEB's body. Even using handy wrenches (not exerting flexion torques like the long tube spanners) some SB was found very hard to loosen, some easy. This indicated that in case of corrosion or thread damage high torques, near to the nominal 70 Nxm, might be necessary to apply while screwing in a bolt or a SB, before it comes into contact with the lid and starts pressing it. One SB was found improperly assembled due to too narrow tolerances in that batch fabricatiion. Nevertheless the MK3r nb 41 broke properly as did the MK3r nb 39.

3. 1rst Exercise in the New Transfer Bay

Several weeks after the June demonstration, BNFL, Euratom, and IAEA confirmed the 50 MEBs field-test for which Ispra had to prepare in a very short time all the necessary equipment and the 50 SBs. It was decided to organise a first exercise in November 1990, in the New Transfer Bay (NTB) with a first batch of SBs, aimed at the sealing of 6 MEBs within a week. We had enough material to manufacture the seals and perform DA and QA tests in our laboratory. The handling and reading tools would have been same as in June, except that the aluminium multisection tube was kept for the "grasping tool" and a one-piece stainless steel tube was prepared by BNFL with Ispra end fittings in order to accomodate the Ispra reading-head.

Within November we could obtain and supply BNFL with the certification documentation on the material used in SB's manufacturing. We had a series of "new" MK4r SB manufactured, assembled and measured.

The 1rst day (13/11/90) was dedicated to the preparation of the tools and equipment and to the measurement of two SBs nb 009 and 010.

On the 2nd day (14/11/90) two SBs were installed and reverified on two 1176 MEBs brought into the NTB on a double bogy. One seal was withdrawn because it gave indication (bad correlation) of a possible incorrect mounting at the manufacturing stage. It was replaced and no other problem was encountered.

On the 3rd day (15/11/90) two other SBs were installed and reverified on two other 1176 MEBs.

The reading equipment used to perform the verification/correlation process was same as above (see **2.**) for the 2nd day (Generation II without the TOSHIBA 1600). On the 3rd day, we used for the first time an active connection between the SHARP/BMC and the TOSHIBA and a new software (SLFD 90.4 LOCAL/ REMOTE) , offering the possibility to work either from one computer or the other (Generation III). **Table II** gives the data and correlation factors pertinent to this exercise.

date inst:	14/11	14/11	15/11	15/11
MEB type:	1176	1176	1176	1176
MEB nb:	491	463	772	607
SB type:	MK4r	MK4r	MK4r	MK4r
SB nb:	008	007	009	010
corr(A*3):	.952	.967	.979	.986

trsd: L09; reading-head: RH4/AD4; 5sec/rev

TABLE II

4 2nd Exercise in the New Trasfer Bay

The second exercise in the New Transfer Bay was scheduled for week 17 (23 and 24/4/91). Between November 90 and April 91 we were able to eventually produce and acquire the signatures (phase 0 and phase 1) of 60 MK4r Sealing-Bolts. We eventually managed to have the mechanical parts **(fig.2)** manufactured in due time.

The acquisition of the signature was made with the reading ultrasonic/electronic equipment shown in **fig.3**. This time, a new ultrasonic KRAUTKRAMER instrument, fully monitored from the TOSHIBA laptop computer, has been used along with a TOSHIBA expansion box containing a new A/C converter board. In fact, for the first time since 1986, we have abandonned the use of the complex SONIC + SHARP 1500 + BMC. This new equipment (Generation IV) along with a series of SBs could be sent in due time through the Safeguards Directorate of Luxembourg.

The first day (23/04/91), we could install and reverify two MK4r SBs on two MEBs already in position, before 2 p.m.

BNFL managed to swap the two sealed MEBs with two others from the deep section in less than half an hour. We were said that these ones had their bolts already retightened beforehand, which saved time. These two MEBs were sealed and the SBs reverified by 4 p.m. See **Table III** for the data and correlation factors obtained.

date inst:	23/4	23/4	23/4	23/4	24/4	24/4
MEB type:	1176	1176	1176	1176	1176	1176
MEB nb:	531	459*	220	131	455*	395
SB type:	MK4r	MK4r	MK4r	MK4r	MK4r	MK4r
SB nb:	011	012	013	014	015	016
corr(A*0):	.968	.974	.961	.970		

trsd: L09; reading-head: RH4/AD4; 5sec/rev

(*) number readable on the MEB's lid

TABLE III

5. Discussion and Conclusions

JRC-Ispra has started studying a Sealing System for spent fuel containers like BNFL MultiElement Bottles since 1985. With the agreement of BNFL, Ispra has carried out in 1986 and 1987 a long-term experiment in the open-air storage pond of Sellafield. In several occasions SBs were installed and/or reverified on site and the results periodically reported. By the way, the seals, the handling tools and reading equipment were improved.

These results eventually brought the Inspectorates to considering this technique as a possible alternative to NDA measurements for the safeguarding of MEB spent fuel containers. In order to assess the whole system, a real scale test in actual conditions, aimed at the sealing of 50 MEBs leaving the storage pond, was contemplated in 1989 and planned to start in 1990.

We have presented the last demonstration in June, carried out in a non active bay, which has shown the feasibility of the system and in particular its practicability regarding the handling operation and the reverification of the Sealing Bolts on site. It also determined the choice of the MK3r SB as the best candidate among other types studied by Ispra.

After that demonstration the 50 MEBs test could start "as soon as possible". We have presented the two first exercises, each one consisting normally in the sealing of 6 MEBs during a two-days period.

The exercise in November was not fully satisfactory since some mechanical difficulties were encountered with two SBs. But the verification technique worked properly and we could demonstrate progress concerning the data acquisition and processing.

Next exercise was set up in April in order to let enough time to Ispra to prepare 50 SBs. Unfortunately, it was not possible to complete the exercise on 2 of the 6 MEBs scheduled for sealing. Neither was it possible to proceed, the second day, to the breakage and verification of one SB. We were happy with the safe transportation of our equipment from Ispra and could use for the first time a new set of ultrasonic/electronic instruments, on our way to simplify and automatize the verification procedure and reducing the equipment size.

BNFL indicated a fixed schedule for these exercises, planned to occur every second week, possibly within two days. Therefore we are preparing for the exercise in week nb 19, in which 6 SBs will be applied without on-site reverification. This decision was taken in order to keep with the BNFL schedule.

6. References

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[3] B.C. d'Agraives, J. Toornvliet, E. Mascetti, E. Clegg, "Second exercise of the long term experiment with Ispra Sealing-Bolts at the BNFL Sellafield facility". Proceedings of the 11th ESARDA Symposium, Luxembourg, 30 May-1 June 1989, p. 399.

Acknowledgment

We wish to thank Messrs W.T. Stanley and A. Reynolds from BNFL for their personal involvement and assistance to JRC-Ispra staff respectively during the demonstration and the two first exercises.

We also like to thank Mrs C. Andrews and the drivers from Luxembourg DG-XVII who managed to transport our material in due time.

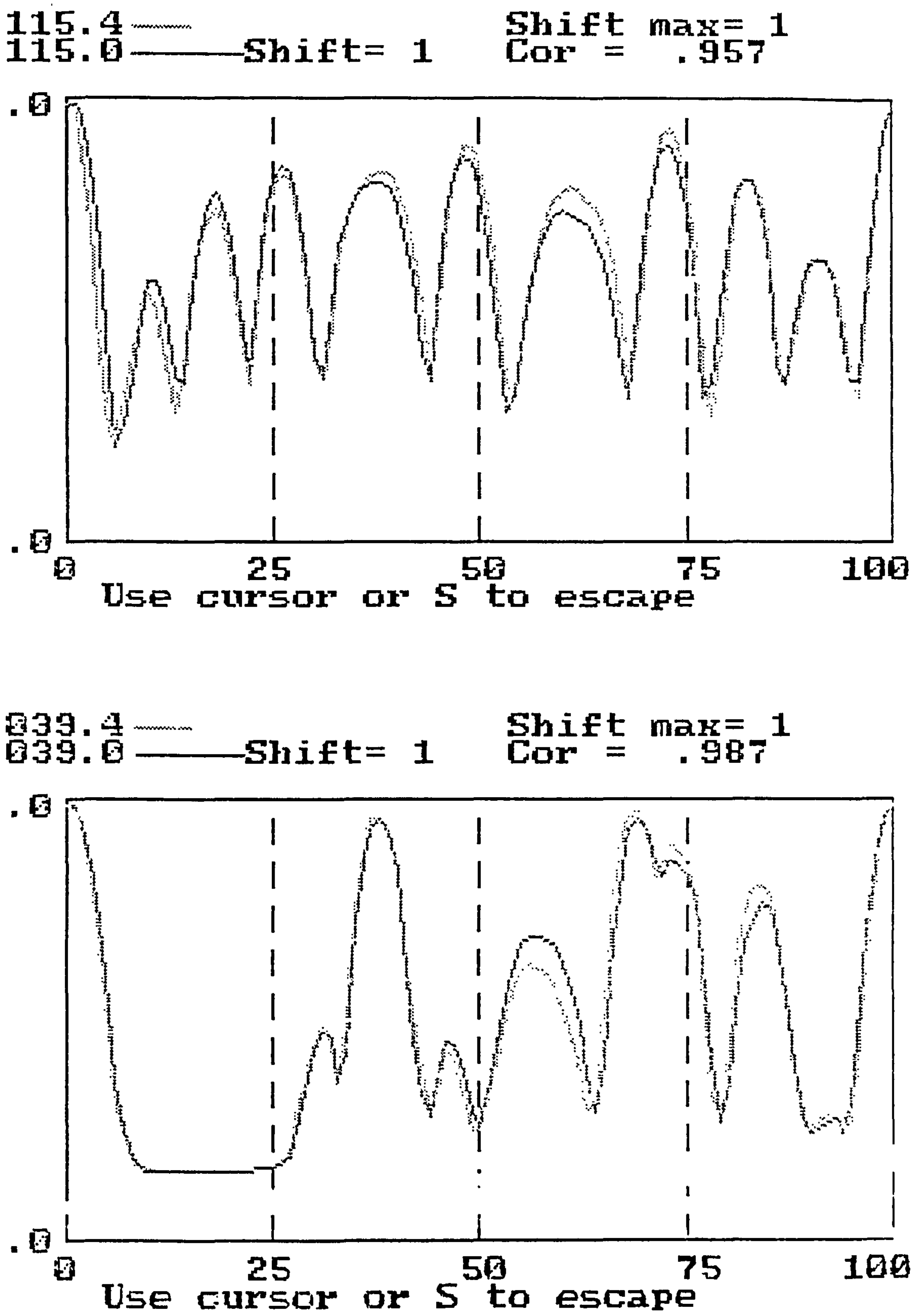

Fig.1: Signatures of SB 115 and 039 remeasured in B142
Phase 4 against Phase 0

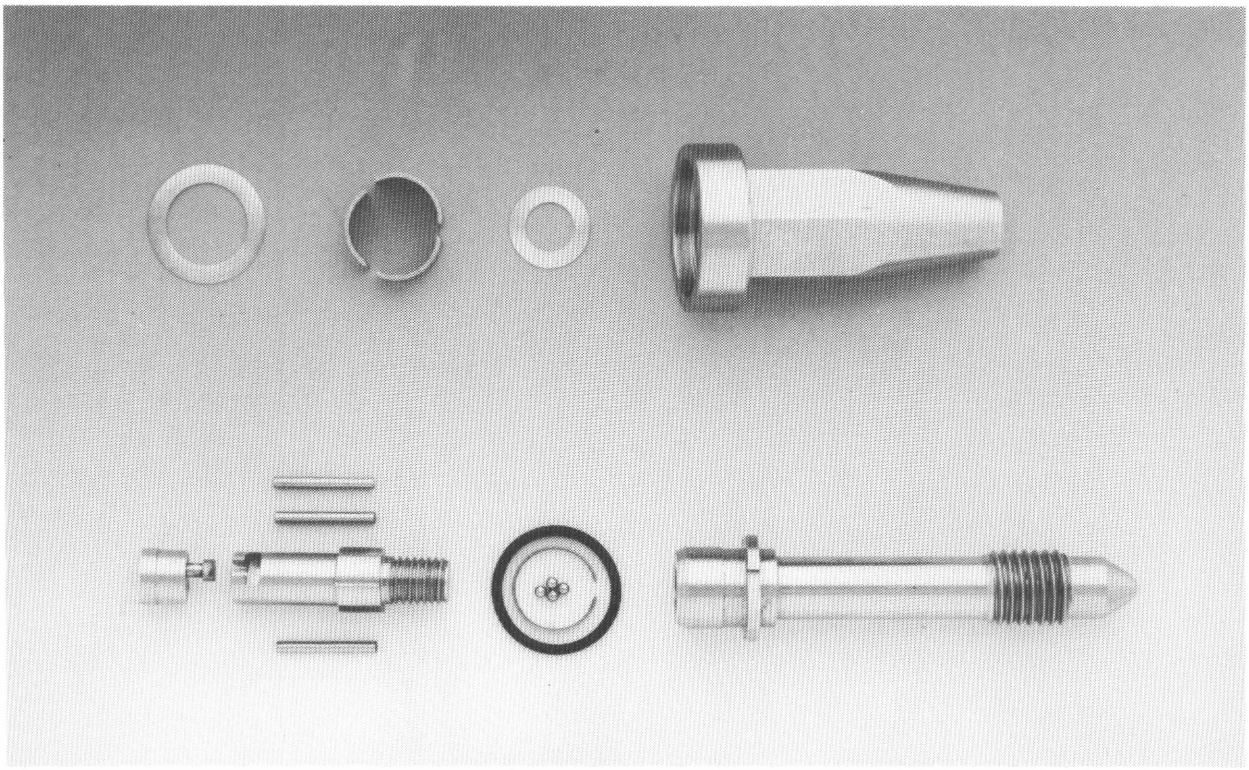

Fig.2: Mechanical parts of a Sealing Bolt MK4r

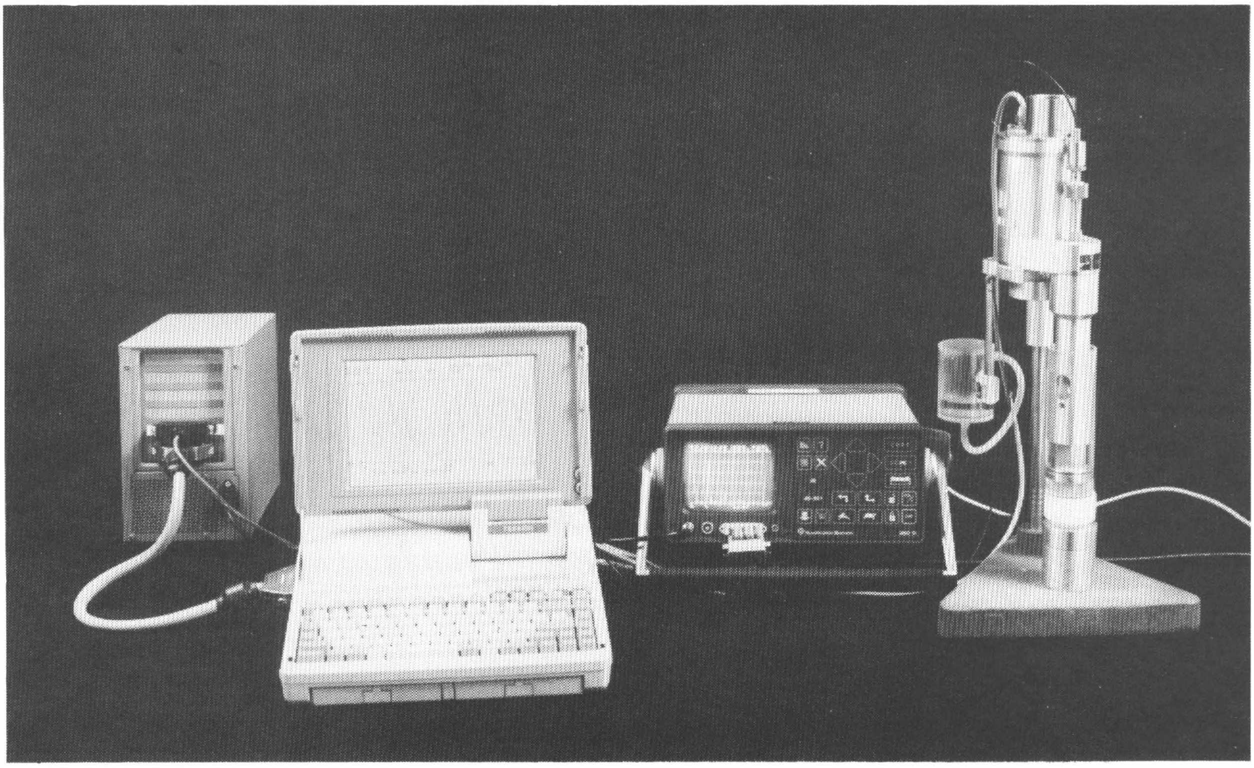

Fig.3: Set of new instruments (Generation IV)
used in the second exercise in B27 NTB

FIELD TEST OF THE TAMPER RESISTANT TV-LINK (TRTL) AT THE T0-FACILITY OF CAP DE LA HAGUE

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Abstract

Under the French Support Programme to the IAEA the containment and surveillance system for the facility complex at Cap de la Hague (CONSULHA) was developed, while under the German Support Programme a method for authentication of video signals was developed. France, Germany, and the IAEA agreed to cooperate in the performance of a field test of the Tamper Resistant TV-Link (TRTL) at the T0-facility. Euratom was invited to participate. The TRTL was connected to CONSULHA on October 13, 1989, and worked until June 13, 1990. During this period, the CONSULHA system was in tests and commissioning. Disturbances like video linearity problems, power failures, loss of video signal, interferences etc. were recorded by the TRTL. The test results of the TRTL have been used to enhance the performance of the TRTL. The paper describes the field test and results.

1. Introduction

In November 1988, representatives from France, Germany, and the International Atomic Energy Agency convened at KFA Jülich and discussed the possibilities for cooperation regarding the implementation of the Tamper Resistant TV-Link (TRTL) into the French CONSULHA containment and surveillance system. The French side was interested in proposing the utilization of the TRTL for authentication of video signals.

It was agreed to plan and perform a joint field test of the TRTL in the T0-facility at Cap de la Hague, provided that the technical demonstration of the TRTL to Euratom and IAEA at the Saclay Nuclear Research Centre in January 1989 was successful and the TRTL was acceptable to the Agency. It was envisaged to test a TRTL with two camera channels. The Agency was to provide the necessary TRTL hardware including a Multiplex TV-System (MUX) with time lapse recorders.

The IAEA representative stated that the existing TRTL was not yet a production type, in particular as regards the miniaturization of the transmitter unit. The French side supported the view that for general applications the existing transmitter unit was too big. This initiated the development of the miniaturized transmitter unit.

It was mutually agreed to install the TRTL at the T0-facility with a field test period of three months. The Agency confirmed provision of the TRTL equipment and a technician for the setup. Under the German Support Programme, the developer was prepared to travel to Cap de la Hague for the setup of the TRTL and subsequently for support during the test period. A preparatory meeting was held at KFA in June 1989 with the participation of the Euratom Safeguards Directorate to discuss the technical details regarding the installation of the TRTL. Euratom confirmed participation in the TRTL field test on the conceptual and technical levels. The participants confirmed that the planned TRTL field test had to be seen in the context of the overall safeguards system for Cap de la Hague, the TRTL being only one feature of the French CONSULHA system.

The goals of the field test were as follows:

- to prove the functional performance and reliability of the TRTL under real facility conditions;
- to collect practical experience in a plant with regard to the appropriateness of the system design;
- to enhance the system performance by introducing improvements on the basis of the test results.

From the results certain conclusions were drawn as regards the further development and practical application of the TRTL. The details of the test are given in reference /1/.

2. Organisation of the Field Test

The IAEA, Euratom, and the French and German Support Programmes shared the responsibilities for the field test in the following way:

- The French Support Programme was to provide the technical infrastructure.
- The German Support Programme was to provide the installation and maintenance of the TRTL system and the evaluation of the results.
- The IAEA was to provide the TRTL hardware including a MUX System with time lapse recorders.
- IAEA and Euratom were to provide their joint team control room.

3. Setup of the System

The TRTL developer specified the technical requirements for the field commissioning of the TRTL in the T0-facility:

1. Control room
   - Space for a 19"/36U cabinet, depth 600 mm; front and rear of the cabinet should be accessible.
   - All cables to enter at the rear or bottom of the cabinet.
   - Specification of signal requirements:
     - video signals: 1 V_{pp}/75R with BNC plug
     - Two TTL signals and ground connected to screw terminals in the cabinet. The TTL signals (for high radiation background) should be active low.
     - Power in - 220 VAC/250 W, 'euro' plug in the cabinet.
2. Access corridor of hot cell
   - Space for a 19"/3U module frame, depth 310 mm
   - The connections to be at the rear of the module frame.
   - Specification of signal requirements:
     - video signals: 1 V_{pp}/75R with BNC plug
     - One TTL signal active high for the 'door open' and ground
     - Power in - 220 VAC/40 W, 'euro' plug in the module frame.

Regarding installation and test requirements of the TRTL, the IAEA confirmed the following provisions:

1. Transmitter Unit
   - The location of installation would be inside a sealed 19" rack made available by the facility operator.
   - The operating voltage range without powering the CCD camera would be 9 - 40 VDC or VAC/approx. 4 W per channel.
   - The operating voltage range to power the CCD camera would be 15 - 40 VDC or VAC/approx. 8 W including CCD camera.

2. Receiver Unit

- The receiver unit would be plugged into the 19″ TRTL-mainframe of 6U height, which would be installed inside the tamper proof 19″-rack of the Agency-owned Multiplex TV Surveillance System (MUX).

- The 19″ TRTL-mainframe would be powered by 110 or 220 VAC, each receiver unit requiring 5 W.

3. TRTL Performance

The TRTL generated its own date and time and inserted it into the video signal (the position could be programmed). It further recorded four different types of 'events', i.e. video signal tampering (partial or total replacement of the camera video signal), any difference of time or date between the transmitter and receiver units, and abnormal operating temperatures inside the camera housing. Plain text for these events was inserted into the video signal and thus recorded on video tape. As a backup measure, the number of events and the time and date of their occurrence were also stored in a memory notebook. A PC could be used to retrieve these data from the memory notebook through a serial interface and to store them on a floppy disk for further evaluation.

With an external request from the gamma and neutron detectors installed at T0, the receiver unit also generated a user definable plain text, e.g. 'high gamma' or 'high neutron' and inserted this text into the video image. The triggering of the external devices, e.g. video recorder, was not part of the TRTL.

Figure 1 shows a schematic setup of the TRTL system, cameras, fibre optics system, and video system. The TRTL was installed as a component of the Agency-owned Multiplex TV Surveillance System (MUX) at the T0-facility at Cap de la Hague. Two cameras faced the interior of the unloading cell for spent fuel transport casks. The TRTL-transmitter units were mounted in a rack in the access corridor of the hot cell. The TRTL-receiver units were mounted in the MUX-rack on a different floor of the plant. The camera signals were processed by the TRTL-transmitter unit and then converted into optical signals. These signals passed through optical fibre cables connecting the hot cell facility with the safeguards control room which housed the recording units and the authenticating electronics.

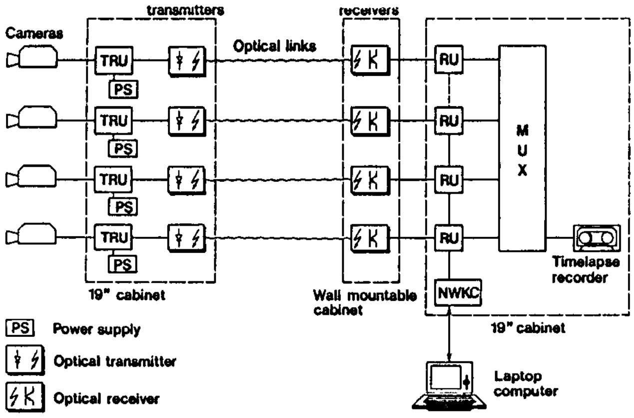

Fig. 1. Schematic Setup of a TRTL System, Cameras, Fibre Optics System, and MUX

The technical specifications of the fiber optics system were as follows:

Optical fibre	GOLD 50 125
material	glass
diameter of fibre	50/125 µm
diameter of cable	1 mm
linear attenuation	at 850 nm <3 dB/km
linear attenuation	at 1300 nm <1.8 dB/km
operating temperature	- 50, + 125 °C
tensile strength	20 K
Transmitter	ETI-09 M
transmission band	- 3dB, 30 Hz to 9 MHz
power	±12 V/30 mA
input impedance	75 Ohm
input power	1 V peak-to-peak
wave length usual	850 nm
signal/noise	45 dB
transmission power	- 22 dBm
Receiver	RTI-09 M
features	see Transmitter
reception threshold	- 32 dBm
differential gain	10%
differential phase	10 deg
Connectors	SOCAPEX 906 505 5007

The IAEA had specified a 1 % non-linearity for the fibre optic system.

4. Performance of the Field Test

The installation of the TRTL and startup of the field test took place on 12/13 October, 1989. Two camera channels were available for the test. A subsequent visit to Cap de la Hague was scheduled by the French and German Support Programmes for 7 November, 1989. At that time the MUX system at Cap de la Hague was not fully operational. It was not used for inspection purposes yet and therefore unsealed.

The Agency further stated that it had not been the intention to incorporate the TRTL into the CONSULHA system prior to completion of a successful field test and a comprehensive qualification programme. Only at a time when the TRTL had the status of 'inspection use', would the Agency discuss with Euratom whether the TRTL should be part of the mutually agreed surveillance system at Cap de la Hague.

The meeting of 7 November, 1989, was attended by representatives of the French and German Support Programmes and the Agency. It yielded the following results.

The three-month field test of the TRTL started on October 13, 1989. Two cameras in the dry cell (basket and cask) were connected to the TRTL.

These two cameras would not be used for safeguards purposes as the CONSULHA acceptance tests were scheduled for January 1990.

The French side offered to collect the data from the TRTL system during the test period of the TRTL until mid-January and send the diskettes to the developer in Germany.

The German side provided a technical note on the Tamper Resistant TV-Link /2/.

The test data proved the reliable performance of the TRTL which registered several types of event data.

One of the two cameras had to be removed for upgrading the mechanical tamper resistance. A first adjustment of the linearity of the optical link was carried out but not completed. Further work on the fibre optic cables was planned to reduce the attenuation.

The developer stayed at Cap de la Hague in February, 1990, for performance monitoring and maintenance of the TRTL. He stated that the TRTL had operated over the whole test period, showing that the video signal could not be authenticated due to non-linearity of the video signal transmission in the fibre optic link. Linearity problems with the fibre optic link were still present.

A linear dependence between the signal transmitted from the camera side and the signal received at the recorder side is an indispensable condition for successful authentication.

The TRTL had recorded the following events:

- The DATA flag had occurred because the receiver unit had not been able to authenticate the data from the transmitter unit due to the above mentioned non-linearity problems.
- There had been a camera failure of three days.
- There had been a frequent loss of video signals.

A power failure in the PC had introduced a false date/time into the TRTL.

The field test of the TRTL was completed at Cap de la Hague on 13 June, 1990.

5. Results and Consequences

The field test had been extremely valuable, as the developer had had the opportunity to become acquainted with real operating conditions in an industrial plant. The lessons learnt were:

1. Safeguards equipment should be delivered to a plant in a fully operational state, i.e. completely cabled and tested.

2. Prior to installation, the quality of the available video signal should be verified for conformity with international video standards.

In particular, the following problems were faced at La Hague:

1. Overloading caused by the video signal;
   The level of the video signal was not within the limits of international standards (white level > 0.7 V_{pp}). Transmitter Unit and Receiver Unit were affected.
2. Spikes in the video signal (Electromagnetic Interference);
   Transmitter Unit and Receiver Unit were affected.
3. Non-linearity of the transmission line;
   Receiver Unit was affected.
4. Longterm loss of the video signal due to camera servicing by the operator's subcontractor;
   Transmitter Unit and Receiver Unit were affected.
5. Practical problems with the downloading of the encryption key.

The following actions were taken as a result of the La Hague test:

1. Design of a TV-Pattern Generator (TPG) to feed standard grey areas into the video transmission line (CCIR, EIA standards). The TRTL can be used to monitor the linearity of the transmission line. For installation of the TRTL, the technician should be able to utilize the TPG.
2. Further development of the TRTL software and hardware.

The major efforts of the design enhancements were associated with the new software rather than the hardware. The hardware design was frozen, whereas the software was subjected to further improvement of the performance of the system. In the meantime, discussions on a complete optical surveillance system which included the TRTL led to the development of the Multi-Camera Optical Surveillance (MOS) System.

6. Summary

At the Cap de la Hague T0 facility the Tamper Resistant TV-Link (TRTL) was connected to the CONSULHA System on October 13, 1989, and operated until June 13, 1990. During this period, the CONSULHA System was in the commissioning and field test.

Two camera channels were available for the TRTL field test. The facility operator recorded the system event data on computer diskettes which were conveyed to the developer of the TRTL. From time to time the developer travelled to Cap de la Hague for maintenance of the TRTL and review of the data in the electronic notebook.

The TRTL operated throughout the test period. Event flags were properly registered nearly every day. The test conditions were influenced by the fact that the cameras were frequently serviced which resulted in loss of video signals. The TRTL indicated linearity problems associated with the optical links. The quality of the video signals as well as interferences also posed problems. Therefore, the question arose of how to measure the quality of a video transmission line with regard to the requirements imposed by the TRTL. During the field test, it was not always possible to reliably verify the authenticity of the signals. The developer tried to cope with the non-linearity of the fibre optic transmission. The threshold was set from 5% to 15%. But still, the TRTL gave tamper flags. This indicated that the non-linearity exceeded 15%, which was confirmed in a non-linearity test (result was approx. 25%).

The video information was recorded on the MUX System. The video tape and the records of February to June 1990 were given to the Agency for evaluation in order to make a correlation between the data stored in the TRTL notebook and on video tape.

The test results of the TRTL have been used to enhance its performance.

The TRTL authentication system is now a major component of the Multi-Camera Optical Surveillance System (MOS) developed under the German Support Programme to the IAEA. Many of the practical problems faced in the TRTL field test at Cap de la Hague were taken into account. MOS measures and reports on all analog values associated with the transmission line. A commercial pattern generator can be used to assess the quality of the transmission line. Another feature introduced into the authentication method is the securing and reporting in case of power interruptions. The system will be much more tolerant with regard to video interference.

7. References

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/2/ G. Neumann, B. Richter; Brief Technical Notes on the Tamper Resistant TV-Link (TRTL), BMFT/IAEA Joint Programme, report no. 194, Jülich, December 1989.

VERIFICATION OF THE BURN-UP OF SPENT FUEL ASSEMBLIES BY MEANS OF THE CONSULHA CONTAINMENT/SURVEILLANCE SYSTEM

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Abstract

CONSULHA is a containment/surveillance system which has been developed as part of the French Support Programme for the IAEA Safeguards in cooperation with EURATOM and was designed to meet the IAEA EURATOM requirements for the verification of nuclear materials. This system will make it possible to count movements and verify irradiation of spent fuel assemblies in industrial facilitities such as reprocessing plants and nuclear reactors.

1. Principles and Particularities of CONSULHA

CONSULHA is a system of original design associating video surveillance with the measuring of neutron and gamma activity levels. This technique makes it possible to ascertain that a fuel assembly is visible, and enables it to be tagged to prove that it actually is a spent fuel assembly./1/ /2/

2. Results Obtained in Service

CONSULHA was installed at the COGEMA plant at La Hague in September 1989. During the 1989-1990 campaigns, it enabled inspection of the movement of 1962 PWR and BWR fuel assemblies unloaded in workshop T0. In this unloading station, it was necessary to adapt the system to the existing installation. Despite this constraint, it was confirmed that it was possible to verify the burn up of the fuel assemblies. The semi-quantitative results obtained make it possible to propose a burn up verification system.

The operating software was created in collaboration with the IAEA and EURATOM Inspectors, on the basis of an actual verification situation. The inspectors normally have video recordings obtained with cameras which are sequentially saved on a magnetic tape, nuclear readings and data which are supplied by the operating organization and which have to be checked.

Using the readings obtained with the fuel assemblies unloaded at T0, the relationship between the burn up announced by the operating organization and the results of neutron and gamma activity measurement were determined. A first series of curves, without correction for cooling time, made it possible to verify the burn up with an accuracy of the order of 20% despite the different cooling times. If the raw values are corrected to allow for the cooling time, determined by only gross gamma activity measurements, accuracy is improved.

3. Criticism of Results

The results obtained with CONSULHA installed at T0 were used in the knowledge that they would not be obtained under the best conditions, for the following reasons:

- It was impossible to optimize the positioning of the detectors due to operating constraints.
- This resulted in low neutron detection effectiveness and poor statistical accuracy.
- The distance between the fuel assemblies and the detectors is not constant because of the different positions of the assemblies in the storage basket.
- Gamma activity measurement was carried out using a detector measuring total gamma radiation.

4. Conclusion

Verification of the burn up of spent fuel assemblies involves problems at a number of levels where there is a place for cheap, unsophisticated equipment easy to use. It is for this reason that an improvement programme has been set up in order to simplify the equipment, particularly the detectors.

In veiw of the particularly encouraging results obtained at T0, it can be considered that the basic industrial elements are already available for creating a burn up verification device.

Certain improvements were envisaged from the start of the project:

- optimizing detector installation,
- improving gamma activity detector performance,
- adding gamma activity detector performance,
- adding software to utilize the results for safeguards purposes,
- associating the results with video review.

These four points will be considered in detail during this presentation in the light of the work in progress.

5. Present Approach

In association with the IAEA and EURATOM safeguards authorities, French and German universities and laboratories (the PHASE Laboratory of INP2 Strasbourg and the Laboratory of the Univerisity of Dresden), and industrial partners (Intertechnique, Quartz et Silice and Dialectica), three techniques have been tested:

- measurement of burn up using neutron detectors and gamma detectors with energy selection,
- measurement of burn up using a single neutron and gamma activity detector by means of a single detector using a doped scintillation glass enabling neutron/gamma discrimination by energy selection,
- verification of burn up by measurement of the photoelectric peaks of the gamma radiation of caesium-134 and caesium-137 using a CdTe detector operating at room temperature.

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VIDEO TECHNICAL CHARACTERISTICS AND RECOMMENDATIONS FOR OPTICAL SURVEILLANCE

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Abstract

The application of new video surveillance electronics to safeguards has introduced an urgent need to formulate and adopt video standards that will ensure the highest possible video quality and the orderly introduction of data insertion. Standards will provide guidance in the application of image processing and digital techniques.

Realistic and practical standards are a benefit to the IAEA, Member States, Support Programme equipment developers and facility operators, as they assist in the efficient utilization of available resources. Moreover, standards shall provide a clear path for orderly introduction of newer technologies, whilst ensuring authentication and verification of the original image through the video process.

Standards emerging from IAEA are an outcome of experience based on current knowledge, both within the safeguards arena and the video parent industry which comprises commercial and professional television. This paper provides a brief synopsis of recent developments which have highlighted the need for a surveillance based video standard together with a brief outline of these standards.

1. Introduction

In recent years, optical surveillance has become increasingly important as a means of maintaining nuclear material accountancy by the IAEA. Initially, image surveillance was accomplished with photographic film cameras. However, recent difficulties in film procurement, together with an upsurge in technical developments in television and a general reduction in the relative costs of video components, have culminated in making today's video technology indispensable to the Agency.

The electronic processing of images is very different to that employed in the older mechanical and chemical systems associated with film technology. Therefore, it is reasonable to assume that procedures and strategies adopted for the newer technology will also be different. This is especially evident with the amalgamation of analogue and digital electronics for processing images where these techniques are now commonplace. Video technology, of a kind which is unique to the safeguards arena, is being developed by the IAEA and Member States through Support Programme activities. Therefore, equipment must satisfy the requirement of the users, in this case the Inspectorate. Features must be provided which will aid the inspector's work efficiency and maintain the Agency's credibility.

Through the development activities of member states assistance programmes, the Agency has begun to evaluate and introduce new technology into optical surveillance safeguards equipment with the dual aims of credibility and efficiency.

It is from this background that a need for optical surveillance video standards has now emerged. This need has been recognized for some time within the Development Division of the Department of Safeguards of the IAEA. In May 1990, the Australian Safeguards Assistance Programme provided a Cost Free Consultant to the Agency for the specific six month task of establishing optical surveillance videc standards and recommendations for implementation in safeguards.

2. The Need for Standards

Standards guide governments, agencies, manufacturers, consumers and the general public alike and facilitate the orderly conduct of these enterprises. A common direction or purpose for each endeavour is thus assured. In consequence, video standards are of benefit to the Agency, Member States, independent equipment developers and facility operators as they assist in the utilization of the available resources by unifying equipment and procedures that aid installation and maintenance as well as lower costs.

A standard does not preclude anyone working outside its jurisdiction or bounds. However, its very existence requires that one be aware of it together with its limits and scope, and possibly more importantly, be cognizant of the reasons for its existence or development. In many instances, standards are mandatory where information is interchanged or hardware merged, Similarly, this will be the case for IAEA safeguards equipment. Of course, standards are subject to periodic review as technology or procedures change and are revised or withdrawn.

Optical surveillance, like any other branch of engineering, requires standards to be a comprehensive and established activity. Until recently, it had not been necessary for IAEA development to apply any special video standards, primarily because the application of video technology to the safeguards area was comparatively small compared to the total television industry. Moreover, application of television systems was considered a fairly straight forward procedure.

This philosophy has now changed. It has been realized that there is a necessity for orderly and efficient introduction of standards into optical surveillance framework.

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The first aim is the formulation and adoption of basic video standards that will ensure the highest possible video quality at an acceptable level of equipment cost. The maintenance of better video quality means consistent, high quality surveillance data for the Inspectorate.

The second aim is the orderly introduction of electronic data insertion into the video signal. There are considerable potential benefits for the Agency to fully utilize electronic data insertion. It would be possible to electronically append the following information to the video signal:

- Scene time and date;
- Facility sensor/camera location
- Authentication data
- Temperature/humidity data
- Tamper indicators
- Other data from containment and surveillance devices

Once appended, the data is recorded with the video for subsequent retrieval during the review process. Because the data is in an electronic form, it can be easily read by a computer system. This would greatly facilitate all future review systems which use computer techniques or image processing.

Until now, no unified television standards existed within the safeguards community; nor do they appear to be in common use throughout the surveillance community in general. Furthermore, standards in the broader television community cannot be directly applied to IAEA optical surveillance without modification, as they are not directly applicable; nor do they reflect the developments or changes that are being tailored to the specific requirements of video quality and data insertion for safeguards applications.

Standards need to flow on to ensure the orderly introduction of these newer technologies whilst ensuring authentication and verification of the original image through the video process. The planned use of back-end and front-end image processing techniques and the incorporation of sophisticated authentication systems have applied additional motivation for the immediate establishment of video standards specifically devised for the IAEA's optical surveillance systems but based, where possible, on standards already in use by the television industry. Such a procedure will minimize re-engineering.

Wherever possible, the proposed IAEA standards shall recommend the use of current national and international standards as its basis. This procedure was adopted to ensure a consistency and to maximize compatibility with locally available hardware. By being compatible, the Agency's equipment will be automatically compliant with much of the commercially available video equipment. This methodology is essential if the manufacture of specialized items of plant is to be minimized.

3. The Problems of Multiple Television Systems

There are many different television systems operating in world. As safeguards is an international and cooperative effort, careful attention must be paid to ways of ameliorating the effects of this non-standardization in any system developed for optical surveillance. To do this we must examine the existing systems so that we can extract the maximum compatibility for the proposed standards.

The International Radio Consultative Committee (CCIR), has considered some of the different television systems in Report 624-3 /1/. The systems in more common use are considered in the following table:

Table 1 Television systems in common usage

SYSTEM	LINES/ FRAME	FIELD FREQ (Hz)	LINE FREQ (Hz)	BW (MHz)
M	525	60 (59 94)	15,734.264 +/- 0 0003%	4.2
N	625	50	15,625 +/- 0 00014%	4.2
B,G	625	50	15,625 +/- 0 0001%	5 0
H	625	50	15,625 +/- 0 0001%	5.0
I	625	50	15,625 +/- 0 00002%	5 5
D,K	625	50	15,625 +/- 0.0001%	6 0
K1	625	50	15,625 +/- 0 0001%	6 0
L	625	50	15,625 +/- 0.0001%	6.0
Rec. 472	625	50	15,625 +/- 0 0001%	5 0/ 5 5/ 6 0

NB' BW = Bandwidth
Figures based on CCIR data

Needless to say, the establishment of video standards for the Agency's optical surveillance will require close liaison between developers and users, similar to the consultative process adopted by the CCIR, the International Electrical Commission (IEC) and other standards setting organizations.

Existing safeguards surveillance systems use many devices whose systems and electrical standards vary widely at present. In general terms, performance of these devices can be classified into the distinct categories shown in Table 2.

Although a survey has not been undertaken of inspectorate establishments worldwide, there is solid evidence to assume that classes of equipment range from high quality industrial through to domestic grade. In practice, much of the present Agency surveillance equipment is classifiable between industrial and commercial CCTV grade.

A requirement of the Video Standards Task is to adopt, as far as possible, a small but appropriate number of standards. These standards shall have as many compatible threads running through them and each being applicable to that part of the world where the Agency wishes to implement safeguards. The aim is to permit the various parts of a surveillance system,

Table 2 Categories of performance

A	*Full broadcast quality* - Used in television broadcast stations
B	*Industrial* - High quality, high resolution - Medium to high fidelity - Resolution: ≥ 400 lines, S/N 28 - 40 dB
C	*Commercial* - Used for domestic and property surveillance - Medium fidelity - Resolution: 240 - 400 lines, S/N 28 - 40 dB
D	*Domestic* - VHS, Beta and home video cameras - Low fidelity - Resolution: < 300 lines, S/N 28 - 40 dB

developed by different organizations, to be interconnected with a minimum of incompatibilities. A unified set of video standards will also help to coordinate and arbitrate on the introduction of new data insertion methods and video authentication.

To this end, it is proposed that two major strands be adopted: namely, one based on the American NTSC 525 line, 60 Hz standard for use in countries where 60 Hz mains is used for power and lighting, and the European PAL 625 line, 50 Hz system in countries where 50 Hz predominates. Both of these general standards will require adaption or modification for use in safeguards work as there are features that are now required by safeguards video that simply don't exist in the normal television arena.

It is recognized that due to the specific requirements of IAEA surveillance, some IAEA video equipment is breaking new ground, thus generating an urgent need for a standard approach. This approach is not exactly the same as the broadcast, industrial or domestic television industries. Furthermore, it must be appreciated that image processing is one of the most rapidly developing areas of surveillance video and therefore must be given considerable room to develop.

4. What Needs Standardizing?

Any optical surveillance standard adopted by the Agency must be consistent with, and an integral part of, the methodologies used by the IAEA for safeguards work. In addition, the standard must maintain or enhance confidence factors for safeguards surveillance. However, it should not be tighter than is necessary if it is to be readily accepted and implemented. It is understood that any standard has to be realistic to be workable. Adherence to it by all parties involved should present as few problems as possible. At the same time, it must be able to evolve as technology evolves and as techniques improve. It must also minimize installation expenditure, equipment replacement, maintenance and staff retraining.

It is envisaged that the IAEA video standards and recommendations shall be an "umbrella" document and combine IAEA specific methodologies with the relevant IEC and CCIR or, where necessary, other references /2/. By way of a defined methodology, it shall provide for revisions so that when any change occurs in the "reference standard", applicable revisions to the IAEA document shall take place.

For the time being, the proposed mandatory standards will be limited to that section of the video chain, loosely referred to as the *transport*, which refers to the synchronizing or timing, amplitude, linearity and transmission bandwidth parameters. Fortunately, this is the kernel of any television system and therefore the most relevant part. A reasonable approach is to breakdown the standard into its constituent parts appropriate to that required for optical surveillance. A proposed system is shown in Table 3.

The standard has been sectioned into major and minor categories. Whilst this method might not be ideal, it delineates the sections that have to be defined in the first iteration of the standard. It is anticipated that the minor items will be expanded as time permits.

Table 3 Video standard - Sectional parts

MAJOR SECTIONS:		
1.	*System*	* PAL * NTSC * Other (MAC, HDTV?)
2.	*Transport*	* Synchronizing pulses * Synchronizing timing * Amplitude and linearity * System bandwidth * Colorimetry (PAL & NTSC systems) * Linear line and terminal equipment
3.	*Type of performance*	* Quality (broadcast, industrial, etc.)
MINOR SECTIONS:		
4.	*Peripheral equipment performance*	* Cameras * VCRs * Digital storage systems * etc.
5.	*Processing*	* Image processing * Non-linear correction * Special effects (matting, etc.) * Mixing (linear and non-linear)
6.	*Test Procedures*	* Test and measurement methods * Calibration

Major subsets within the standard will be applicable to a specific environment or country, or type of standard (low resolution, colour, PAL, NTSC, etc.) and,

Minor subsets will define the component sections of such a standard (for example camera video and video authentication links may have more than one set of definitions).

Whilst there is a diversity of opinion as to the required fidelity for optical surveillance, it is widely agreed that the industrial grade, defined in Table 2, will provide a level of performance acceptable for the majority of surveillance work. Overall broad performance, from the camera lens and imaging device, through to the display device viewed by the inspector, can also be defined in terms of resolution and signal to noise ratio.

Minor items (listed by subsystem) need to be considered in the next and subsequent iterations of the standard. However, Table 4 is a brief list of some items which will be mentioned in the forthcoming Standards and Recommendations document /2/.

Table 4 Peripheral equipment performance

- Facility lighting
- Field of view
- Camera filter
- Camera lens
- Camera (monochrome and colour)
- Video cable (baseband use)
- Fibre optic (transmitter, receiver and cable)
- Video authentication link
- Video distribution amplifier
- Clamping (processing) amplifier
- Video tape recorder (12.65mm Beta)
- Video tape recorder (12.65mm VHS)
- Video tape recorder (12.65mm S-VHS)
- Video tape recorder (12.65mm VHS time lapse)
- Video tape recorder (8mm)
- Video optical (laser) disk recorder (digital)
- Digital video image processing and storage
- Electronic data insertion
- Testing methods
- Frequency of tests
- Calibration Equipment (Laboratory)

5. Vertical Interval Planning

Broadcast services use the blanking interval for many purposes: vertical interval test signals, Teletex, cuing and other timing processes. Optical surveillance also uses this area for data and authentication signals with the result that the vertical interval blanking region has become so congested that some of the active video, or picture region, is used for switching by multiplexer and VCRs.

Much consideration is being given to how best to balance the requirements of existing industrial video systems with those required for surveillance data and authentication. However, resolution of the differences in vertical interval use by optical surveillance systems, such as MIVS /3/, COSMOS /4/, MUX-II /5/, TRTL /6/, MOS /7/ and other image processing systems /8,9/ remains to be fully resolved.

6. Evaluation of the Video Standard

Following Agency in-house review, evaluation procedures shall be adopted which will seek comment from Member State experts. Where necessary, the standard shall be revised before becoming an active document. Further iterations of the standard will be made as circumstances warrant.

7. Immediate and Interim Video Standards

It is important to note that newer technologies must not be introduced for their own sake, but rather for the practical reasons such as the limitations of existing equipment. Many other examples can be cited, such as the increasing amount of surveillance material for review, limited inspection resources, inefficient review techniques and other ergonomic and economic reasons.

What can be done immediately? Options have to be available, prior to the standards being introduced, to continue to operate already installed systems, which may be outside the standard, until they can be totally replaced by new generation equipment falling within the standards. Such actions are considered necessary for practical reasons such as equipment budget forecasts and planning. The proposed standards will undoubtedly increase pressure to upgrade some surveillance systems. However, buying equipment that is obviously outside the standard should be avoided.

8. Conclusion

Many aspects which affect the setting of the Video Surveillance Standards and Recommendations are already under consideration by the Agency. A key issue is the introduction of video authentication systems. The successful introduction of optical surveillance data reduction systems will depend on both data insertion and video quality aspects. Avoiding delays in implementation will result in appropriate equipment specifications for the Agency's present and future optical surveillance systems.

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EURATOM EXPERIENCE WITH VIDEO SURVEILLANCE I

SINGLE CAMERA AND OTHER NON-MULTIPLEXED SYSTEMS

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Abstract

The Euratom Safeguards Directorate (ESD) has been using a number of single camera video systems (Ministar, MIVS, DCS) and non-multiplexed multi-camera systems (Digiquad) for routine safeguards surveillance applications during the last four years. This paper describes aspects of system design and considerations relevant for installation. It reports on system reliability and performance and presents suggestions on future improvements.

1. Introduction

In nuclear power stations and nuclear reprocessing plants, surveillance units equipped with a twin MINOLTA camera unit using Kodak MFX film are used to maintain the continuity of knowledge of material which has been independently verified at some previous point in time. These surveillance units are periodically exchanged and brought back to LUXEMBOURG for processing and subsequent film review.

The MFX Super 8 Kodak film is no longer commercially available and therefore the Minolta camera units need to be replaced by other surveillance systems based on video technology within the next few years.

The use of MOX fuel elements in LWR reactors has led to additional needs for video surveillance.

2. Equipment

Twin Minolta unit

The present EURATOM film camera surveillance system consists of a camera, timer, tamper-resistant sealable enclosure and mounting brackets that allow the proper positioning of the cameras. In practice each enclosure contains two cameras each with their own power supply and timer, viewing and same area. Two cameras are used for increased system reliability.

Each camera is triggered by individual external timers that activate the cameras to expose one frame of film on a preselected interval.

The cameras, and timing units are placed in an environmentally tight enclosure containing dessicant to control the humidity in the interior of the enclosure. The cameras are modified MINOLTA model XL-401 or XL-601 Super 8 cine camera. The interval electronics have been modified so that single frame operation is the fixed mode of use and it is initiated only by an external timer.

EURATOM has approximately 12 years of experience in the use of film camera surveillance. The first units were deployed in 1979. The original film cartridge (Kodak MFX) has a capacity of 7200 frames.

Ministar

The MINISTAR (MINI Surveillance Television And Recording) system was designed by SANDIA National Laboratories, Albuquerque, New Mexico, USA for applications requiring a small economical and portable video surveillance system.

The MINISTAR consists of three separate sub-systems :

- a camera housing containing a solid state camera with lens
- a Video Recording Assembly
- an external battery charger/accessories case.

The three-component system allows for a distributed installation. All three components may be located separately from each other.

The Video Recording Assembly (VRA) contains a SONY SL-2000 video recorder, a Control Electronics Module (CEM) and 12V features. The VRA also incorporates several tamper protection features. Power interruptions, loss of video signal and cable disconnection are detected and recorded. A tamper switch activates each time the case is opened, triggering a recording with inserted numbers in the video. The system can operate up to 4 hours on batteries during mains power failures.

Modular integrated video system (MIVS)

The Modular Integrated Video System (MIVS) was developed by Sandia National Laboratories in Albuquerque, New Mexico, USA under the U.S. support programme to the IAEA. It was developed to produce a simple, highly reliable closed circuit television (CCTV) system to replace the ageing XL-401 Twin Minolta Film Camera System. It was to replace the Minoltas in facilities where main plant power was readily available and in situations where it was desirable to have the CCTV camera separated from the CCTV recording console.

The specifications for the MIVS were approved in 1986; the operational modules were developed in 1987; and the field evaluation of the system began in 1988.

The MIVS design specifications required it to have a simple user interface, be easy to maintain, have high tamper resistance, have minimum facility impact, and achieve the same level of reliability as the Minolta for long-term, unattended surveillance. The system reliability of the Minolta is 0.98 with a 90 percent confidence level while recording 7 200 scenes over a 90 day inspection interval. In contrast, the MIVS has the capacity to record 26 000 scenes. This greater number allows the interval between scenes to be reduced and thus increases the surveillance coverage. The MIVS uses two video tape recorders (VTRs) operating in parallel.

The MIVS consists of small, unobtrusive housings, has very low power consumption, is simple to install, and in general has a very low facility impact. The equipment consists of modules plugged into a base plate, thereby facilitating maintenance in the field. Field maintenance involves simply replacing one of the four basic modules and returning the used module to the central maintenance facility for repair and reconditioning. No special tools are required for the field maintenance. It is expected that the recorder modules will be replaced on a routine basis, perhaps about once every two years or after approximately 180 000 scene recordings.

The MIVS user interface has been "human engineered" to ensure that it is as simple, understandable, and fault-proof as possible. The human interface involves pressing one of four "soft keys" at the command of the menu on an 8-line by 40-column screen. Even if surveillance settings are not complete, when the case is closed the MIVS will commence surveillance in the "forced start" mode.

Finally, a significant amount of attention has been given to making the MIVS tamper resistant. A unique and sophisticated line authentication system is used to verify the integrity of the cable between the camera and the recorder. The MIVS case itself is constructed of anodized aluminium that would be difficult to damage and repair without being detected. Seal wire guides are installed to assist in sealing the MIVS cover, and the mating surface is constructed to prevent entry without opening the case.

When a mains power failure occurs the MIVS will continue to operate on back-up battery power for approximately 3 hours (at 5 minute scene intervals).

Digiquad-System

The EURATOM video surveillance system DIGIQUAD is a modular video system based on commercially available components, mounted in a 19 inch cabinet.

Figure 1 shows a schematic overview over the system. The DIGIQUAD-system consists of 4 CCD-cameras, 2 monitors, 2 GYYR-time lapse recorders in parallel, one recorder being used for back-up, 1 DIGIQUAD processing unit, 1 power supply with battery back-up.

The two outputs from the DIGIQUAD unit, output 1 for single pictures and output 2 for the quad pictures (the images of up to four cameras are combined into one image in a four-way split screen) are recorded on video tapes. The recording interval of the time lapse recorder can be set between 2.5 min and 0.02 sec. between fields or groups of fields.

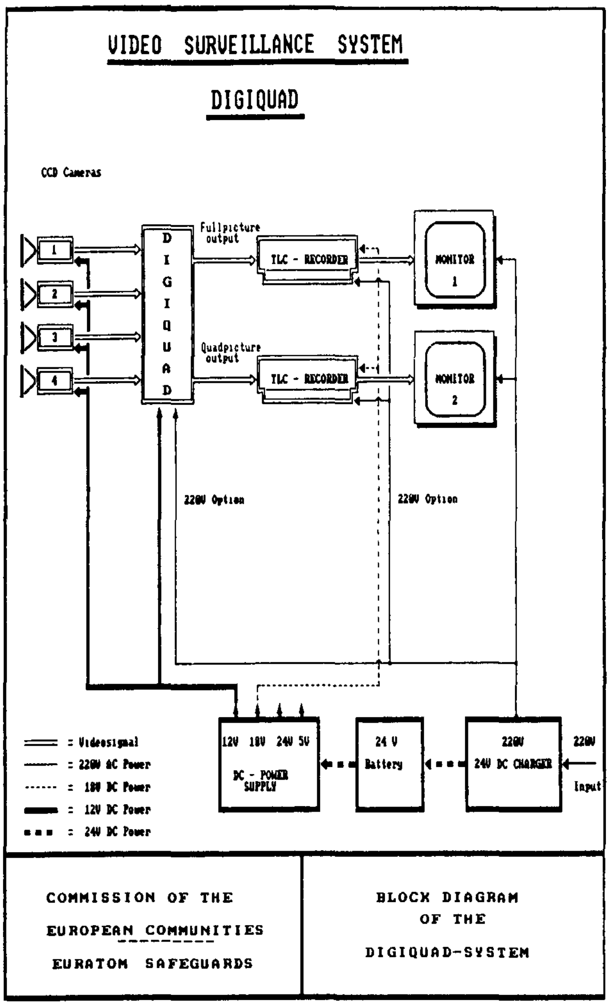

Figure 1

The power supply has a back-up battery for approximately 12 hours back-up time.
A closed dual circuit cooling system has been installed on the rear door to ensure sufficient cooling while avoiding the intake of air into the cabinet.

3. Operational experience

General

The traditionally used twin Minolta units are very tamper resistant but the limited frame capacity and radiation level restricts their application. This situation is clearly improved by today's availability of CCTV systems.

The advantages of the video systems over the old twin Minolta units are :

- the quality of the frames which give much better detail with the CCD cameras
- the transport of the tapes for the video system is easier than transporting the suitcase containing a twin Minolta unit (approximately 20 Kg)
- no time spent for preparation and opening of the units before installation and after removal because only two small Video cassettes are exchanged
- no time spent for the processing of the MFX films of the twin Minolta camera
- possibility of on-site reviewing with video systems.
- the separation of camera and system unit means that the inspector does not have to access cameras in difficult locations.

On the other hand, all video systems currently used require mains power and will only operate between 2 hours (Ministar), 4 hours (MIVS) and 12 hours (Digiquad) on back-up batteries. The installation of video systems involves not only the fixing of a camera in a sealed camera housing but also the laying of cables in cable trays, possibly through wall penetrations to a conveniently placed central system cabinet which may take the form of a suitcase, a wall-mounted case or a 19" industry standard instrumentation rack. The facility must supply mains power preferably from a non-intreruptible power supply. If possible an alarm signal should be relayed to the control room if a loss of mains power is detected on the line supplying the video system. The cable connections into the system cabinet must be protected against accidental or deliberate disconnection.

As mains-powered equipment, video systems must comply with national regulations on electrical safety.

Reliability

Table 1 shows 1990 reliability figures for the four single camera and non-multiplexed multi-camera systems in use during that year. It lists the number of systems installed in the field, the recording capacity and the total number of operating hours (approximate figures) for each type of device. The reliability figures are also shown in Figure 2. The reliability parameters were calculated as the relative number of surveillance failures caused by intrinsic system faults. All intrinsic failures observed for the Minolta systems were caused by simultaneous failures of both cameras (Table 2). The Ministar failures were brought on by malfunction of the single recorder, both intrinsic MIVS failures were caused by faults in the central module.

Of the four surveillance failures observed for Digiquad systems during 1990, three were triggered by power failures followed by a malfunction of the DC power back-up system. The fourth loss of surveillance was very probably caused by camera failure.

Review

New review tables for Minolta films and dedicated review stations are used for review. Even though in-field review is technically possible with video systems, it is the policy of the Euratom Safeguards Directorate that all video cassettes should come back to headquarters for safeguards review or at least for random technical reviews.

The review of a Minolta film typically takes one hour. Reviewing video tapes from systems set at the same recording interval as a Minolta system will normally take longer because several fields are recorded per scene whereas only one image is taken per scene by the Minolta camera.

This can be partially alleviated by reviewing the video cassette at higher tape speed. The quad picture recording of the Digiquad systems allows fast simultaneous review of four cameras. Review speeds have to be reduced, however, or recorded sequences have to be reviewed several times if there are relevant activities visible on more than one of the four pictures of the split screen.

Review of a Digiquad tape takes between ½ hour (static situation) to 6 hours (dynamic situation e.g. loading of LWR MOX).

Figure 3 shows the 1990 reviewing effort.

Table 1

EQUIPMENT DATA FOR 1990						
Type	Number systems	Type capacity	Total of operat hours	First used	Current status	Reliability 1990
Twin Minolta	86	7200 frames	775 440	1979	End 1996	99.2
Ministar	7	21000 scenes	21 600	1984	End 1992	93.4
MIVS	4	26000 scenes	34 944	1989	25 to be installed	97.5
DIGIQUAD	10	270000 frames	87 360	1989	version 2 installed	98.5

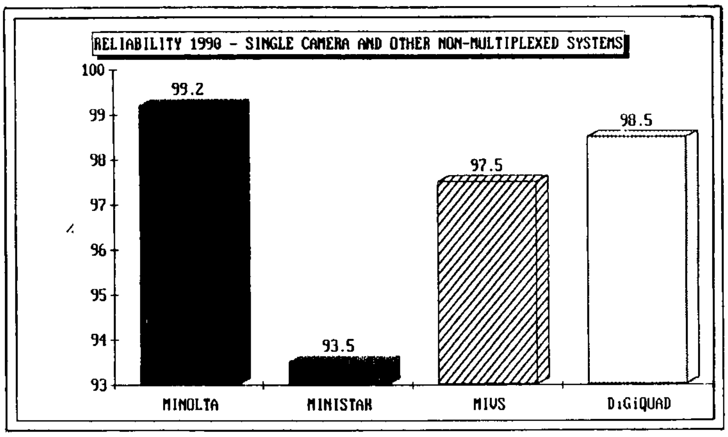

Figure 2

Table 2

FAILURE TABLE FOR EURATOM EQUIPMENT FOR 1990								
	External				Internal			
Type	Power loss	Lighting loss	Irradi failure	human error	camera failure	module failure	total	number reviews
Twin Minolta	0	9	1	2	3	0	3	360
Ministar	0	0	0	1	0	2	2	30
MIVS	0	0	0	0	0	2	2	80
DIGIQUAD	3	0	0	0	1	3	4	230

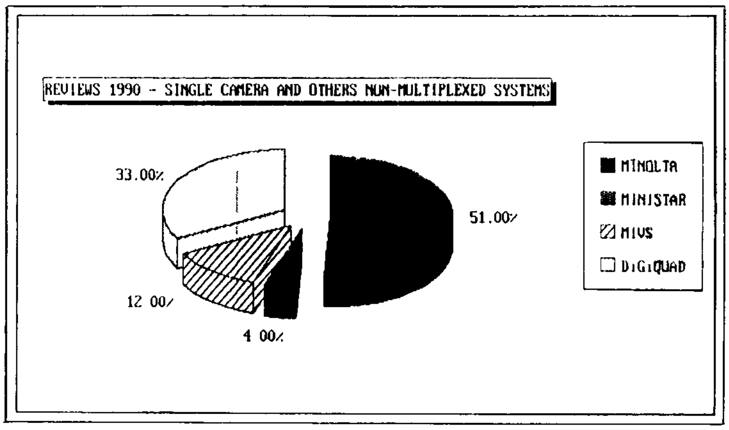

Figure 3

4. Further Developments

A new version (version 2.0) of the Digiquad system has been developed in which the problems with the DC power back-up were eliminated and a much higher degree of redundancy was built in. First tests at headquarters have demonstrated more than 17 hours of battery operation and two such systems have been installed in the field. In a further upgrade step all Digiquad systems will be fitted with special data loggers (Rustrack Rangers) which will record key system parameters such as mains voltage, current load, DC voltages, temperature, shock and humidity in battery buffered RAM. Rather like a flight recorder this device will normally only be read out if a system crash has occured.

A new automatic PC-based review station for MIVS tapes has been developed by Aquila and will be delivered soon. Another review station based on neural network technology is being tested by the IAEA and Euratom. This station was developed by Sandia and the University of New Mexico.

After it has been "trained" this station can automatically recognize items of safeguards interest. For both stations the idea is to image-process tapes in unattended mode and restrict human review to those images where the review system has detected the presence of particular items or the movement of sufficiently large items.

Following suggestions from Aquila, discussions have started on a stepwise upgrade of the MIVS system to a digital video system.

The transfer of video system status through telephone modems is currently being tested at headquarters in Luxembourg.

5. Conclusions

Over the last five years the Euratom Safeguards Directorate has tested, installed and put into routine use a number of single camera video systems and non-multiplexed multi-cameras video systems. The reliability of these systems has not yet reached that of the twin-Minolta units but improvements in system redundancy and module reliability should make it possible to attain this goal during 1991/1992 at least for the most recent devices. The main reasons for surveillance failures during 1990 were lighting and power failures followed by module failures. Safeguard video systems must be shown to comply with regulations on mechanical and electrical safety. Mains power from a non-interruptible supply must be assured. It is expected that some form of automated review or pre-review will be introduced on a test basis during 1991/1992.

NAVIGATION OF A TELE-OPERATED VEHICLE IN A SAFEGUARDS ENVIRONMENT

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Abstract

A tele-operated system is being developed to reduce the amount of human intervention in a nuclear material pilot storage facility. The system consist of two major components: a remotely guided vehicle carrying a manipulating arm, and a central graphical workstation to be used as the system's operator console. This paper describes the work that has already been done in terms of navigation of the vehicle. Special attention was given to the interface between the operator and the computer. It provides in each moment all the information needed, such that the operator may consider himself inside the storage area. Range information and TV images will be used for navigation, accurate position determination, and to authenticate the vehicle's location. Safety considerations that are part of the design, are also discussed.

1. Introduction

Developments in computing and electronic technologies have widened the options on how to design fissile material storage facilities. Storage access, inventory and surveillance philosophy, must be reassessed taking into account new available technologies.

The SAOV project (Advanced Storage of Valuable Objects) /1,2/ aims to build a pilot storage facility for nuclear materials, integrating a remotely operated manipulating system, with the following operational objectives:

- remote inventory
- remote handling and transport
- high security
- provide adequate data for high level material management

An overall view of the project has been presented elsewhere /3/. This paper details some aspects related to the navigation of the vehicle, namely, the man-machine interface, safety and security. Before actually proceeding to the topic, there is a short section describing the system's architecture and the equipment available.

2. System's Architecture

The system is divided in two major blocks (Figure 1): the remotely operated vehicle carrying on board a manipulating arm and associated equipment, and the system's operator console. This division emphasizes the design of a proper man-machine interface, granting the operator access to all the necessary information to guide and control the vehicle and equipment.

The remote handling system is composed of the following functional modules:

* System's Operator Console:
  - Colour Graphical Workstation (Unix, NFS, X11, Motif);

* Vehicle:
  - 2 motorized wheels, 68020 VME computer, optical encoders;

* Manipulating Arm:
  - 6 degrees of freedom, 68020 VME computer, optical encoders;

* Vision System:
  - TV camera, computer controlled pan-and-tilt, zoom and focus, image frame grabber;

* Range System:
  - Laser range finder with RS232 interface, pan-and-tilt;

* Ultrasonic Obstacle Detection System
  - (under consideration);

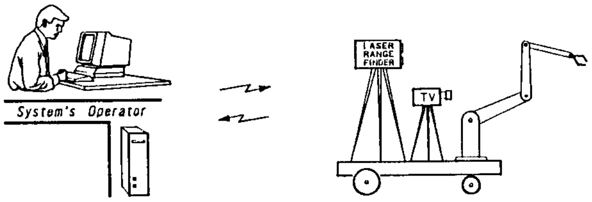

Figure 1: Computer Controlled Remote Handling.

The architecture of the system is represented in Figure 2. Commands are transmitted via an RS232C serial link to the computer on-board of the vehicle. This computer sorts out the commands and redirects them to the proper destination, i.e., to one of the real-time tasks that control vehicle's movements (speed and position), or to a second computer controlling the manipulator arm. Other tasks are associated to two pan-and-tilt units (TV camera and laser range finder), to focusing and zooming, etc...

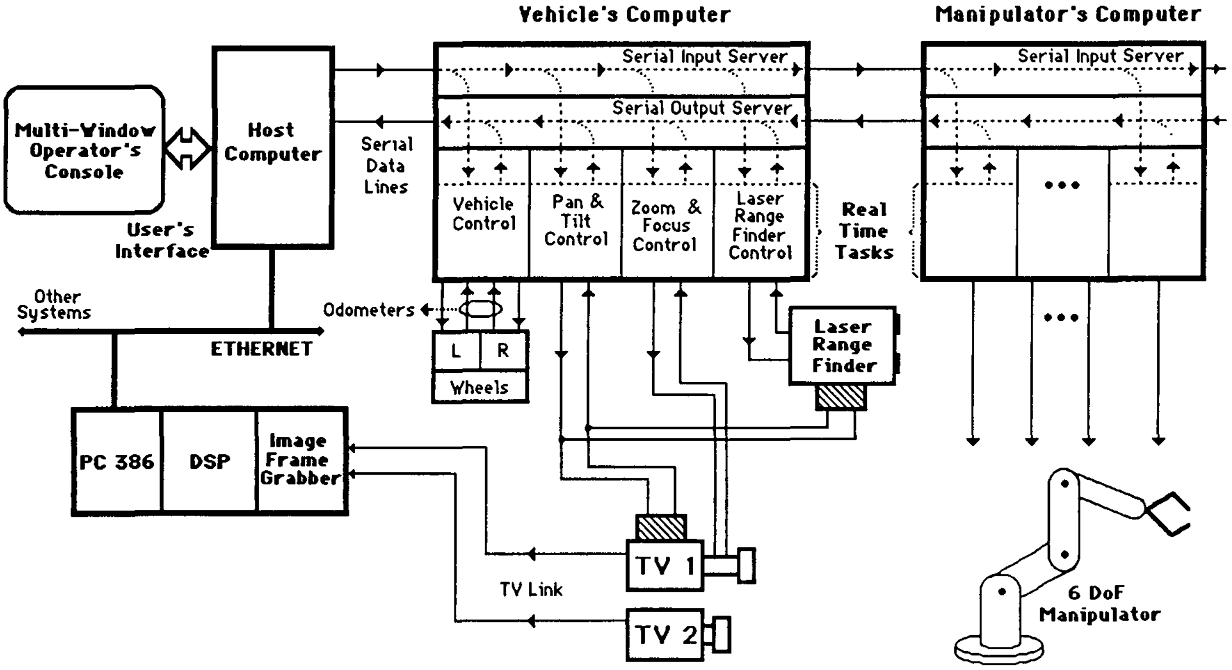

Figure 2: System's Architecture.

3. Navigation

Three navigation development phases have been devised. These phases correspond to increasing levels of complexity, each one being based on the developments of its predecessor.

I - **Manual**: the system's operator controls the movements of the vehicle directly, i.e., he/she interactively instructs the computer which commands are to be sent to the vehicle.

II - **Semi-Automated**: the operator sets some intermediate objectives to the computer, e.g., "Go to the end of the corridor". The host computer will then instruct the vehicle which actions to take and which sensorial feedback is needed.

III - **Automatic**: this mode will be objective driven in essence. The operator sets one objective, such as, "Go to locker G3". This mode includes a strategy planning module as well as a navigation one.

Navigation of a remotely operated vehicle must be seen in a broad sense. In fact, it consists of all those low level modules that are necessary for vehicle motion control (e.g., Move or Stop), and a set of middle and high level tools to help both the operator when guiding the vehicle, and the system when operating in a semi or fully automated mode. Trajectory planning and path finding, will not be considered in this paper.

3.1 Positional Feedback

Some considerations on the vehicle, positional feedback and equipment available, are necessary before proceeding into the description of the navigation tools.

The vehicle has two independently motor driven wheels (at the back) and two free wheels (at front) and uses differential steering. The on board computer with a real-time operating system, manages communications, and controls vehicle's movements, i.e., its speed and position. Motion feedback is achieved by means of two odometers, each one equipped with an incremental optical encoder (accuracy of 1 mm). Information from the odometers is read by a multi axis controller board. The DC wheel motors are driven by pulse width modulation (PWM) servo amplifiers.

A major difficulty with this type of positional feedback exists when, by any reason, the wheels slip or lose contact with the ground. This happens on irregular surfaces, or when the vehicle is subject to sudden movements. The information read from the odometers no longer corresponds to reality, and other sources of positional information must be found.

It is possible to say that within this application, the environment is well structured. Indeed, room dimensions, as well as object (e.g., file cabinets or boxes) dimensions and localizations can be well known in advance. It is thus possible to have a 3D volumetric representation (3D model) of the room and its contents. The laser range finder (LRF) is expected to play a major role in position calibration. Since the LRF provides direct distance measurement, it is possible to make a 360 degrees horizontal range scan (Figure 3). From the analysis of these data, precise position calibration (±25 mm for a range of 20 m) may be obtained. This is sufficient for navigation purposes.

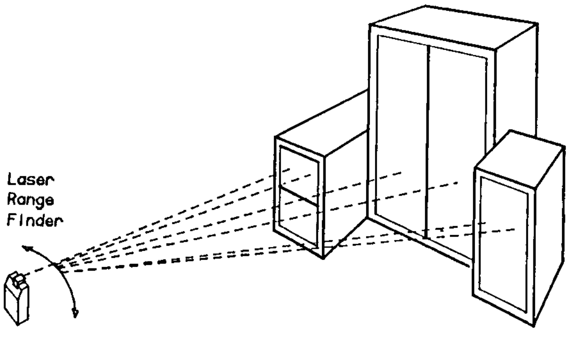

Figure 3: Scanning with a Laser Range Finder.

3.2 Navigation Tools

Different types of navigation tools are envisaged. First, one must consider the man-machine interface used to control the vehicle. Next, are those graphical tools that help the system's operator to manoeuvre the vehicle (and manipulator) and provide a feeling of being inside the storage area (tele-presence), e.g., plan maps, or 3D representations of the environment. Finally, come those tools (Watch-dogs) that assist the operator in avoiding or detecting emergency situations, such as collision detection. Each type of tools is detailed below. Tools for Safeguard purposes and safety considerations are referred separately.

4. User Interface Modules

The interface between the system's operator and the computer is paramount to the success of the project. Ease of use, security and safety are also dependent on the quality of the interface. Indeed, all commands to the remote system (vehicle and manipulator) are issued via the computer. In the same line, it is up to the computer to display all the information acquired by the on-board sensors, e.g., TV images, range information, etc..

It is our goal to integrate all the information that is relevant to the operator in a single multi window screen. In order not to become overloaded, it is up to the operator to select at any moment the information that is going to be displayed on the screen. Some moments later, other information may be selected, and the previous one iconized.

4.1 Vehicle Steering and Manoeuvring

At present, the mouse is used as an interactive device, and three vehicle control interfaces have been developed. All modes have an on-line help facility.

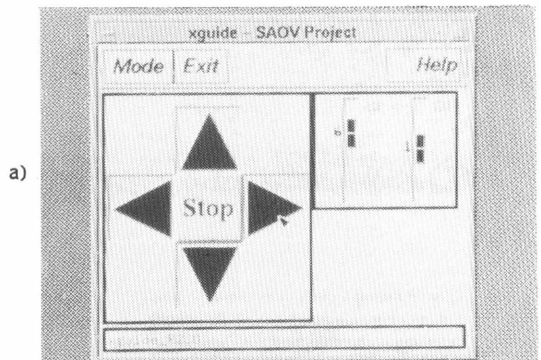

Figure 4: Vehicle Control Interface:
a) Arrow Mode; b) Sense Mode.

* **Arrow mode**: Four motion arrows (North, South, East and West) and a Stop area are displayed, as indicated in Figure 4a. It is up to the operator to click onto those arrows. The number of clicks and the arrows pressed are input to a finite state machine that will set the speed for each of the wheels. Two speed indicators display the speed on each wheel.

* **Sense Mode**: This mode emulates digitally a "joy-stick". The neighbourhood of the central STOP button is "paved" with a set of small buttons, as seen in Figure 4b. To each button corresponds a combination of speeds in the two wheels. This combination is then output to the vehicle, determining its speed and direction.

* **Drag mode**: This mode uses the three mouse keys to control both the vehicle speed and direction. The central key must be always pressed to enable motion, whereas the two lateral keys are associated to direction changes. The vehicle starts moving forward by sliding the mouse forward, and backwards when sliding the mouse backwards. Speed and acceleration can be controlled by interpreting in software the movements of the mouse.

From our experience, it can be said that the Sense mode is better suited for manoeuvring, whereas the two other modes are more indicated to straight motion. The Drag mode has the advantage that the user can control the vehicle without moving his/her eyes out of the room window.

In a near future, the use of a six degree of freedom space ball will be investigated for vehicle control.

4.2 Navigation Tools

Two types of tools are considered: Visual and Graphical. Visual tools display the images acquired by the TV cameras on board of the vehicle. Two cameras are planned. The first one, situated on the rear of the vehicle has a wide angle lens. The second camera will be installed on a computer controlled pan and tilt unit, and has a zoom lens for detailed perception. Both zoom and focus can be adjusted by the computer.

At present, a PC386 computer acquires images from the two cameras and sends the image data via the Ethernet network to the operator's console. It is expected that in a near future hardware to display live video on the workstation console becomes available.

An important tool is the environment plan viewer. The plan displays the room, its objects and the position of the vehicle. The vehicle control program sends to this program a constant data stream specifying the vehicle's location and orientation. The plan viewer accepts this data and updates the vehicle's position. This provides the user with the necessary visual feedback (Figure 5).

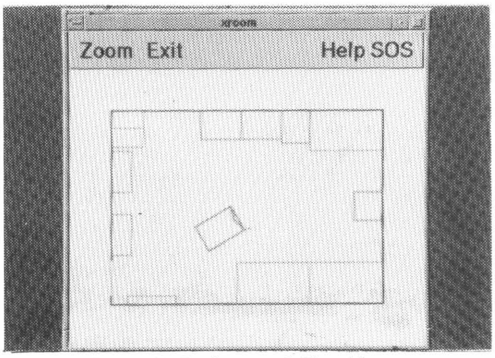

Figure 5: Environment Plan Viewer.

Another tool that will prove to be important for navigation purposes, as well as for protection (see below) is a 3 dimensional graphical (volumetric) representation of the environment, including the vehicle. Since the viewpoint is selectable, it will be possible for the operator to display a perspective of the environment as seen from the vehicle, or of the vehicle as seen from an imaginary point (e.g., ahead of the vehicle and looking backwards), as indicated in Figure 6. This tool uses the PHIGS library to represent 3D graphical information /4,5/.

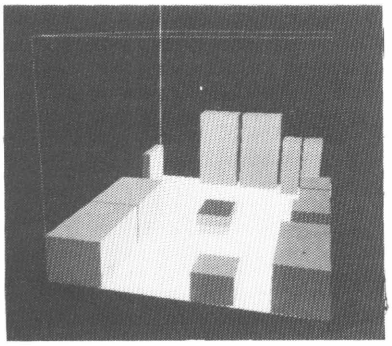

Figure 6: Environment 3D Viewer.

4.3 Computer Aided Protection

A major concern in tele-operation is the safety of the vehicle and instrumentation. It is important to provide the means for their protection. A major cause for accidents stands from collisions, involving either the vehicle or the manipulator. Considering that the volumetric representations of the environment, vehicle and manipulator are known, it is possible to extrapolate current movements, predict possible collisions, and alert the operator. This is mostly important when displacing the manipulator. When moving the vehicle, it is possible to display distances to surrounding walls. This will be useful information for the operator.

5. Safeguard Implications

Safeguard philosophy relies strongly on data verification and authentication. This is normally achieved by means of feedback, and by cross certifying data originated with different sources. One of the objectives of this project is to test the feasibility of computer aided tele-operation techniques for operations such as storage, retrieval and inventory of fissile materials. Safeguards considerations have influenced some design issues, namely the possibility for environment validation, and some aspects concerning security and safety.

5.1 Environment Validation

A major application of the laser range finder is the calibration of the vehicle's position. It is possible, however, to use the LRF to build range images. These images are acquired by scanning the LRF inside a solid angle, which will determine the image spatial resolution. Each pixel in a range image represents the distance from the LRF to an object. Objects in a range image can be identified by means of their surface edges, which correspond to discontinuities in the distance from the object to the LRF. From a technical point of view, there are indications that for these particular applications, edge detection in range images is more reliable and direct than with tonal (grey level) images. Some preliminary results are rather encouraging in what concerns image segmentation, edge detection and surface extraction /5/. Figure 7 shows a synthetic range image, to which noise has been added, $\sigma=1.5$, and the processed contours.

It is then possible to try to match the range image from the environment, with its 3D model. Indeed, if the computer "knows" where the vehicle stands, it can create a synthetic range image of the environment as perceived by the laser range finder. This image will be used as a reference when comparing with the actual range image acquired with the LRF. A match between the two images will corroborate the vehicle's position. This facility may prove to be of interest for validation purposes.

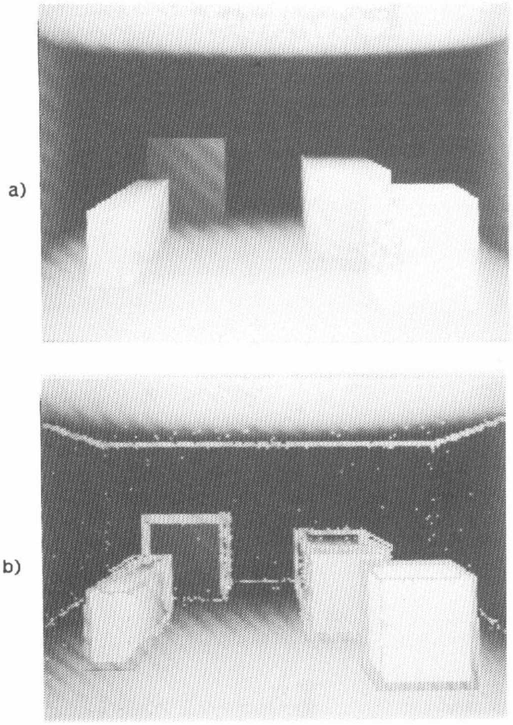

Figure 7: Range Images: a) Synthetic Range Image; b) Processed Contours.

5.2 Security and Safety

As mentioned above, mechanisms must exist to assure the vehicle's integrity. The existence of tools helping the operator can improve his/her ability driving the vehicle and manipulator. However, the design of the system must incorporate features enhancing the overall security and safety. The following topics have been considered:

* **Communications Loss**: Background tasks must exist both in the host and in the vehicle, to monitor the communications between the vehicle and the host. If there is a communication loss (e.g., due to cable breakdown), the vehicle must stop immediately its own activities and enter a standby state.

* **Vehicle Self-Parking Capability**: The vehicle must always keep track of its own moves, in order to be able to return by its own means, i.e., without host instructions and assistance, to a predefined parking place. This capability should be triggered whenever an irreversible problem is detected, e.g., loss of communications.

*** Emergency Commands Bypass**: One of the tasks of the computer in the vehicle is to manage communications and redirect commands issued by the host to some of the running tasks. Part of this function must take into account security features, namely the existence of emergency commands. These commands must bypass all the command queues, in order that their execution start as soon as possible. This concern applies both to the input command server in the vehicle, and to the host command dispatcher.

*** Ultra-Sound Obstacle Detection**: It is possible to install a belt of ultra-sound sensors around the vehicle to detect possible collisions. A real-time task should analyse ultra-sound data and detect objects that are in the way of the vehicle. Though ultra sound sensors do not provide information with good spatial resolution, their usage is important for the simple task of obstacle detection.

6. Final Considerations

Some of the work described is still in progress. There is still a lot of research and development to be made before some conclusive ideas can be extracted. Therefore, it is difficult at this stage to have precise conclusions, and to know on the impact on Safeguards. Furthermore, since the project aims to investigate new technologies and concepts in a particular area of application, feedback from the final users will be of utmost importance. Some basic ideas seemed to have been well accepted and successful on its implementation, namely, the case for an integrated presentation of relevant data into a single screen as shown in Figure 8.

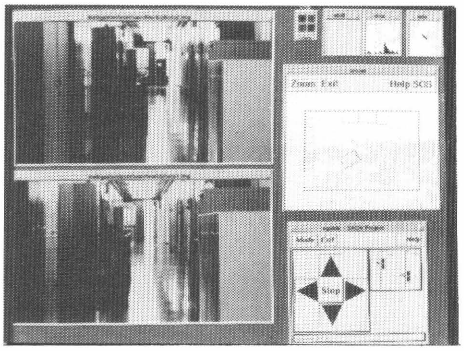

Figure 8: Integrated View of Relevant Information: TV Images, Plan View, Vehicle Control, Clock, etc...

In case of a combined use of such system by an operator and Safeguards authorities, the laser range finder with the associated software assures the necessary authentication of the vehicle's position.

In spite of the early stages of the project, some findings can already be mentioned, such as:

* It is important that at the end of the manipulator, there is a telescopic section, in order to reach the deeper parts of a locker, or file cabinet;

* For the same reasons, there is a need for the manipulating arm to have an elevating section, probably near its base, to reach upper shelves.

* In terms of the vehicle, there is the need to synchronize navigation commands with the vehicle's response. At the moment this is done in a slow manner. This problem is caused by the existing command interpreter in the vehicle, which should be considerably improved, in order to stand to Safeguards requirements.

* There is also the need to develop a specific multi-task manager to control the tasks assigned to the on-board computers (see Figure 2).

* The presentation of direct live video into the workstation screen, rather than transmitting video from a PC, would certainly improve the visual feedback to the system's operator, and hence, the ease of navigation.

7. Acknowledgment

The authors wish to express their gratitude to Messrs. M.Cuypers and D.Landat for all the fruitful discussions that helped to define some system characteristics and the operational test procedures.

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LA FLUXMETRIE THERMIQUE AU SERVICE DE LA GESTION EN TEMPS QUASI-REEL DES STOCKAGES INDUSTRIELS DE PLUTONIUM

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M.Neuilly (CEA/IPSN)

Abstract

This paper presents Thermal Fluxmetry as an aid to Near Real Time Monitoring of Plutonium storages.The device used (multi task in real time),

- collects continuously data with disk saving during aquisition and control,
- compares the consistency of the Thermal Flux measured on each container with the one calculated from the data given by the inventory book,
- displays quantitativily the result of the above comparison in the barchart mode,
- on request, displays the past thermal evolution of one or several containers.

This system was tested in a real plutonium storage at the Cadarache center.

I.- Introduction

Un stockage est constitué d'un ensemble d'emplacements dans lesquels se trouvent des emballages de transport de plutonium ou un ensemble d'alvéoles verticales ou horizontales dans lesquelles se trouvent des emballages de plutonium agréés pour le stockage.

Il existe deux types de stockage:

* les stockages où la matière arrive à l'issue de son élaboration puis est expédiée dans un autre établissement au bout d'un temps plus ou moins long,

* les stockages où la matière, arrive pour être mise en oeuvre. Ce type de stockage est associé à des chaînes de fabrication et permet d'entreposer aussi bien le produit à mettre en oeuvre, le produit fini ainsi que tous les produits intermédiaires (produits en cours d'élaboration, rebuts,...).

Quel que soit le type de stockage, le temps de présence des agents dans les stockages de plutonium, doit être réduit autant que faire se peut, d'où l'automatisation fréquente des opérations de transfert et de stockage.

Dans le cas où les conteneurs de plutonium en stockage sont équipés de capteurs thermiques, il est possible de suivre le combustible nucléaire en temps quasi-réel.

En effet, toute perturbation de la structure thermique du conteneur (par addition ou soustraction de tout ou partie de la matière calogène) entraîne des modifications des gradients thermiques et par suite, des signaux enregistrés. Il existe ainsi une relation permanente caractéristique entre le signal et l'intégrité thermique du conteneur.

II.- Principe

II.1.- Fluxmètrie thermique

Nous considérons le cas de la détermination expérimentale de la puissance thermique émise de façon quasi constante par un objet calogène dont le volume ne peut être considéré comme petit. Cette puissance thermique est évacuée par la surface externe d'aire et de géométrie invariable de l'objet. Il s'établit un flux thermique sortant de l'objet calogène.

Tout flux thermique F qui traverse une paroi de conductance K provoque entre la face d'entrée et la face de sortie une différence de température DT : F = K * DT. Une telle paroi constituera un fluxmètre si la différence de température DT peut être évaluée, K étant connue ou, au moins, constante pour une paroi donnée.

Si le fluxmètre entoure complètement l'objet calogène il constitue un calorimètre/1//4//11/. Les faces d'entrée et de sortie sont le plus souvent concentriques, de ce fait, on parle respectivement des enceintes (faces) interne et externe. Souvent, la face de sortie est en contact avec un puits de chaleur maintenu à température constante.

D'une manière générale, la conductance K est fixée une fois pour toutes lors de la construction du fluxmètre et comme il est très difficile de calculer la valeur exacte de K, le fluxmètre est étalonné avec des flux connus. En conséquence, pour un flux donné issu d'un objet calogène de géométrie donnée, un système fluxmétrique est caractérisé par:

- la valeur de K puisqu'elle fixe la hauteur de DT
- le type des capteurs qui transforment DT en signal électrique,
- le type d'enregistreur qui dépend de la hauteur du signal électrique,
- la jonction fluxmètre-enregistreur, liée également à la hauteur du signal électrique.

Il n'est donc pas possible de réaliser un appareil universel et très performant, travaillant dans un domaine très étendu de puissance thermique avec des échantillons de toute taille dans n'importe quel environnement. L'appareil devra être adapté au type de mesure envisagé/2//3/.

Alors que dans un calorimètre on prévoit une construction qui permet de négliger le flux thermique qui ne passe pas par le fluxmètre détecteur/1/, dans ce que nous avons appelé "FLUXMETRE", le rapport: $\frac{\text{(flux détecté)}}{\text{(flux total émis)}}$ est par principe très faible.

Ce principe est le suivant: si nous décomposons le flux thermique total émis par l'objet calogène en un grand nombre de flux unitaires, nous pouvons en sélectionner quelques uns pour modéliser le flux total: * cette modélisation ne sera réaliste que pour des objets calogènes de même géométrie et de puissance calorifique du même ordre,

* cette modélisation nécessite une étude thermique préalable de l'objet calogène et de son environnement.

Si donc on a pu modéliser le régime de fuite thermique de l'objet calogène au moyen de quelques flux unitaires seulement et si les capteurs thermiques sont de faible surface, la mise en place des fluxmètres sur l'objet à mesurer ne perturbe pas l'équilibre thermique aussi bien interne qu'externe de celui-ci, alors la mesure des différences de température est rapidement significative et exploitable.

Dans ce type de réalisation il n'y a plus de chambre de mesure donc il n'existe plus à proprement parler de faces d'entrée et de sortie "du fluxmètre détecteur" ; la face externe de l'objet calogène représente la face d'entrée d'un fluxmètre détecteur fictif somme de quelques petits fluxmètres unitaires. La face de sortie peut ne pas exister; dans ce cas, des structures, de préférence métalliques, environnant l'objet calogène vont permettre de reconstituer une face de sortie fictive.

II.2.- Fluxmètrie thermique et stockage de plutonium:

Pour équiper un stockage de capteurs thermiques permettant d'effectuer des mesures quantitatives de l'énergie calorifique dissipée par chaque conteneur, il faut tout d'abord connaître les diverses fluctuations thermiques s'y produisant habituellement au cours du temps, afin de pouvoir choisir et afficher les seuils d'alarme permettant de détecter les anomalies de gestion de façon sûre et sans générer de fausses alarmes.

Lorsqu'un conteneur va être placé dans une alvéole de stockage, il est caractérisé, et ses caractéristiques sont indiquées sur le bon de mouvement interne qui est émis avant le transfert. Ces caractéristiques sont, en particulier, la quantité et la qualité du plutonium, ainsi que le numéro de l'alvéole où ce conteneur va être placé. L'ordinateur qui prend en compte le transfert peut calculer la puissance calorifique théorique dégagée par ce conteneur. En effet cette puissance est fonction de la masse de plutonium et de sa composition isotopique, paramètres connus. L'ordinateur peut ainsi afficher la puissance que devra dissiper le conteneur, en face du numéro de l'alvéole.

Une fois que l'équilibre thermique du conteneur dans l'ambiance de l'alvéole a été atteint, il y a affichage sur l'écran de l'ordinateur de la puissance calorifique mesurée à l'aide des capteurs et comparaison de cette puissance avec celle attendue. Il est alors possible de déceler, en quelques minutes, une erreur d'adresse (le conteneur n'a pas été placé dans la bonne alvéole) et en quelques heures, une différence significative (par exemple 6%) entre la puissance mesurée et la puissance attendue.

Cette différence peut aussi provenir d'une erreur sur la masse de plutonium ou d'une erreur sur la composition isotopique annoncées. Il y a dès lors automatiquement demande de vérifications.

Si aucune anomalie n'est détectée, la puissance calorifique dégagée est suivie au cours du temps. Dans la pratique, les enregistrements des signaux fournis par les capteurs "Mesure" et les capteurs "Référence" équipant un conteneur en stockage, montrent que la valeur de "DT" (T°mesure - T°référence) reste pratiquement constante quelle que soit l'amplitude des variations thermiques lentes qui se produisent dans les stockages rencontrés.

S'il y a une variation brusque de la valeur de cette puissance, une alarme se déclenche. Cela veut dire que toute intrusion non programmée dans l'alvéole déclenche un signal d'alarme. Par intrusion on entend ici non seulement l'ouverture ou l'enlèvement du conteneur, mais aussi une simple ouverture de la porte ou de l'opercule de l'alvéole.

III.- La transmission des données.

Deux types de transmission des données ont été expérimentées : la transmission par fils et celle par radio.

III.1.- La transmission par fils.

Nous rappelons que chaque conteneur instrumenté est relié par un câble comportant au moins trois conducteurs à un ensemble électronique. Cet ensemble comprend :

- un scrutateur de voies permettant d'interroger tel ou tel conteneur de manière séquentielle ou aléatoire,
- un multimètre permettant de connaître les valeurs ohmiques des capteurs de référence et mesure,
- un ordinateur qui va calculer les différences de température et gérer le système de gestion des informations..

Cette transmission par fils présente l'inconvénient, en version mobile d'étaler un écheveau de fils sur le sol du lieu d'implantation et en version fixe dans un stockage, d'avoir à effectuer des travaux importants lorsque le stockage n'a pas été précâblé à la construction, ce qui est le cas général. Nous avons donc été amenés à concevoir et faire réaliser une transmission par radio:

III.2.- La transmission par radio:

Chaque conteneur est équipé d'un dispositif de faible volume, que nous appellerons "BALISE" permettant d'une part de mesurer les valeurs ohmiques des thermistances (Référence et Mesure) et, d'autre part, d'émettre des signaux. Trois versions avec des applications distinctes ont été développées :

III.2.1.- Transmission continue:

Les thermistances "Mesure", "Référence" et une résistance "Etalon" interne à la balise, sont interrogées par un scrutateur à trois voies et les valeurs ohmiques sont transmises de façon continue par la balise, après codage, au calculateur qui, après réception et décodage, évalue la puissance thermique émise par le conteneur.

Chaque conteneur est donc équipé, pour la durée de la mesure d'un ensemble de capteurs et d'une balise qui fonctionne sur un canal hertzien propre. Le nombre maximum de conteneurs, qui peuvent être équipés dans une même salle de stockage, est alors limité par le nombre de canaux hertziens disponibles soit de l'ordre de quelques dizaines. L'émission de chaque balise étant continue, la scrutation de chaque conteneur, en particulier pendant les périodes riches en mouvements de matière nucléaire, pourra se faire à la fréquence désirée.

L'autonomie électrique d'un émetteur alimenté par batterie est supérieure à 8 heures d'émission continue (en l'absence de recharge permanente par élément photovoltaïque) et la portée utile est de 50 mètres.

L'existence de l'étalon interne affranchit la transmission de toute dérive éventuelle des codeurs, décodeurs, émetteurs ou récepteurs quelle qu'en soit la cause et permet d'effectuer une mesure dont la précision n'est limitée que par la technique de codage.

La balise a pour dimensions H . l . p : 10 x 6 x 5 cm dans sa version préindustrielle. Ce type de balise est plus particulièrement adapté aux contrôles périodique d'inventaire (par exemple annuel).

III.2.2.- Transmission périodique ou d'alarme:

Les fluxmètres thermiques se comportent et sont gérés comme des "Scellés thermiques" dans l'application "Confinement-Surveillance" de stockages longue durée de préférence.

Dans ce dispositif, chaque "balise" sonde pendant un temps très bref les ensembles de thermistances "Mesure" et "Référence". Elle calcule la différence de température et la compare à une valeur de consigne, spécifique du conteneur suivi et de son contenu, valeur qui est introduite dans la balise à la mise en surveillance du conteneur.

En cas de valeur anormale, une deuxième puis une troisième mesure sont effectuées pour vérification. En cas de confirmation de l'anomalie, un signal d'alarme est émis avec transmission sur une fréquence (commune à tous les émetteurs qui dépendent d'un même récepteur de surveillance,appelé "Centrale") de la nature de l'alarme (signal pas conforme ou tension d'alimentation faible) et du n° de l'émetteur en alarme.

Les différentes périodicités (temporisation à la mise en route de la surveillance, durée d'une mesure, intervalle de temps entre deux mesures d'un groupe de trois, intervalle de temps entre groupes de trois mesures...) sont toutes adaptables aux cas particuliers.

Pour une interrogation toutes les deux minutes et en l'absence d'émission du signal d'alerte, la durée de vie de la batterie est de deux ans environ (en l'absence de recharge photovoltaïque). Dans la version de présérie que nous avons expérimentée, les dimensions de la balise sont: L x l x h: 15 x 9 x 3 cm et la centrale permet de gérer quelques centaines de balises simultanément.

III.2.3.- Transmission sequentielle ou aléatoire:

C'est le type de transmission qui a été choisi pour la "Gestion en temps quasi-réel d'un stockage de plutonium" objet de la présente communication.

Chaque balise est interrogée dans l'ordre et à la fréquence choisis par l'ordinateur de gestion; ainsi les valeurs des différentes périodicités ne sont pas figées à la construction de la "Balise" en fonction des spécificités du stockage et des conteneurs gérés. Cette souplesse d'utilisation est mise à profit pour changer le rythme des interrogations de "Balises", par exemple, en fonction des périodes d'activité et d'inactivité du stockage.

Une "Balise" sonde les chaînes de thermistances "Mesure" et "Référence" ainsi que sa propre tension d'alimentation, code ces trois valeurs et les transmet sur demande à l'ordinateur de gestion.

En l'absence de recharge photovoltaïque, chaque balise de la version non industrialisée, que nous avons expérimentée, peut être interrogée quelques milliers de fois. Le nombre de balises gérées par un même ordinateur n'est pas limité et les dimensions (réductibles) de chaque balise sont provisoirement de L x l x h : 15 x 9 x 3 cm.

IV.- Présentation des données:

Les données de température "Mesure" et "Référence" ayant été archivées dans l'ordinateur de gestion, sont traitées et, au choix, affichées en temps réel ou rappelées pour analyse d'un évènement.

IV.1- Visualisation en temps réel:

La copie d'écran de la figure 1 présente l'aspect habituel de l'écran de contrôle lors d'un mouvement de matière nucléaire.

Les cages 5 et 7 sont en alarme, car une heure et demie aprés leur chargement le DT mesuré est déjà supérieur au DT calculé à partir de la fiche d'inventaire.

La cage 8 est en alarme depuis deux heures et pour encore quelques minutes car elle a subit une suppression de matière caloactive, opération qu'il convient de justifier.

La cage 6 est en attente d'équilibre car quatre heures après son chargement elle n'a pas encore exactement atteint le palier de DT attendu.

On peut noter que dans notre cas où seulement neuf cages centrées sont en surveillance, les barregraphes sont inutilement détaillés et sur l'actuelle présentation en noir et blanc les informations non chiffrées sont difficilement exploitables. En fait cette présentation a été développée pour des matrices de 6x9 à 8x8 avec la légende latérale ou 4x12 à 6x12 avec une légende sur trois lignes horizontales.

Si le nombre de conteneurs à surveiller est supérieur aux possibilités des matrices disponibles, l'écran de visualisation peut représenter, sequentiellement, autant de matrices que nécessaire; il semble toutefois préférable de choisir un nombre et une disposition d'écrans de visualisation qui permettent une représentation spatiale semblable à la disposition spatiale des conteneurs .

IV.2.- Visualisation pour étude d'une situation:

En fonction des connaissances acquises sur le comportement thermique des conteneurs et/ou celui de la salle de stockage, les valeurs de consigne, de DT et de ses limites acceptables (±6%) sont affichées dans les cases de la Figure 1. En situation d'alarme, il est nécessaire, dans un but d'analyse, de pouvoir visualiser l' "Historique" des comportements thermiques des conteneurs pendant une période choisie. Sans interrompre l'enregistrement de la surveillance, il est possible de faire s'afficher différents historiques comportant des effets de ZOOM sur la variable "température" ou sur la variable "temps". On peut choisir de tracer l'historique d'un conteneur ou l'historique de plusieurs conteneurs contigus.

Ce dernier mode de représentation est en général plus explicite, car de la comparaison des différents enregistrements plus d'enseignements peuvent être tirés.

Il est à noter que si les capteurs "Référence" sont, comme ici, directement dans l'air ambiant leur grande sensibilité aux fluctuations de température de l'ambiance (qui les rend aptes à servir de détécteur d'intrusion) entraîne une instabilité permanente du signal DT. Si, en revanche, on désire une meilleure précision dans le contrôle quantitatif il suffit de positionner les capteurs "Référence" sur une structure métallique, proche des conteneurs, dont la température est représentative de l'environnement de ceux-ci.

V.- Mise en oeuvre expérimentale

V.I.- Installation technique:

Le magasin de matières fissiles du Centre de Cadarache équipé des cages centrées FS52 (utilisé pour les démonstrations du système) est typique des stockages longue durée évoqués en III.2.2. Il n'est pas prévu d'y ouvrir les cages centrées de transport pour en modifier le contenu, enfin les séjours répétés de personnels n'y sont pas recommandés.

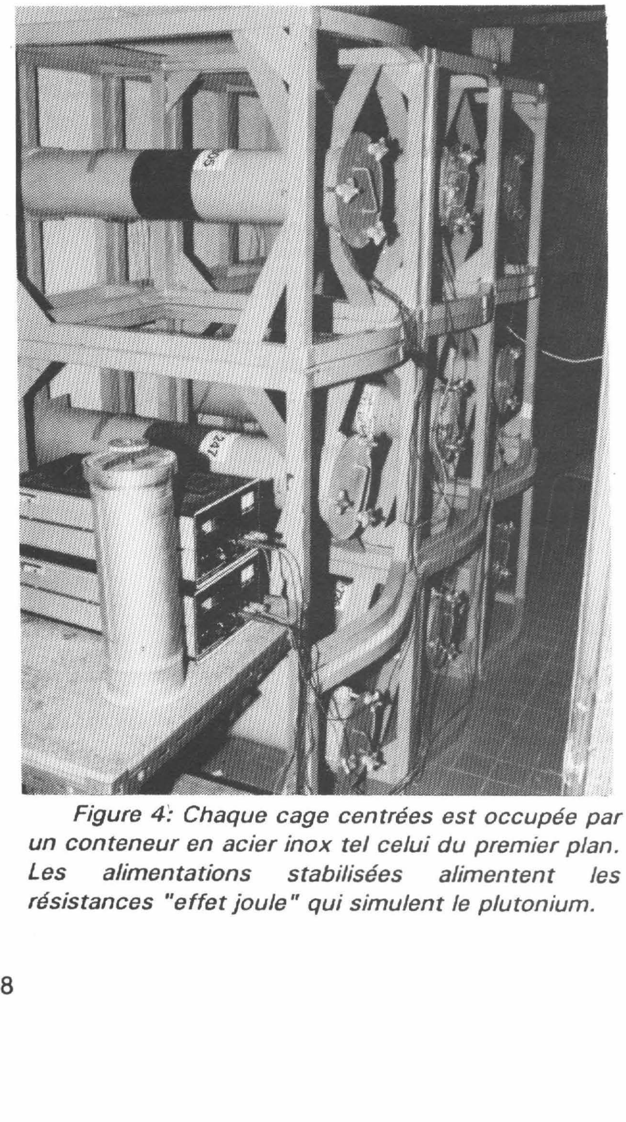
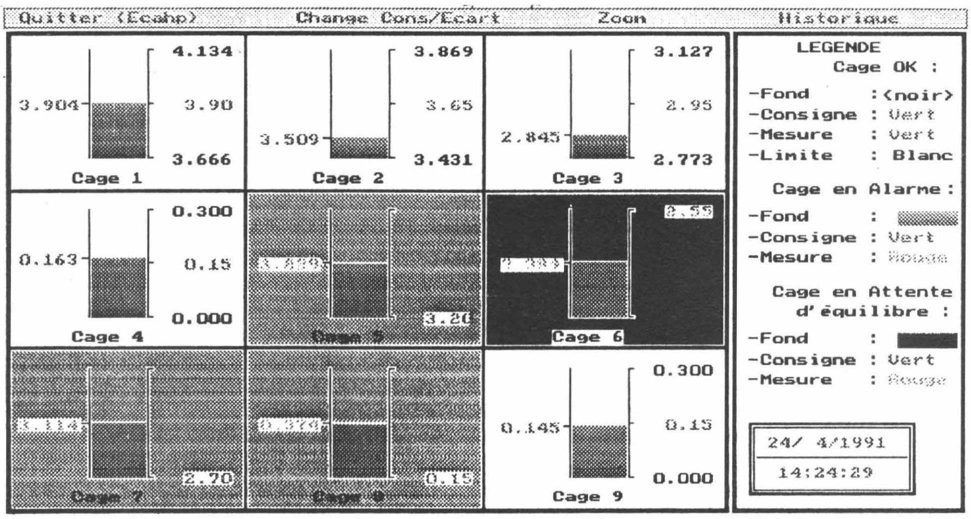


Figure 4: Chaque cage centrées est occupée par un conteneur en acier inox tel celui du premier plan. Les alimentations stabilisées alimentent les résistances "effet joule" qui simulent le plutonium.



LEGENDE

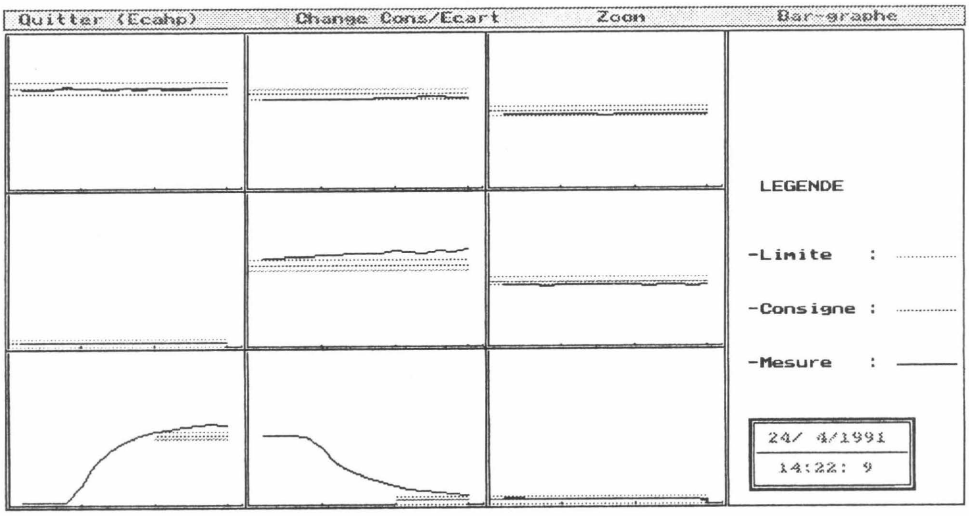
Cage OK :
-Fond : <noir>
-Consigne : Vert
-Mesure : Vert
-Limite : Blanc

Cage en Alarme :
-Fond : ▨
-Consigne : Vert
-Mesure : Rouge

Cage en Attente d'équilibre :
-Fond : ■
-Consigne : Vert
-Mesure : Rouge

24/ 4/1991
14:24:29

Figure 1: L'écran dans le mode suivi en temps réel. La présentation comporte, dans la version réelle, un code couleur qui est explicité dans la légende .



LEGENDE

-Limite : ·········
-Consigne : ·········
-Mesure : ———

24/ 4/1991
14:22: 9

Figure 2: Rappel des DT précédant la situation décrite par l'écran de la Figure 1.

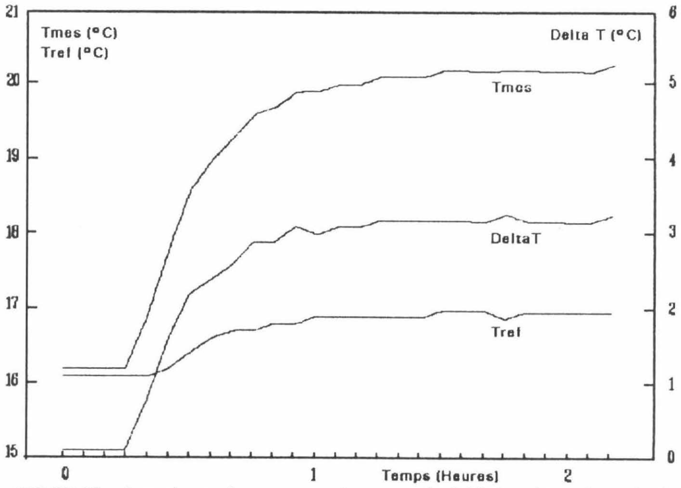


Figure 3: ZOOM" des deux heures qui ont précédé la situation de la Figure 1, cage 7.

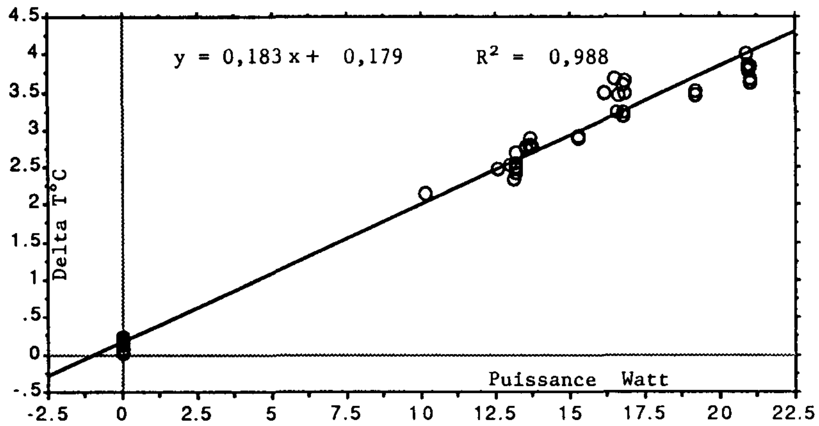


Figure 5: Différence de température entre la surface du fût des cages centrées et l'air ambiant pour différentes puissances thermiques produites dans les cages

Parameter:	Value:	Std. Err.:	Std. Value:	t-Value:	Probability:
INTERCEPT	.179				
SLOPE	.183	.003	.994	72.025	.0001

Confidence Intervals Table

Parameter:	95% Lower:	95% Upper:	90% Lower:	90% Upper:
MEAN (X,Y)	1.999	2.081	2.006	2.074
SLOPE	.178	.188	.179	.187

Aussi ce stockage, s'il se prête bien à la présentation du système de Gestion, ne peut être un lieu d'expérimentation.

Pour la mise au point du système de "Gestion en temps quasi-réel des stockages industriels de plutonium" nous opérons dans un stockage modélisé en laboratoire. Ce lieu expérimental est constitué d'une salle dont la climatisation peut être modulée à volonté. La puissance installée (en chauffage, réfrigération, et ventilation) associée à la possibilité de régulation fine de la température (à 0,1 °C près si nécessaire) permet d'y simuler les comportements climatiques des stockages dont on considère l'équipement comme possible.

Dans cette salle d'expérimentation nous avons disposé, à titre d'exemple, des cages centrées en acier peint, de type DV21, présentées sur la photographie de la figure 4 dans une des dispositions envisageables. On peut noter que la simulation du plutonium est obtenue avec des éléments chauffants par effet Joule noyés dans du sable dans des conteneurs inox dont on voit un exemplaire sur la même figure.

Afin que le système que nous developpons puisse fonctionner, il est indispensable que l'ensemble constitué par les conteneurs de plutonium en place dans les casiers de stockage ou, ici, les cages centrées instrumentées de capteurs thermiques se comporte comme des fluxmètres.

La figure 5 ci-dessus montre que le DT mesuré sur le fût central des cages centrées est bien proportionnel à la puissance calorifique comprise entre 0 et 20 Watts à ±6% dans des conteneurs en acier inoxydables prévus pour recevoir jusqu'à trois kilogrammes de combustible sous forme d'oxyde. L'emplacement et la construction des capteurs thermiques sont spécifiques du type de stockage.

V.- Conclusions

Dans l'état actuel de développement de notre système d'aide à la "Gestion en temps quasi-réel de stockages de Plutonium" les problèmes d'ordre strictement technique (équipement informatique, équipement électronique de mesure et de transmission de celles-ci, logiciel de gestion de l'intallation électronique) ont reçu une solution. Le logiciel a été conçu de façon modulaire afin de permettre souplesse et portabilité. Il conviendrait maintenant d'adapter le logiciel à un stockage réel siège de fréquents mouvements de matière caloactive afin qu'il puisse faire la preuve de son utilité.

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EVALUATION D'UNE PREMIERE SERIE DE SCEAUX TITUS 1 PRODUITS INDUSTRIELLEMENT

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ABSTRACT

The Titus 1 seal is an on-site verifiable ultrasonic multipurpose seal which has been developped in JRC-Ispra in the 80'. it can be directly and automatically verified by means of the ASTUS* portable equipment.
On a first production of 75 seals, manufactured in 1990 by the French company ELCA, the Seals and Identification Techniques laboratory of the Institute for System Engineering and Informatics of the JRC-Ispra has carried out an evaluation study, in particular regarding: the unicity and the stability of the signatures; the behaviour at extreme temperatures, under thermal cycles, and under mechanical shocks.
The points where the production is satis factory and those where the quality should be improved are discussed.

1)INTRODUCTION

Le sceau Titus 1, issu d'études communes CEA-CCR Ispra (Brevet 14278), est destiné à être soumis à des conditions climatiques variées ainsi qu'à subir des chocs mécaniques durant son utilisation.Cet article a pour objet de présenter le comportement d'un tel sceau dans de telles conditions d'utilisation.

2)MATERIEL ET TYPE D'ESSAIS (Ref.1):

L'obtention du signal électrique de signature des sceaux Titus 1 est réalisé par l'excitation périodique, au niveau du scellé,d'un transducteur céramique,collé contre la partie interne supérieure du sceau, émettant un train d'ultrasons,qui après de multiples réflexions dans la partie métallique du sceau, sertissage compris, crée en retour sa caractéristique ultrasonore. La signature définitive du sceau Titus n'est obtenue qu' apres sertissage de celui-ci.

75 de ces sceaux (Figure 1) nous ont été fournis par la société Francaise Elca; sceaux qui, dans cette étude, ont été lus par un système analyseur de signal "Data 6000". (Ref.2)

Avant sertissage, nous avons étudié l'unicité de chacune des identités à une température de 20°.

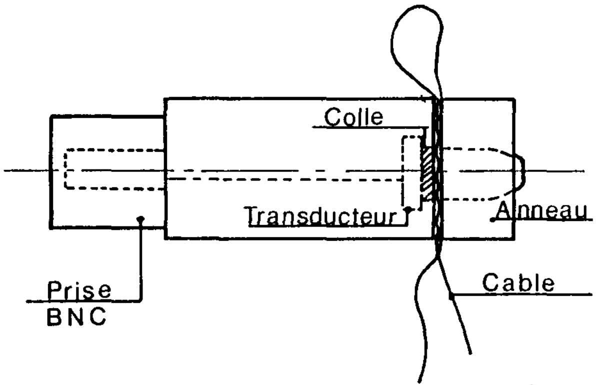

Figure 1: Sceau Titus 1 ech. 2/1

Parmi ces 75 sceaux, nous en avons choisi arbitrairement 10 qui ont été scellés et étudiés plus en détail dans leur comportement lorsqu'ils sont soumis à 4 cyclages thermiques allant de -10 a +50 degrés (Figure 2), en comparant leur signature relevée après chaque cycle thermique avec leur même signature mais prise à 20 degrés avant toute sollicitation de ce genre.
Nous avons également étudié la possibilité de vérifier de tels sceaux (identification de leur signature) à des températures extrêmes telles que -10 et +50 degrés. Ces différents essais ont été réalisés a l'aide d'un four à résistance et d'une enceinte frigorifique nous permettant des températures de -20 a +200 degrés. Afin de vérifier à tout moment la température des sceaux, chacun d'eux a été instrumenté avec un thermocouple de type chromel-alumel placé en surface et relié à un thermomètre digital.

Durant son emploi, le sceau Titus 1 peut être soumis à des chocs mécaniques; afin d'étudier sa sensibilité à de telles contraintes, nous avons effectué les essais suivants,avec les sceaux précédemment cités:
-Chaque sceau a été soumis à quatre séquences de 25 chutes libres avec réception sur une plaque d'acier; les deux premières séquences ont été réalisées avec une hauteur de départ de chute de 28 cm., soit une énergie potentielle de 0.042 J, (la masse moyenne d'un sceau étant de 15.4 g); les deux séquences suivantes avec une énergie potentielle de 0.130 J (H= 86 cm.). Après chaque sequence de 25 chutes, la signature de chaque sceau a été relevée et comparée à elle même,mais prise à 20 degrés avant toute chute.

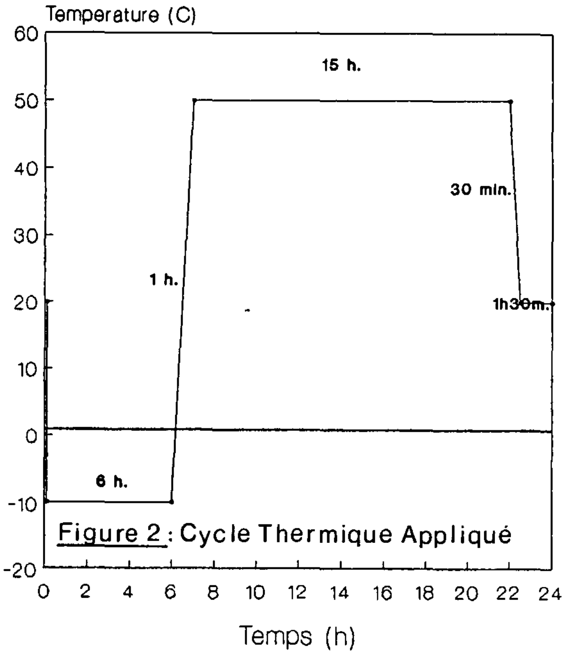

Figure 2 : Cycle Thermique Appliqué

5)RESULTATS:

5-1)Unicité des identités avant sertissage

En effectuant les cross-corrélations, suivant la méthode de Bravais-Pearson, des 75 signatures relevées à 20°C, soient 2730 calculs,il apparait que le coefficient de corrélation obtenu (Kcorr) est compris entre -0.3 et 0.9; la majorité des coefficients étant située entre 0.1 et 0.75. (Figure 3). De plus, en relevant deux fois la signature d'un même sceau,le coefficient est supérieur à 0.96. Il semble donc que l'unicité des 75 sceaux Titus 1 soit satisfaisante à 20 degrés.

5-2)Comparaison avant-après sertissage

En comparant les signatures des 10 sceaux sertis avec elles mêmes, mais relevées avant sertissage nous obtenons des coefficients de corrélation variant de 0.645 à 0.910 avec une large majorité sous la barre des 0.9.Il est donc très difficile de reconnaitre un sceau après sertissage en comparant sa signature avec celle obtenue avant cette opération; ceci étant dû au fait que le sertissage engendre une déformation de la partie supérieure du sceau dont le retour de l'onde caractéristique de signature dépend. Ce phénomène constitue une sécurité importante qui interdit pratiquement toute reproduction du sceau, celui-ci ne possédant son identité définitive qu'une fois installé.

5-3)Tenue aux cyclages thermiques

En comparant les signatures des 10 sceaux sertis obtenues après chacun des quatres cycles thermiques présentés Figure 2, avec leur homologue relevée avant ces essais, nous obtenons des coefficients de corrélation tous supérieurs à 0.960; les cross-corrélations effectuées entre les dix sceaux étant toujours inférieures à 0.796.
Les sceaux Titus 1 supportent bien des variations de température de -10 à +50°C pour autant que l'identification soit faite à température ambiante.

5-4)Comportement aux températures extrêmes

A 50°C, il est aisé de reconnaitre la signature d'un sceau Titus en la comparant avec elle même mais prise à l'ambiante;par contre, à -10 degrés (Figure 4),cette vérification est devenue impossible, la signature ayant trop changé.

Des essais complémentaires ont permis de préciser la gamme de températures dans laquelle les sceaux Titus 1 peuvent être vérifiés (peu de changements dans leurs identités):soit 0 à +50 C

5-5)Sensibilité aux chocs mécaniques

Parmi les dix sceaux sertis ayant été soumis aux séquences de chutes libres, deux d'entre eux ne possedaient plus de signal de retour (signature) après les 25 premières chutes d'une hauteur de 86 cm., et un troisième était dans le même état à l'issue de la seconde séquence. Les 7 restants présentent de legères altérations de leur identité qui n'empèchent pas leur vérification.

6)CONCLUSION

Sur une population de 75 sceaux Titus non sertis produits industriellement:
-L'unicité des identités est satisafaisante à 20°
-La reconnaissance des signatures est aisée.

Sur un échantillon de 10 sceaux Titus 1 sertis, nous avons vérifié que:
-Aucune défaillance n'a été relevée entre avant et après sertissage.
-Les signatures sont stables dans le temps à température de 20 degrés.
-Le sceau Titus 1 ne possède son identité définitive qu'une fois serti, ce qui est un important facteur de sécurité.
-Il semble que le sceau Titus 1 supporte bien les variations de température dans une gamme de -10 à +50 Degrés , aucune défaillance n'ayant été relevée.L'identification, après de telles variations thermiques, se fait aisément, à condition de relever signature à température ambiante.
-La vérification d'un sceau Titus 1 à -10 degrés n'est pratiquement pas possible;la gamme de températures admises étant de 0 à +50 degrés.
-Le sceau Titus 1 présente une fragilité certaine aux chocs mécaniques.

Si la fragilité mécanique des sceaux Titus 1 était amoindrie,soit par un progrès technologique soit par une protection extérieure (caoutchouc), et si l'on réussissait à réduire ou compenser les modifications de signatures à basses températures (ex: amélioration du collage du transducteur), le sceau Titus 1, tel que produit actuellement de façon industrielle, vue sa grande simplicite d'utilisation,pourrait être utilisé conformément aux espoirs des concepteurs(Sceau tous usages).Sinon, on devra limiter son utilisation dans des domaines où les conditions sont moins contraignantes.

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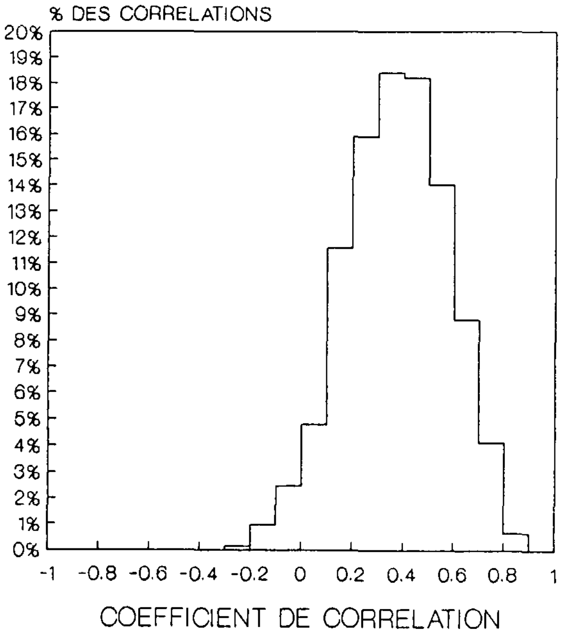
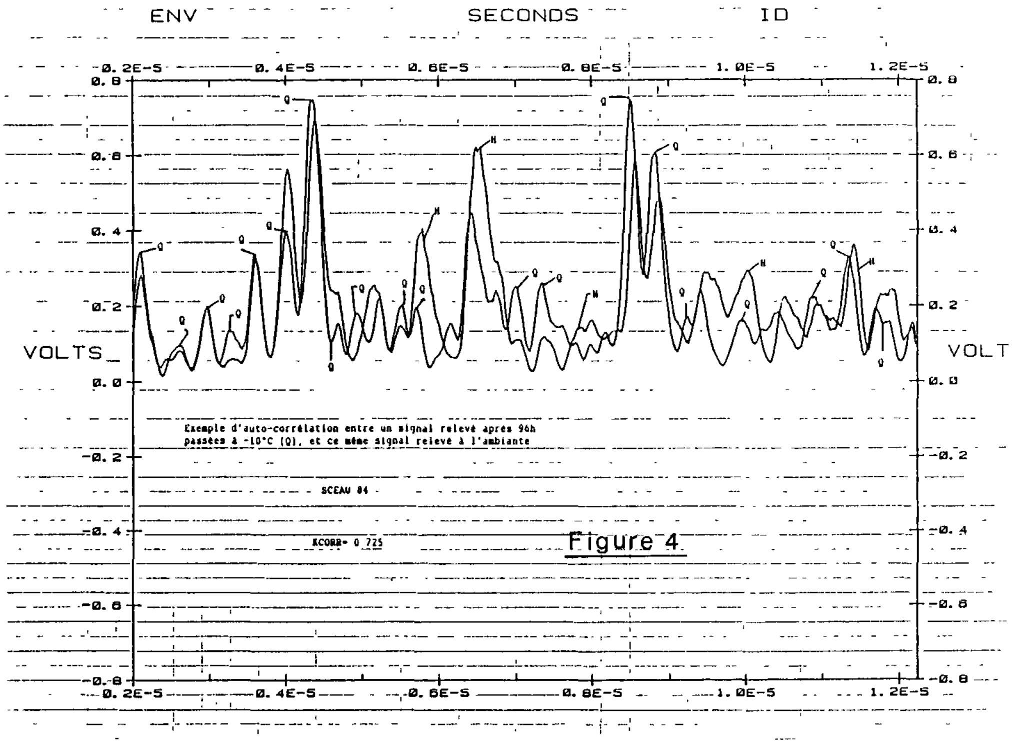
/2/-R.Denis-"Verification of Titus seal identities using the "Analogic Data 6000" system". Note Technique Ref. 173/40-422/88/rd/rmb-Commission des Communautés Européennes-C.C.R. Ispra-Février 1988.

Figure 3: HISTOGRAMME DES CORRELATIONS AVANT SERTISSAGE DES 75 SCEAUX TITUS



2730 CORRELATIONS



Exemple d'auto-corrélation entre un signal relevé après 96h passées à -10°C (Q), et ce même signal relevé à l'ambiante

SCEAU 84

%CORR= 0.725

Figure 4

SYSTEME ASTUS DE VERIFICATION DE SCEAUX
TRANSPORT TITUS I

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Abstract

ASTUS, a system for acquisition and processing ultrasonic signatures of TITUS I seals has been developed. TITUS seals are used to verify the integrity of the fissile material's container sealing after transport.

An autonomous portable reading case permit to take seals signatures at the starting point and to transmit these reference signatures to a central safeguards computer by phonic modem. Then, at the terminal point with a similar reading case, an authority takes again the signature of seals and immediately transmit these signature to the central safeguards computer. The central computer processes the data in real time by autocorrelation and return its verdict to the terminal point.

1. Introduction

Le système ASTUS (Acquisition de Sceaux de transports Ultra Sonores) a pour but d'assurer la non-violabilité de la fermeture de conteneurs lors d'un transport. A cet effet, la caractérisation, ou signature, ultrasonore d'un scellé placé sur le conteneur est relevée sous forme électrique avant puis après transport, dans le but de décider après comparaison, de l'intégrité du sceau.

Au lieu de départ, une phase d'acquisition à l'aide d'une valise portable et autonome permet la prise en compte de plusieurs signatures. Ensuite, une phase de transmission est effectuée par voie téléphonique vers le Poste Central de Sécurité. Ce dernier stocke chaque signature horodatée constituant un fichier de référence.

Au point d'arrivée du transport, ou en cours de route, il est possible à l'aide d'une autre valise de même nature, de prélever de nouvelles signatures et de les transmettre vers le Poste Central via une ligne téléphonique. Un logiciel de traitement en temps réel par corrélation détermine alors l'intégrité du scellement par rapport aux signatures de références.

2. Principe du système

Les sceaux ultrasonores sont de type TITUS I issus d'études communes CEA-CRR ISPRA (Brevet n° 14278 en 1984). L'obtention du signal électrique est réalisée par l'excitation périodique, au niveau du scellé, d'un transducteur céramique émettant un train d'ultrasons, qui après de multiples réflexions dans la partie métallique du sceau, sertissage compris, crée en retour sa caractéristique ultrasonore.

Les valises sont constituées d'un PC portable, THOSHIBA T 1600, équipé d'une carte modem. Elles possèdent un petit interface permettant l'excitation des scellés et l'échantillonnage des signatures sur 1024 points avec une dynamique de 8 bits. L'autonomie de plusieurs heures permet l'acquisition et la transmission d'une centaine de signatures.

Les transmissions téléphoniques se font soit en mode acoustique, soit en mode électrique à 1200 bauds avec compression de données. La durée d'un transfert est de l'ordre de 30 secondes par signature.

Le Poste Central est l'organe de décision sur l'intégrité d'un scellement. Il est constitué d'un PC Compacq 386 équipé d'un disque dur de 84 Moctets. Le traitement tient compte des différences de mesures, biais et facteur d'échelle, retard, dues aux chaînes d'acquisition de chaque valise. L'influence de la température est aussi compensée dans la gamme d'utilisation 0°C à 50°C. Le traitement par couple de signatures, référence et test, élabore le taux d'autocorrélation qui est comparé à un seuil préalablement déterminé par études statistiques.

La Société ELCA (électronique du Capitole, TOULOUSE) a mené à bien la fabrication industrielle des scellés et développé avec le CEA les modules d'acquisition et de transmission des signatures ainsi que les logiciels de traitement des données. Le système présenté est une première version qui est en phase d'évaluation opérationnelle sur des transports représentatifs dont les premiers résultats seront présentés. Il sera possible d'effectuer des démonstrations (acquisition de signatures et vérification de celles-ci entre AVIGNON et PARIS).

3. Description de l'ensemble

Le systeme d'acquisition et de contrôle des sceaux TITUS I est constitué de deux sous-ensembles :

. Un ensemble portable.
. Un poste central de contrôle.

3.1. Ensemble portable

L'ensemble portable est constitué des sous-ensembles suivants :

. Un générateur ELCA.
. Un PC autonome TOSHIBA T 1600.

L'ensemble portable réalise l'acquisition des signatures ultrasonores analogiques des sceaux TITUS I et la transmission des signatures vers le post central par liaison téléphonique.

Générateur ELCA

Il réalise :

- l'acquisition des signatures ultrasonores analogiques des sceaux TITUS I

  Echantillonnage : 50 MHz
  Nombre de points : 1024
  Plage d'échantillonnage : 4 s à 24 s

- la transmission des signatures par liaison série RS 232 vers le PC autonome

  Vitesse de transfert : 9600 bauds
  Format : 8 bits de données
  2 bits de stop pas de parité

Le générateur est alimenté à partir de la barrerie du TOSHIBA T 1600.

PC autonome TOSHIBA T 1600

Le portable autonome TOSHIBA T 1600 réalise deux fonctions :

- l'acquisition des signatures. Il pilote le générateur ELCA par liaison série,
- la transmission de ces signatures vers le poste central par liaison téléphonique ou série.

Caractéristiques du TOSHIBA T 1600 :

- Microprocesseur 80 C 286 (16 bits) 12 MHz,
- 1 Mo de mémoire RAM,
  . Fonctionnement : 5 à 35°C
- 1,4 Mo de disque souple 3" 1/2,
  . Poids : 5,2 kg
- 20 Mo de disque dur,
- Coprocesseur arithmétique 80 C 287, 8 MHz,
- Autonomie 2 à 5 heures.

Modem électrique/acoustique :

$$1200 - 2400 \text{ bauds} \begin{cases} \text{commandes HAYES} \\ \text{correcteur d'erreur MNP4} \\ \text{compression de données MNP5} \end{cases}$$

. Durée d'acquisition d'une signature = 10 s
. Transmission d'une signature vers le poste central = 30 s
. Test en auto-corrélation = 15 s

Acquisition des signatures

Le portable autonome TOSHIBA T 1600 utilise le port série repéré SER-A pour dialoguer avec le générateur, en vue d'acquérir les signatures des sceaux.

Les signatures sont ensuite normalisées et stockées dans un fichier sur disque dur.

Les informations contenues dans le fichier sont les suivantes :

- nom du sceau,
- date de l'acquisition,
- température,
- 1024 points normalisés,
- checksum des 1024 points.

Transmission des signatures vers poste central

La transmission peut s'effectuer soit par liaison série soit par liaison téléphonique.

- Liaison série :
  La transmission par liaison série ne peut s'effectuer que si le T 1600 est proche du poste central. Dans cette configuration, le T 1600 dialogue à 9600 bauds avec le port série du poste central.

- Liaison téléphonique :
  La transmission par liaison téléphonique s'effectue lorsque le T 1600 est éloigné du poste central. Dans cette configuration, le T 1600 utilise son modem interne pour dialoguer à la vitesse de 1200 bauds.

Lors de la demande de transmission, le T 1600 va chercher dans le répertoire SIGNAT le fichier correspondant à la demande avec l'extention ".CTL". Si celui-ci n'existe pas, il va transmettre le fichier avec l'extention ".REF". Si aucun des deux fichiers n'existe, alors aucun fichier ne sera transmis.

Après la transmission du ou des fichiers (maximum 8), le T 1600 passe en attente des résultats provenant du poste central.

Tous les résultats issus du poste central sont visualisés sur l'écran.

Le poste central de contrôle mémorise sur disque dur les signatures reçues et les analyse pour délivrer les messages suivants :

"SCEAU INTACT"
ou
"SCEAU SUSPECT"

3.2. Le poste central

Le poste central a été réalisé autour d'un PC COMPACQ Deskpro 386 S fonctionnant à 16 MHz.

Equipé d'un coprocesseur arithmétique, il offre une puissance de calcul satisfaisante pour effectuer le contrôle des signatures des sceaux.

Une imprimante à aiguilles permet d'écrire les résultats provenant des tests.

Un modem externe relié par liaison série permet de recevoir par liaison téléphonique les signatures et de transmettre les résultats du contrôle à l'ensemble portable.

Caractéristiques du poste central :

- Microprocesseur 386 SX, 16 MHz,
- 2 Mo de mémoire RAM,
- 1,2 Mo de disque souple 5" 1/4,
- 84 Mo de disque dur,
- Coprocesseur arithmétique 387 SX, 16 MHz.

<u>Méthode retenue pour les calculs des taux d'auto-corrélation</u>

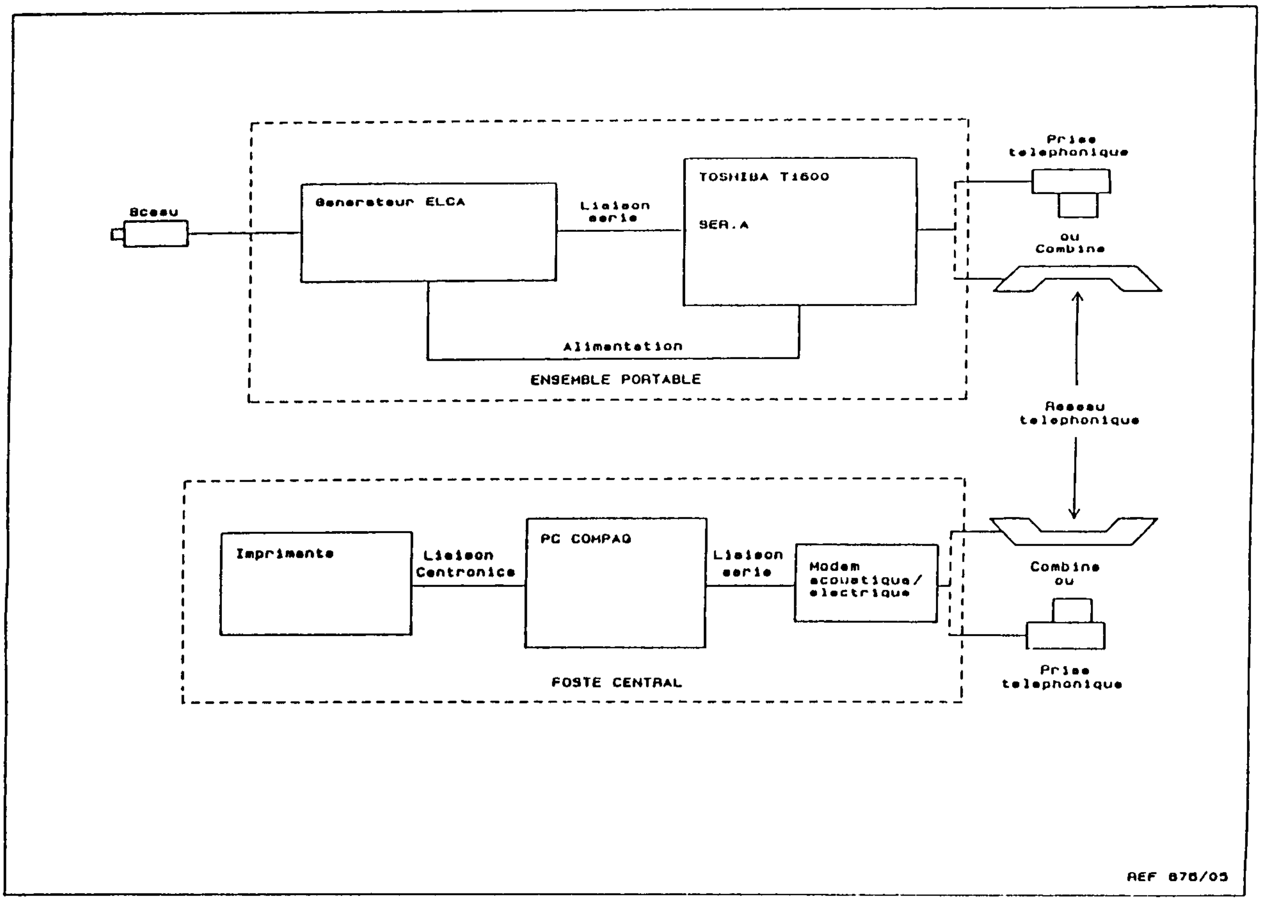
C'est la méthode de calcul mise au point par ISPRA qui a été retenue.

Les signatures haute fréquence (1024 points) issues du sous-ensemble portable sont filtrées (convolution triangle).

Lors de ce filtrage, seuls les 512 premiers points sont pris en compte, ceci afin d'éviter les problèmes dus aux variations de température (étirement de la signature avec la température).

Les enveloppes du taux de corrélation obtenu après la compensation en température doit être supérieur à x % pour déclarer le scellé INTACT ; dans le cas contraire, le scellé est déclaré SUSPECT.

La durée nécessaire à l'acquisition des signatures et à son contrôle est inférieure à 1 minute.



<u>FIGURE 1</u> : SYNOPTIQUE (Poste Central - Ensemble portable)

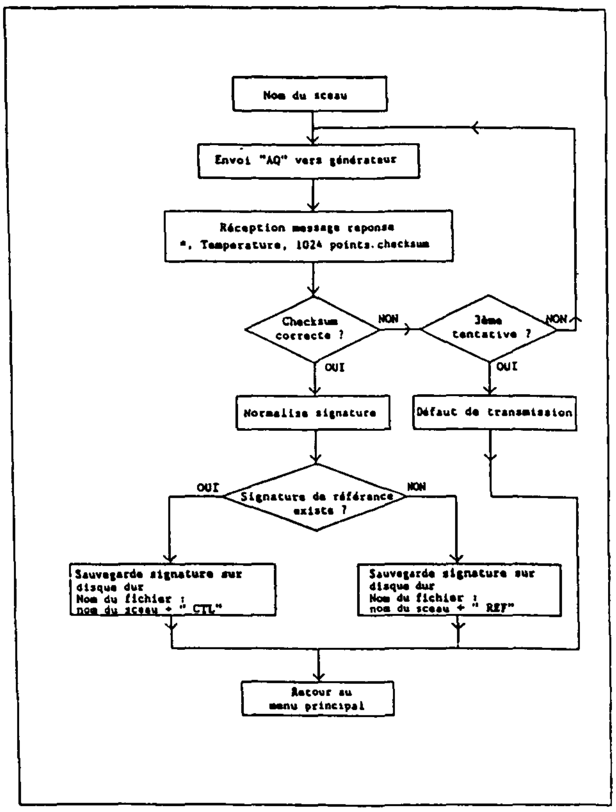


FIGURE 2 : Logigramme d'acquisition des signatures

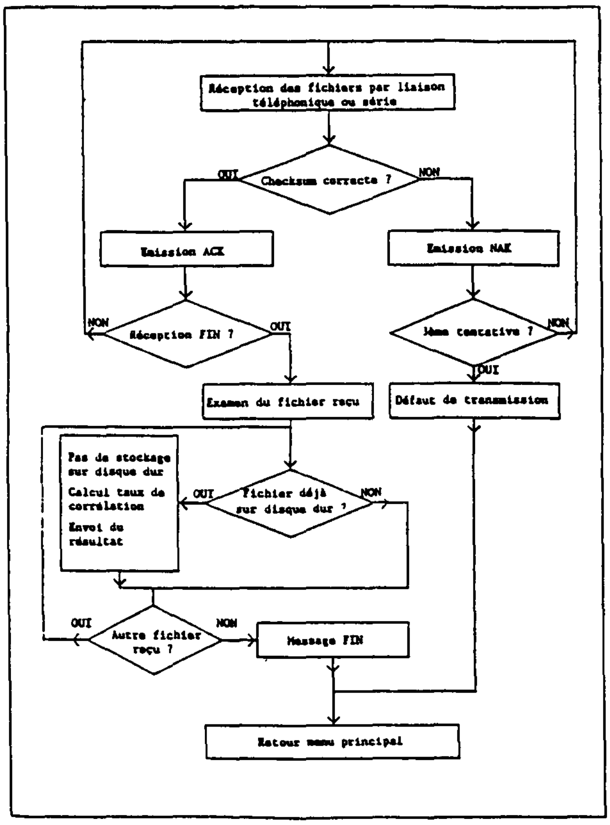


FIGURE 3 : Logigramme de traitement des signatures

IMPROVING IMAGE REVIEWING WITH A NEW SYSTEM (EMOSS, MEMOBANK AND MOVICOM DEVICES)

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Abstract

The need is obvious for selecting the relevant images and/or items of information from among the overwhelming amount produced by C/S systems, mainly video cameras. Image reviewing in-field provides timeliness while image reviewing at headquarters is used for confirming the validity of the conclusions drawn. There is also, in many cases, a need to improve the quality of the images.

The Emoss and Memobank devices developed by Hymatom could be the core of this new system, with its optimized digital storage which greatly improves image quality.

These devices, which are compatible with existing systems such as MIVS and MUX, have the following advantages:

- comprehensive storage of scenes is comparable to the video tape recording,
- intelligent digital storage facilitates in-field reviewing,
- recoverable disc storage makes it possible to carry out the review at Headquarters,
- printing of the events helps reviewing.

The Emoss and Memobank can be triggered by external information from other systems.

1. Introduction

At the present time, film cameras are being progressively replaced by video cameras. In spite of this, the increased number of optical devices associated with the extension and increased complexity of new nuclear facilities makes reviewing many video tapes an increasingly time consuming task for the inspectors, both on-site and at Headquarters. Also, it has been proved by scientific experiments that the optimum time for carrying out a review with the necessary accuracy is approximately 40 minutes and that, after one hour and a half, reviewing becomes ineffective and the inspectors can then miss important events due to fatigue.

It is therefore urgent to avoid this waste of human resources, especially as the inspectors have other important tasks to perform.

2. Possible recording improvements

The equipment must be capable of storing as many pictures as are now stored on video tape. Information such as the date, time and location of the camera as well as external events such as video detection, detection of radioactivity, detection of the breaking of a seal etc. must be stored in parallel and it must be possible to use them to locate the picture. This system must also be easy to use and the necessary training must not last too long.

The false alarm rate must be sufficiently low to ensure that the effectiveness of the system is not adversely affected.

The linking of the events and the information as described above, as well as the pictures recorded, should considerably reduce inspector reviewing time.

The fact that all pictures would be recorded would also make it possible to carry out a random reviewing of certain scenes on site and to supplement the reviewing scenes with external events. It also makes it possible to cover certain failures (e.g. another camera which records a zone of activity whose field of vision partially overlaps that of the first camera).

Complete sequential recording also offers freedom from any failures of associated systems which trigger recording, for example video detection, nuclear detection, seals etc. Under these conditions, reviewing will be total.

3. Proposed equipment for video reviewing

Recent progress in digital techniques make it possible to envisage a new form of organization for optical containment/surveillance systems. The equipment is available and many hundreds of units have been installed, some for surveillance in nuclear installations.

The Emoss or Memobank devices could form the core of this new system as it offers optimized digital storage and higher picture quality.

The Emoss system, originating from the Memobank, has been developed by Hymatom for the Euratom Safeguards Directorate.

These devices are compatible with the systems currently used by the IAEA to make video recordings, such as MUX and MIVS.

Storage of all the scenes required for the inspectors is comparable to the system used in a video recorder. Intelligent digital storage facilitates on-site reviewing. The removable storage disk makes it possible to transfer data to Headquarters. The printing out of the different items of information or events considerably expedites the location of the picture, and hence the reviewing process.

The system also reacts to external information and can be connected:

- to a video detection system of the Movicom type,
- to another measurement system whose results can be overlaid on the video picture or shown on another display screen, in order to avoid excessively complicating the original picture.

The external events associated with the video recorder considerably reduce the number of pictures that need to be reviewed by the inspectors on site, and possibly at Headquarters.

The equipment is now on the market and can be used as it is. Movicom has been in routine use by Euratom inspectors since the end of 1989, and has proved to be extremely effective and to have an extremely low false alarm rate.

Within the framework of the French Support Programme for IAEA Safeguards, the Memobank with Movicom equipment is planned to be installed at the Cap de La Hague reprocessing plant, possibly in parallel with the IAEA MUX system already in place.

CONCLUSION OF THE I.C.T. BENCHMARK EXERCICE

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ABSTRACT

The I.C.T. Benchmark exercice made within the R.I.V. working group of ESARDA on reprocessing data supplied by COGEMA for 53 routines reprocessing input batches made of 110 irradiated fuel assemblies from K.W.O. Nuclear Power Plant was finally evaluated.

The conclusion are :

- all seven differents I.C.T. methods applied verified the operator data on Plutonium within about one percent,

- anomalies intentionally introduced to the operator data were detected in 90 % of the cases,

- the nature of the introduced anomalies, which were unknown to the participants, was completely resolved for the Safeguards relevant cases,

- the false alarm rate was in a few percent range.

The I.C.T. Benchmark results shows that this technique is capable of detecting and resolving anomalies in the reprocessing input data to the order of a percent.

1 - INTRODUCTION

Dans le cadre du groupe de travail R.I.V. (Reprocessing Input Vérification) de l'ESARDA, le CEA a organisé, à l'aide de deux jeux de données de dissolution de référence un exercice d'intercomparaison de performances des différentes techniques existantes utilisant les corrélations isotopiques pour la vérification des inventaires d'entrées d'une installation de retraitement de combustibles irradiés.

Les résultats des analyses chimiques de dissolution des combustibles retraités à l'usine de LA HAGUE ont été fournis par la COGEMA (composition isotopique de l'Uranium et du Plutonium et rapport Pu/U du combustible dissous présent dans le dissolveur).

Les caractéristiques physiques des combustibles irradiés dans le réacteur électrogène d'OBRIGHEIM ont été fournies par K.W.O. (masse initiale et composition isotopique de l'Uranium avant irradiation et taux de combustion atteint à la date du dernier déchargement).

Le premier jeu de données, noté jeu A, a permis d'évaluer les précisions atteintes lors de l'établissement de l'inventaire d'entrée à l'aide des techniques de corrélation isotopique spécifique à chacun des sept laboratoires participant à l'exercice d'intercomparaison. Les résultats détaillés ont été publiés lors du Symposium ESARDA de Venise.

Le deuxième jeu de données, noté jeu B, devait permettre d'estimer les capacités de chacune des techniques utilisées, à détecter et à diagnostiquer la présence d'anomalies volontairement introduites dans le jeu originel, et dont la présence pouvait avoir un impact variable sur la précision ou la fiabilité de l'inventaire d'entrée établi à l'aide de la méthode dite du "bilan gravimétrique".

Nous présentons dans ce papier les résultats obtenus pour ce jeu B "perturbé" ainsi que les conclusions finales déductibles de la synthèse des résultats obtenus sur l'ensemble de l'exercice dit "ICT-Benchmark".

2 - RAPPELS

Nous présentons dans ce chapitre un ensemble de brefs rappels synthétiques concernant les objectifs visés, les différentes techniques évaluées, et les méthodes d'intercomparaison utilisées.

2.1 - Description des objectifs visés

Le premier objectif visé est l'estimation des précisions atteintes par chaque laboratoire lors de l'établissement, par ses soins, d'un inventaire d'entrée utilisant sa propre technique de corrélation isotopique, appliquée à un jeu de données de dissolution commun.
Pour cet objectif la grandeur la plus significative est la masse de Plutonium obtenue par chaque participant.

(Le même travail a été effectué pour les masses d'Uranium, mais les résultats sur les masses de Plutonium sont plus intéressants à expliciter).

Il a été convenu que les valeurs des masses servant de référence à l'exercice d'intercomparaison sont celles qui ont été obtenues lors du retraitement des combustibles du jeu A par le Service Laboratoire de LA HAGUE, à l'aide de sa méthode d'établissement d'inventaire gravimétrique d'entrée.

Le deuxième objectif visé est d'estimer la capacité et la sensibilité que chaque laboratoire peut déployer dans la détection "d'anomalies" éventuellement présentes à l'intérieur de données de dissolution.

Dans cet esprit, un ensemble d'anomalies types de toute nature a été volontairement glissé à l'intérieur des données de dissolution constituant le jeu B.

Le but essentiel est de détecter, avec le minimum de fausse alarme, le maximum d'anomalies introduites qui induisent des perturbations significatives sur les estimations des masses d'inventaire d'entrée. Les effets engendrés par ces anomalies peuvent être assez différents selon les types de corrélations isotopiques utilisées.

2.2 - Contribution des différents laboratoires

Huit laboratoires ont contribué au bon déroulement de cet exercice d'intercomparaison sur les corrélations isotopiques.

Nous présentons sur le tableau n° 1 les différents laboratoires participant à l'exercice.

Tableau n° 1

N° 1	JAERI	(Japan)
N° 2	MOL	(Belgique)
N° 3	JRC	CEE Karlsruhe
N° 4	CEN-CADARACHE	CEA (France)
N° 5	ARGONNE	DOE (USA)
N° 6	AIEA	AIEA Vienna
N° 7	EURATOM	CEC Safeguards
N° 8	LA HAGUE	COGEMA (France)

Le Service laboratoire de l'usine de LA HAGUE a fourni les données analytiques de référence.

Sept laboratoires ont participé à l'exercice sur le jeu A, et six sur le jeu B.

De plus le CEA s'est chargé de la coordination de l'exercice et du développement des méthodes statistiques nécessaires à son dépouillement.

2.3 - Principes fondamentaux des corrélations isotopiques

Une corrélation isotopique est une fonction liant plusieurs grandeurs caractéristiques d'un combustible irradié (composition isotopique, Pu/U, taux de combustion, etc...) présentant une invariance maximum aux variations de l'historique d'irradiation du combustible (effet de spectre neutronique, temps de refroidissement, puissance spécifique d'irradiation...). Elle permet d'évaluer des grandeurs peu accessibles ou que l'on désire recouper avec des valeurs obtenues par ailleurs.

2.4 - Description des Techniques I.C.T. des différents Laboratoires

Les corrélations isotopiques se caractérisent principalement par le choix des fonctions de corrélation utilisées (p. ex. (Pu/U)/(DU235)) et par le choix de la banque de données de génération de ses coefficients de corrélations (banque d'origine empirique ou théorique).

Il existe, en effet, deux grandes familles de techniques de corrélations isotopiques.

La famille des corrélations isotopiques empiriques, qui recherche à l'aide d'outils statistiques des corrélations fortes dans un ensemble de données toutes d'origines expérimentales, et regroupées en banque de données plus ou moins hétérogènes.

Ces banques sont constituées de résultats d'analyse chimique sur différents combustibles irradiés retraités.

L'efficacité de ces corrélations est très fortement liée à la qualité et à l'ampleur du domaine couvert par sa banque de données analytiques.

La famille des corrélations isotopiques calculées, où la sélection des fonctions de corrélation a été effectuée en tenant compte des lois d'évolution du combustible sous irradiation.

L'optimalisation de l'invariance de ces fonctions a été effectuée, à l'aide des codes de calculs neutroniques en usage dans le domaine de la physique des réacteurs nucléaires.

Les banques de données utilisées pour générer les coefficients de corrélations ont été créés spécifiquement par type de combustible différents et sont en général très bien qualifiées dans le domaine usuel de conditions de combustion du combustible dans les réacteurs électrogènes de puissance.

La bonne qualité de ces corrélations réside dans la qualification des outils de calcul neutronique utilisés.

2.5 - Description des Données Communes Utilisées

L'exercice a porté sur un total de 110 assemblages de combustible irradiés déchargés du réacteur d'OBRIGHEIM et retraités par la COGEMA en son usine de LA HAGUE.

Le jeu A, intégralement d'origine, se compose de 24 dissolutions effectuées à l'usine de LA HAGUE en 1981.

Le combustible a été déchargé en 1977 et en 1978 du réacteur d'OBRIGHEIM, après avoir atteint des taux de combustion compris entre 25000 et 35000 MWj/tm (0).

Le jeu B, intentionnellement perturbé, se compose de 29 disso-lutions effectuées à l'usine de LA HAGUE en 1980.

Le combustible a été déchargé en 1974 et en 1975 du réacteur d'OBRIGHEIM, après avoir atteint des taux de combustion de même ordre que pour le jeu A.

Tableau n° 2
DESCRIPTION OF ANOMALIES INTRODUCED IN SET B

(11 batches have been changed)

N°	Type of modification	Original data of dissolution
3b	1 assembly has been exchanged	n° 215 instead of n° 196 => modify <Er> of batch
6b	Isotopic analysis of U	0,018 1.008 % 234U 235 U 0,0386 98,588 236 U 238U
8b	1 assembly has been exchanged	n° 196 instead of n° 215 => modify <Er> of batch
12b	Isotopic analysis of Pu	1.340 61.223 23,215 239Pu 10,064 1,158 % 242Pu
14b	Burn-up of 1 asembly modified Pu/U measured	n° 159 B.U. declared = Pu/U measured = 8,372e-3
17b	Isotopic analysis of U	0,017 1,051 % 235U 0,376 98,556 238U
18b	Isotopic analysis of Pu	1,454 60,646 238Pu 239Pu 23,492 10,106 240Pu 241Pu 4,302 242Pu
21b	1 assembly has been exchanged	n° 143 instead of n° 313 => modify <C.T., B.U., Er>
26b	Burn-up of 2 assemblies modif.	n° 278 B.U. declared = n° 279 B.U. declared =
27b	Isotopic analysis of Pu Pu/U	1,300 60,412 23,089 10,925 4,274 % 8.31e-03

2.6 - Description des Méthodes Statistiques d'Intercomparaison

Le modèle utilisé, appelé Grubb-Neuilly, a été décrit en détail lors de la présentation des résultats obtenus pour le jeu A [1].

De plus un article spécifique a été publié dans le bulletin de l'ESARDA [2].

La caractéristique principale de ce modèle d'analyse statistique, plus général que le modèle de Grubb, est de permettre la séparation de la partie indépendante de l'erreur aléatoire de la partie corrélée, lors de l'estimation de la variance de chaque laboratoire.

3 - RESULTATS OBTENUS

Après un bref rappel des conclusions déduites précédemment, examinons les résultats obtenus globalement sur les deux jeux A et B pour chacun des objectifs visés.

3.1 - Description des anomalies intentionnelles

Les altérations des données originales ont été choisies de manière à simuler, aussi bien par leur nature, que par leur intensité, les diverses erreurs pouvant éventuellement et naturellement se glisser dans des données de dissolutions.

Le tableau n° 2 présente en détail les caractéristiques des anomalies introduites. Celles-ci concernent onze dissolutions du jeu B.

Elles consistent en particulier en des interversions d'assemblages de combustibles irradiés ayant des caractéristiques différentes, par exemple pour les dissolutions 3b, 8b, 14b et 21b, (taux de combustion, date de déchargement, teneur isotopique en Uranium initial historique d'irradiation), ou en des modifications des analyses des compositions isotopiques de l'Uranium ou/et du Plutonium par spectrométrie de masse, par exemple pour les dissolutions 6b, 12b, 17b et 18b, ou de modification des résultats de double dilution isotopique du Pu/U, pour les dissolutions 14b et 27b.

NB. :

- Tous les assemblages ont une teneur initiale en 236U (entre 150 et 250 ppm). Cette donnée, qui n'a pas été donnée par le fabricant, n'est pas une anomalie introduite. Cet effet a été détecté par plusieurs laboratoires et corrigé.

- L'assemblage n° 139 du lot n° 15b était manquant dans les données de dissolution distribuées.

Les anomalies introduites peuvent se classer en deux catégories assez distinctes :

- les anomalies qui ont une influence sensible et directe sur des grandeurs significatives au sens du Contrôle des Garanties, comme la masse de Plutonium par exemple. Huit anomalies sur les onze introduites appartiennent à cette catégorie (3, 6, 8, 14, 17, 21, 27 et 29) (nous noterons par la suite ce type d'anomalie : "significative"),

- les anomalies qui n'ont qu'une influence sur les contrôles de cohérence interne spécifique à chaque technique de corrélation isotopique et/ou qui ne sont pas significatives pour le Contrôle des Garanties. Trois anomalies appartiennent à cette catégorie (12, 18 et 26) (nous noterons ce type d'anomalie : "non significative").

Dans le tableau suivant n° 3, nous présentons l'évaluation quantitative approximative des effets induits sur les masses d'Uranium et de Plutonium de l'inventaire d'entrée par les diverses anomalies qui ont été intentionnellement introduites. (Seules significatives dans le cadre du contrôle de garanties).

	Table n° 4			Table n° 5			Table n° 6		
Laboratories	Set A 24 dissol.			Set B 18 dissol.			Set B 26 dissol.		
JAERI	0.8	0.9	1.2	0.4	0.8	0.9	0.6	0.8	1.0
MOL	1.6	0.9	1.8	1.2	0.7	1.4	1.3	0.7	1.5
JRC	1.5	1.1	1.8	0.7	0.6	0.9	0.7	0.8	1.1
CADARACHE	0.3	0.6	0.7	0.0	0.9	0.9	0.2	1.2	1.2
ARGONNE	0.9	0.6	1.1	0.7	1.5	1.6	1.1	0.2	1.1
IAEA [*]	1.3	0.9	1.6	0.0	1.7	1.7	0.8	1.7	1.9
EURATOM	1.5	1.2	1.9						
Kind of error	c	r	T	c	r	T	c	r	T
c = correlated r = random T = total									

(*) AIEA T = 1.2 (reevaluation of results)

Intervalle de Confiance de 95 % par loi de Xhi2 contenant la valeur VRAIE de la VARIANCE concernant la masse de PU donne par les laboratoires				
	Table n° 7		Table n° 8	
Laboratories	Set A 24 dissol.		Set B 26 dissol.	
JAERI	0.93	1.68	0.78	1.38
MOL	1.40	2.52	1.18	2.07
JRC	1.40	2.52	0.86	1.52
CADARACHE	0.54	0.98	0.94	1.66
ARGONNE	0.85	1.54	0.86	1.52
IAEA	1.24	2.24	0.94	1.66
EURATOM	1.48	2.66		
Confidence interval of 95 % calculated bv Xhi2				

Tableau n° 3

Set B No of batch	Fissile material involved	Variations of masses in g.
03	U235 (0)	720.
06	U235 final	300.
08	U235 (0)	720.
12	no effect	*
14	Plutonium	120.
17	U235 final	270.
18	no effect	*
21	U235 (0)	240.
	+ Pu 241	30.
26	no effect	*
27	Plutonium	170.
29	U235 (0)	240.
	+ Pu241	40.

(*) Internal consistensy check only.
(+) Double effect on U <u>and</u> Pu masses.

3.2 - Performances des estimations des masses de Plutonium

Il a été convenu dès le début de l'exercice, d'examiner l'ensemble des résultats obtenus par tous les participants, par rapport aux résultats obtenus par le laboratoire de LA HAGUE pris comme référence (Lab. n° 8 tableau 1).

L'estimation de la variance, des erreurs aléatoires de chaque laboratoire (n° 1 à 7), à l'aide de la méthode statistique Grubb-Neuilly, a été calculée pour les deux jeux A et B.

Nous présentons ces estimations de variance dans les tableaux n° 4, pour les 24 dissolutions du jeu A et, dans le tableau n° 5 pour les 18 dissolutions non perturbées intentionnellement du jeu B.

Nous présentons dans le tableau n° 6 les résultats pour les 26 dissolutions du jeu B tel qu'il a été fourni.

Les dissolutions 3, 8 et 21 ont été enlevées, car certains laboratoires, n'ont pu fournir de résultats, faute d'avoir alors, une banque de données suffisamment étendue.

Cette présentation explicite la décomposition des variances estimées en leur partie purement aléatoire et leur partie corrélée.

A l'examen des tableaux 4 et 5, on peut observer la sensible diminution de la partie corrélée de l'erreur estimée de la variance, entre le jeu A et le jeu B.

Lors de l'analyse des résultats du jeu A on avait attribué l'origine de l'importance de la partie corrélée de l'erreur estimée sur la variance (pour certains laboratoires), à l'utilisation d'une ou plusieurs fonctions de corrélation présentant une particulière sensibilité à des perturbations spécifiques ayant affecté certains des assemblages lors de leur combustion, ainsi qu'à l'utilisation de banques de données empiriques présentant pour certaines caractéristiques physiques des combustibles la composant de trop fortes hétérogénéités.

Ces perturbations, affectant quelques assemblages composant le jeu A, ont été dues à des séjours épisodiques de ces assemblages combustible, lors de leurs chargements dans le coeur du réacteur, dans un voisinage de combustible de type UO2-PuO2 fortement perturbant sur le plan du spectre neutronique pour certaines fonctions de corrélations. Ces résultats été confirmés par des calculs de neutronique.

Ainsi, nous pouvons constater à l'examen des résultats obtenus sur le jeu B une diminution significative de la partie corrélée de l'erreur estimée sur la variance.

Il semblerait que les conclusions, dégagées lors de l'analyse des résultats du jeu A, aient permis l'amélioration des banques de données empiriques utilisées, en extrayant les données singulières ou en optimalisant le jeu de fonctions de corrélations utilisées.

Nous présentons dans les tableaux n° 7 et 8, les intervalles de confiance contenant pour 95 % des cas la valeur vraie de la variance.

	Table n° 8		Table n° 9		
Laboratories	TRUE ALARMS		FALSE ALARMS		
JAERI	7	3	0	0	0
MOL	6	1	5	1	4
JRC	8	3	1	1	0
CADARACHE	8	3	2	1	1
ARGONNE	7	3	2	1	1
IAEA	7	0	0	0	0
EURATOM	....		....		
nb.& kind of anomalies introduced	8 s	3 ns	0	0 f	0
Nb. batches	29		29	1	28
s = safeguards significant f = first dissolution batch ns = internal consistency check					
Conclusion MEAN 7.2 True TOTAL 6 False on ALARMS detection => 90 % of anomalies with 3 % of false alarm					

	Table n° 10	Table n° 11	Table n° 12
Laboratories	Batch n° 12	Batch n° 18	Batch n° 26
JAERI	Error on Pu242	Isotopic of Pu	No detection
MOL	....	No detection	No detection
JRC	Isotopic of Pu	Isotopic of Pu	Burn-up
CADARACHE	Isotopic of Pu or Cooling time	Isotopic of Pu or Pu/U	Burn-up or U236 (0)
ARGONNE	Pu242/Pu241	Pu analysis	U235 or Pu/U
IAEA	No detection	No detection	No detection
REAL ANOMALIE introduced in dissol. data	242Pu/239Pu Change 18 g of Pu239 into 18 g of Pu241	Change Plutonium Isotopic Composition	Change Predicted Burn up of fuel bundles
Total diagno.	4 <1>	4 <2>	3 <3>
TRUE	4	4	2
FALSE	0	0	1

<1> 1 not answer and 1 not detected
<2> 2 not detected
<3> 3 not detected

Il est certain qu'à ce stade, l'ensemble des laboratoires, participant a la totalité de l'exercice, atteint une variance globale pour le Plutonium de l'ORDRE DU POUR CENT.

3.3 - Performances des détections d'anomalies introduites

Nous présentons dans les tableaux n° 8 et 9, les taux des vraies et fausses alarmes comptabilisées lors de l'analyse des résultats fournis par les sept laboratoires participants.

Pour chaque laboratoire nous indiquons le nombre d'alarmes vraies, (c'est-à-dire concernant une dissolution où une anomalie a bien été intentionnellement glissée), et le taux de fausses alarmes, (c'est-à-dire concernant des dissolutions sur lesquelles aucune modification de données ou de résultats n'a été effectuée).

On considère donc, par définition, qu'il n'existe pas d'anomalie dans les données examinées hormis celles qui y ont été délibérément introduites.

Nous avons décomposé les anomalies dans les deux catégories explicitées précédemment.

Il existe huit anomalies de type "Contrôle des Garanties" et trois de type "contrôle de cohérence interne".

On considère que la convergence des résultats de la majorité des laboratoires vers une détection d'anomalie dans les données de dissolution du batch n° 1, ajouté à la particularité de ce dernier d'être le premier du lot, nous permet d'envisager l'existence d'une perturbation réelle en son sein, qui pourrait fausser nos calculs statistiques.

Ainsi, d'un commun accord, nous avons retiré cette dissolution de l'ensemble examiné pour les calculs du taux de fausses alarmes.

En conclusion on note que 90 % des anomalies ont été détectées par 80 % des participants avec un taux de fausses alarmes inférieur a quatre pour cent.

De plus toutes les anomalies "significatives" ont été détectées. Enfin, 70 % des anomalies "non significatives" ont été détectées par 80 % des participants.

3.4 - Performances des diagnostics d'anomalies de contrôle de cohérence

Pour les anomalies de type "significatif", il apparaît de l'analyse des résultats que, si une détection d'anomalie intervient de la part d'un laboratoire, et que celle-ci est fondée (vraie alarme), son diagnostic a toujours été correct.

Il est important de noter qu'un diagnostic a été considéré comme correct à partir du moment où la source à l'origine de l'anomalie fait bien partie des sources putatives explicitées par les participants.

En ce qui concerne les anomalies de type "non significatives", leur nombre restreint (3) ne permet pas une synthèse statistique des résultats obtenus.

Ainsi nous présentons dans les tableaux n° 10, 11 et 12, les résultats détaillés obtenus par tous les participants sur les dissolutions n° 12, 18 et 26, qui contiennent ce type d'anomalie.

Il est intéressant de noter les alternatives liées, proposées par certains, comme sources possibles de l'anomalie.

En effet, il apparaît assez difficile de dissocier une erreur légère sur la teneur en Pu241, qui se serait produite lors de la spectrométrie de masse, d'une inexactitude sur l'évaluation du temps de refroidissement du combustible (portant essentiellement sur le Pu241).

4 - CONCLUSIONS

Malgré l'existence de différentes approches de la technique des corrélations isotopiques, qui peut être aussi bien entièrement empirique ou faire appel plus ou moins fortement à la physique des réacteurs, il est remarquable de noter la COHERENCE des résultats obtenus par les SEPT laboratoires participant à cet exercice d'intercomparaison.

De plus les données de dissolution utilisées comme jeux de référence pour cet exercice proviennent d'une installation de retraitement industriel (UP-2 à LA HAGUE) et ont été acquises dans un contexte d'exploitation de production normale.

Les deux objectifs fixés, la vérification de l'inventaire d'entrée, ainsi que la détection et le diagnostic des anomalies volontairement introduites, ont été atteints.

En effet, l'écart global obtenu entre l'inventaire d'entrée en Plutonium obtenu avec les données de l'exploitant de l'usine UP-2, et les inventaires en masse du Pu calculées par les sept différentes méthodes des participants à l'exercice est à peu près de UN pour CENT.

Les anomalies qui ont été introduites intentionnellement ont un ordre de grandeur de quelque pour cent.

Nous avons noté que celles-ci ont été détectées dans 90 % des cas.

De plus, l'ensemble des participants a réussi à les détecter dans la même proportion.

Les anomalies de nature à avoir, soit par leur spécificité, soit par leur intensité, un effet significatif au sens du contrôle de garanties sur la détermination de masses de matières fissiles, ont été détectées dans 90 % des cas avec un taux de fausses alarmes d'environ un pour cent.

D'autre part, les anomalies non significatives sur le plan des matières fissiles mais présentant un intérêt pour contrôler la bonne cohérence des données de dissolution utilisées, ont été détectées dans environ 70 % des cas avec un taux de fausse alarme de quelques pour cent.

Enfin, les diagnostics effectués sur les alarmes réelles ont été corrects dans 99 % des cas.

5 - PERSPECTIVES

A l'examen de ces résultats, il apparaît techniquement possible d'envisager l'utilisation des techniques de corrélations isotopiques, dans le cadre de la vérification d'inventaires d'entrée d'installations de retraitement de combustibles irradiés, dans trois secteurs d'application suivants :

1) Les autorités chargées du Contrôle de Garanties (EURATOM Safeguards ou/et IAEA) sont amenées à remesurer des échantillons de solution de dissolution. Dans le cas éventuel ou une différence puisse apparaître entre les résultats des exploitants de l'installation et l'organisme chargé des vérifications, plutôt que de se livrer à des répétitions de mesures, il pourrait être judicieux d'utiliser les résultats obtenus grâce à la technique des corrélations isotopiques.

En effet, celles-ci permettraient soit de remonter à la source de la différence, soit de servir éventuellement de "umpire détermination".

De fait, les corrélations isotopiques sont fortement liées à des grandeurs mesurées et contrôlables comme les teneurs initiales en U235, les masses initiales de combustible, les taux de combustions, etc...

2) Certains types d'anomalies ne sont pas actuellement directement détectables par des mesures de type destructives effectuées sur le combustible irradié dissous.

En particulier, les interversions d'assemblages, entraînant un impact sur les temps de refroidissement présumés du combustible à dissoudre, pourraient être détectées par les corrélations isotopiques.

3) Enfin, l'application de la technique des corrélations isotopiques sur les combustibles AVANT dissolution pourraient permettre de détecter préventivement certains types d'erreurs comme les interversions d'assemblages combustibles.

Ces vérifications, nommées précampagnes, porteraient uniquement sur les données fournies avec les assemblages de combustibles à retraiter (en particulier leur composition isotopique estimée).

CONCLUSIONS OF THE ICT BENCHMARK EXERCISE

Despite the different approachies of ICT (empirical sometimes supplemented by reactor physics). The results of its application are consistent. The ICT methodes were applied to operational data of two reprocessing campaign of UP-2 plant at LA HAGUE.

The method was implemented with two objectives : to verify the material balance at the input point and to detect and explain anomalies, which were intentionally introduced.

The overall agreement for the plutonium material balance between the operators data and the seven methods applied by the participants was about one percent (see table 1).

This agreement was found to be consistent for the set A data, without intentional anomalies and set B with intentional anomalies. The intentional anomalies introduced were in the range of a few percent and were uncovered in 90 % of the cases, and the causes were identified by all of the techniques as shown in table 2. The analysis demonstrated that the intentional anomalies were detected with a 90 % probability and that the source of the anomalies were established.

The anomalies introduced were of two types. In the first type, the magnitude of the anomalies affected the material input balance, whereas in the second type there was no effect on the balance. The results showed that the anomalies of the firts type were detected in 90 % of the cases, and that the anomalies of the second type were found in about 70 % the cases. Referring to table 3. The range of the false alarm rate was found to be within a few percent. The interpretation of the true alarms were correctly identified in about 99 % of the cases.

There are three possible applications to present practice of reprocessing input verification :

1) EURATOM Safeguards and IAEA remeasure samples of the reprocessing input. In case differences between the operators analyses and the Safeguards' laboratory are in the percent range which cannot be resolved by a repetition of the measurement. ICT could be used to detect a possible cause of the difference. Since ICT baes partially on historical data (initial enrichment, fuel weight, burn up, etc...) it could act as an umpire determination.

2) As shown above ICT has the potential to discover errors not to be seen from a direct (destructive) analysis of the dissolved spent fuel such as exchange of fuel assemblies, misreadings of cooling time, fuel weight, burn up etc...

Therefore a check of operator and Safeguard laboratory data by ICT could reveal inconsistencies which otherwise might be not discovered. However this again applies to deviations about the percent range.

3) Since ICT can be applied to the shipper data (when isotopic data are supplied) prior to fuel dissolution, a so called precampaign analysis could focus the verification analysis to the detection of the anomalies:

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THE ROLE OF NDA IN SAFEGUARDING A PILOT CONDITIONING PLANT

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Abstract

The pilot conditioning plant (PKA) planned in Gorleben will, among other tasks, demonstrate that technical facilities for the disassembly and packing of spent fuel for final disposal have reached a standard suitable for industrial-scale operation. As a multi-purpose plant of a pilot nature, it is also intended for the optimization of safeguards measures.

On the way of a fuel assembly from the reactor via long-term intermediate storage to the conditioning plant and eventually to a geological repository, the last possibility of a direct measurement of the nuclear material exists in the conditioning plant. This fact leads to the assumption that a measurement of the final disposal package content should be considered to represent the input measurement of the final repository. The nature of the conditioning process permits a non-destructive assay only. Such a measurement should be performed on bins or canisters which contain the fuel rods of several disassembled fuel assemblies without their structural parts. Preliminary investigations show that passive neutron counting in combination with a gamma measurement is promising.

Apart from the high demands made on a measurement system, complementary C/S methods are to be selected which permit a correlation of conditioned fuel rods with the supplied fuel assemblies.

1. Introduction

In January 1990, a first partial license was granted for the construction of a pilot conditioning facilitiy for spent nuclear fuel and different types of waste at Gorleben. The site has been prepared and construction work has begun recently.

With respect to safeguards, suitable measures have been discussed among experts for several years with a view to the peculiar problems raised by such a facility. Any safeguards approach suggested will have to fit into overall safeguards considerations, beginning at the reactor and ending with the direct disposal of spent fuel /1/.

The multi-purpose nature of the facility may complicate the definition of an effective safeguards concept because of the variety of different operations. On the other hand, this will also offer an opportunity to find a practicable safeguards solution for an industrial scale conditioning facility by conducting tests with selected and adjusted safeguards equipment /2/.

For spent fuel assemblies shipped to and entering the conditioning facilitiy, item accounting may be applied as a relatively simple and adequate safeguards technique. This situation will, however, change once the integrity as well as the identity of the fuel assemblies are destroyed for the purpose of conditioning, and new items are formed suitable for final disposal.

As the nuclear material content of a final disposal cask will be newly defined in the loading cell of the conditioning facility, it seems reasonable to verify the amount of nuclear material, which has thus far only been calculated, at this point by a measurement. Of course, taking into account the nature of the conditioning process, this will have to be a non-destructive assay (NDA) measurement.

The measurement of the final storage package content does not only constitute the actual output measurement for the conditioning facility. Due to the inaccessibility of the material after closing the package, it represents also the input measurement of subsequent facilities, i.e. the final repository.

After the disassembly of fuel elements the fuel rods are either put into bins as a whole, or are chopped to pieces and put into canisters. Just before the filled bins and canisters are inserted into final disposal containers, they have to pass through the NDA measurement position. Using complementary C/S, it can be assured that all nuclear material leaving the facility is recorded quantitatively.

2. The Role of NDA

As can be inferred from the layout of the facility and its mode of operation, the primary purpose of the PKA is the conditioning of high active material, only to be handled under heavy shielding. The safety design of the facility allows to dispense with measurements of the fissile material altogether. This is also true for process control. Thus, operational measurement, or sampling, locations are not planned.

Strictly speaking, safeguards requirements are met once suitable methods have established that all material which has entered the PKA for the purpose of conditioning has fully left the facility, even if in a different shape.

In a geological repository, fuel assemblies are, according to the German reference approach, emplaced in tunnels in hermetically sealed, heavily shielded packages of the 'POLLUX' type. Knowledge of the current inventory of the final repository will certainly be required, but the relevant input measurement would present major technical problems, as a meaningful quantative measurement of consolidated fuel assemblies through heavy shielding will not be satisfactory, according to current experience. The simplest, and also the most sensible, solution would be to carry out the measurement required in the place where the material subject to safeguards is loaded into the final disposal package. This opportunity will present itself for the last time in the conditioning facility. By providing and operating such a measurement point, the PKA is, in fact, carrying out the input measurement for the final repository. It thus assumes a vital safeguards function for the subsequent facility.

The data calculated by the reactor operator will form the basis of material accountancy; these will not have been changed during interim storage in an away-from-reactor store. If a new item 'final disposal package' is formed, it will not make sense to retain the old shipper data, which has most probably changed during long periods of storage. The only reliable method will be to bring the figures up to date by way of calculation. Thus, NDA is automatically assigned the task of verifying the fissile material, determined by calculation, of the fuel rods consolidated in canisters immediately prior to them being put into shielding. Verification measurements do not automatically satisfy the high standards of an accountancy measurement. The achievable accuracies estimated for NDA at this point will entirely suffice to meet safeguards requirements.

NDA equipment will serve safeguards purposes at a number of locations within the PKA. Such equipment will probably be combined with camera systems to follow container movements and to allocate accountancy batches. In this case, NDA will be used to carry out C/S functions, which will not be alluded to here.

3. The NDA Measurement Techniques

There are several possibilities for a practicable NDA measurement, and the most promising has to be selected considering the form of nuclear material, interference with facility operation, reliability, robustness, authentication, etc. In principle, active and passive neutron detection or gamma measurement systems appear to be adequate for this task. Experience shows that only simple and compact measurement systems can be handled in the hot cell area. For this reason the application of an active measurement method appears questionable. The advantages and deficiencies of the different measurement techniques will be discussed in detail.

In the past, experience has been gained with characterizing spent fuel elements by NDA measurement under water in the spent fuel pond. The measurement of spent fuel elements or configurations of fuel pins in dry conditions present new application fields. For this case it has to be checked which boundary conditions are relevant and these have to be considered in selecting an adequate measurement method. A feasibility study on this subject has been carried out by an industrial company.

The basic requirements for this task are:

- The method should provide a high transparency, i.e. it has to comprise and penetrate the major part of the spent fuel configuration.
- The measurement result should characterize the spent fuel configuration.
- The measurement should allow a quantitative determination of the nuclear material content.

The applicability of a gamma measurement to a dense configuration of fuel pins has been studied. After a cooling time of several years the most prominent lines in the gamma spectrum originate from the decay products Cs-134, Cs-137, Ce-144 and the activation product Co-60. The gamma radiation is strongly absorbed by the surrounding fuel pins. The consequence of this effect is that only the outer parts or layers of the fuel pins contribute to the detectable gamma signal. Therefore the transparency of the gamma measurement is very limited and this measurement method alone is not adequate for a quantitative determination of the nuclear material content.

The application of an active neutron interrogation method faces similar problems. The interrogating neutrons have to be thermalized for generating induced fissions. These neutrons are absorbed in the outer layers of the fuel pins. Thus the multiplication by thermal neutrons is constant beyond a certain thickness of the fuel pin configuration and therefore independent of the nuclear material content. For this reason the active neutron measurement is not capable of solving the task.

The passive neutron detection method has been identified showing the highest transparency of all analyzed measurement techniques considered here. The neutrons are mainly originating from the spontaneous fissions of Cm-244 and the even Pu isotopes Pu-238, Pu-240, Pu-242. They are emitted with high energy and therefore penetrate the fuel pin configuration. The detection probability for neutrons produced in the centre of the fuel pin package is about half of that of neutrons produced in the outer layers. Nevertheless, the passive neutron measurement appears to be the most promising method to measure spent nuclear fuel pins packed in bins or canisters for final storage.

4. Determination of the Nuclear Material Content

The evaluation of passive neutron and passive gamma measurement data requires additional information on the enrichment, burn-up and cooling time of fuel elements to estimate the amount of nuclear material quantitatively. These data are normally provided by the reactor operators on the basis of calculations. They may be confirmed by additional NDA measurements of each individual fuel element before disassembly. This could be done by a simple passive neutron and/or gamma detector device. It should, however, be mentioned that NDA of spent fuel in dry conditions is not state of the art.

There is hence a need to correlate data generated by NDA of the consolidated rods of several assemblies in one bin or canister of the final disposal package. Here, a combination of C/S measures with the NDA systems under consideration will contribute significantly to the effectiveness of the safeguards system in the pilot conditioning plant.

5. Conclusions

Is has been recognized that an NDA measurement of spent fuel in a pilot conditioning plant may represent an important component in a safeguards regime for the direct disposal of LWR spent fuel assemblies. The applicability of different measurement techniques, such as gamma measurement, active and passive neutron detection has been studied. The most promising measurement method appears to be the passive neutron detection, possibly supported by gamma measurement. In any case, additional information on enrichment, burn-up and cooling time has to be provided to estimate the nuclear material content quantitatively. Considerable development work will have to be carried out to confirm the theoretical considerations and to assure that a practicable measurement device will be available in time.

6. References

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ANWENDUNG VON PASE ZUR ANALYSE VON ABZWEIGUNGSPFADEN BEI DER DIREKTEN ENDLAGERUNG AUSGEDIENTER BRENNELEMENTE

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Abstract

Direct final disposal of spent nuclear fuel in permanent geological repositories is a novel problem for international safeguards.

In order to allow for the development of an effective safeguards system and to evaluate alternative safeguards approaches, a comprehensive analysis of possible diversion paths is required together with an assessment to what extent the relevant paths can be covered by the safeguards measures under consideration.

The PASE (Probabilistic Analysis of Safeguards Effectiveness) method is currently being developed and investigated in the IAEA to provide a standard procedure to conduct diversion path analysis. Initial experience using PASE to analyze diversion paths for the presently planned Gorleben repository is presented.

Einleitung

In der Bundesrepublik Deutschland ist vorgesehen, ausgediente Brennelemente aus Kernkraftwerken, auf deren Wiederaufarbeitung definitiv verzichtet wird, in einem Salzstock endzulagern. Die Direkte Endlagerung von ausgedienten Brennelementen stellt neuartige Anforderungen an die internationale Kernmaterialüberwachung und erfordert, da hierfür noch kein Modell-Verfahren existiert, die Entwicklung eines neuen Safeguardssystems.

Entsprechend ihrer Safeguardszielsetzung geht die IAEO bei der Entwicklung von Safeguardssystemen von der Arbeitshypothese aus, daß eine gewisse, wenn auch geringe, Wahrscheinlichkeit dafür besteht, daß vom Betreiberstaat einer kerntechnischen Anlage ein Abzweigungs- oder Mißbrauchsversuch unternommen wird. Eine Voraussetzung für den Entwurf eines Safeguardssystems für eine kerntechnische Anlage ist daher eine systematische Analyse möglicher Abzweigungspfade in dieser Anlage.

Die IAEO versucht seit einer Reihe von Jahren, die Vorgehensweise bei der Analyse von Abzweigungspfaden und bei der Bewertung alternativer Safeguardskonzepte in der Entwurfsphase zu standardisieren und zu formalisieren. Derzeit wird hierfür das PASE-Verfahren (Probabilistic Analysis of Safeguards Effectivness) erprobt.

Methodik des PASE-Verfahrens

PASE stellt den Versuch dar, Arbeitsmethoden, die sich bei der probabilistischen Sicherheitsanalyse bewährt haben, analog auf Sicherungsprobleme zu übertragen. Im Jahr 1986 wurde in der IAEO eine Arbeitsgruppe eingerichtet, die mit Unterstützung externer Experten die Anwendbarkeit solcher Methoden für Safeguards untersuchte. Als Ergebnis dieser Arbeiten wurden ein Verfahren und eine Reihe von Computerprogrammen entwickelt, die als "PASE" bezeichnet werden.

Bei PASE wird versucht, den methodischen Ansatz der Fehlerbaumanalyse auf die Analyse auf Abzweigungszenarios anzuwenden. Die Fehlerbaumanalyse ist eine deduktive Methode, die für Zuverlässigkeits- und Sicherheitsbetrachtungen von technischen Systemen angewendet wird. Dabei wird ein unerwünschter Systemzustand (TOP-Ereignis) vorgegeben. Die Fragestellung lautet dann: "Welche Ursachen können zu diesem unerwünschten Systemzustand führen?". Die Ursachen werden schrittweise durch fortgesetzte Detaillierung der deduktiven Fragestellung weiter aufgeschlüsselt. Dabei werden alle Kombinationen von Komponenten- oder Teilsystemausfällen, die zu dem vorgegebenen unerwünschten Ereignis führen können, und ihre logischen Verknüpfungen zusammengestellt. Diese Zusammenstellung ergibt den sogenannten Fehlerbaum.

Voraussetzung für die Erstellung eines Fehlerbaumes ist die genaue Kenntnis des zu beurteilendes Systems. Vor der Durchführung einer Fehlerbaumanalyse muß das System qualitativ so analysiert werden, daß neben dem Verständnis des Verfahrensablaufes detaillierte Kenntnisse über die Funktionsweise der Komponenten- und Teilsysteme gewonnen werden.

PASE übernimmt den Ansatz und die logische Strukturdarstellung des Fehlerbaumverfahrens. Hier ist der unerwünschte Systemzustand (das TOP-Ereignis) die Abzweigung von Kernmaterial aus der betrachteten Anlage. Durch schrittweise Detaillierung der Fragestellung: "Wie kann Kernmaterial aus der Anlage abgezweigt werden?" sollen alle denkbaren Abzweigungsmöglichkeiten systematisch ermittelt werden.

Dabei gelten für PASE die gleichen Voraussetzungen wie auch für die Erstellung von Fehlerbäumen. Grundlage ist zunächst eine genaue Analyse der Anlage. Sie sollte die räumliche und technische Auslegung der Anlage, Art und Form des Materials, daß in der Anlage gehandhabt wird, die Inventare und Bewegungen des Materials und die Prozesse, die am Material ausgeführt werden, umfassen.

PASE bietet, ebenso wie die Fehlerbaumanalyse, zunächst eine strukturierte und systematische Vorgehensweise. Durch die schrittweise Detaillierung soll sichergestellt werden, daß keine relevanten Abzweigungspfade übersehen werden. Ein eindeutiger Nachweis, daß alle Abzweigungspfade vollständig erfaßt sind, ist hiermit jedoch nicht möglich.

Ein wesentlicher Unterschied zwischen der Fehlerbaumanalyse und PASE besteht darin, daß das Objekt der Analyse nicht ein technisches System, sondern der Handlungsspielraum eines potentiellen Abzweigers ist. Ein technisches System hat in der Regel fest vorgegebene Komponenten, Struktur und Grenzen. Dies trifft auf den Handlungsspielraum eines Staates, dem man Mißbrauchsabsicht unterstellt, nur in eingeschränkter Weise zu. Damit müssen für als Voraussetzung für die Anwendung von PASE neben den technischen Fakten auch ein Rahmen für die zu betrachtenden Abzweigungsszenarien vorgegeben werden.

PASE ist als 2-stufiges Verfahren ausgelegt. In der ersten Stufe werden anhand der logischen Strukturdarstellung die möglichen Abzweigungspfade identifiziert. In einer zweiten Stufe wird betrachtet, wie ein gegebenes Safeguardssystem auf einen vorgegebenen Abzweigungspfad reagiert. Dabei wird untersucht, welche Ereignisse eintreten bzw. welche Komponenten des Safeguardssystems versagen müssen, damit eine Abzweigung über diesen Pfad unentdeckt bleibt. Diese zweite Stufe entspricht dadurch wesentlich eher einer klassischen Anwendung der Fehlerbaumanalyse.

Erstellung der Aktionsbäume

Dem unerwünschten Systemzustand im Fehlerbaum entspricht bei PASE die durchgeführte Abzweigung. Den möglichen Fehlerursachen entsprechen die möglichen Aktionen des Abzweigers. Der Fehlerbaum wird bei PASE damit zu einem Aktionsbaum.

Die Elemente des Baumes sind alle Aktionen, die ein potentieller Abzweiger zur physischen Entwendung des Materials, zur Manipulation seiner Buchhaltung, zur Manipulation von Überwachungseinrichtungen und zur sonstigen Verschleierung einer Abzweigung durchführen könnte. Die einzelnen Elemente werden durch binäre logische Funktionen (UND- oder ODER-Verknüpfungen) miteinander verbunden. Der Entwurf des Baumes erfolgt dabei von oben nach unten (top down). Diese Beschreibung der Abzweigungsmöglichkeiten soll ohne Berücksichtigung dessen, ob sie erfolgreich gangbar wären oder nicht und unabhängig von einem konkreten Safeguardskonzept für die Anlage erfolgen.

Da die Erfassung aller möglichen Abzweigungsalternativen in einem Aktionsbaum zu aufwendig ist, wird die unterstellte Abzweigung in ein grobes Ablaufschema unterteilt, deren einzelne Stufen isoliert betrachtet werden können. Solche Stufen können z. B. für eine Abzweigung aus dem Endlager sein:

- physische Entwendung von Endlagergebinden oder Material aus Endlagergebinden
- Rücktransport des Materials nach über Tage oder aus der Anlage hinaus
- Substitution von entwendeten Endlagergebinden
- Manipulation von C/S-Geräten
- Aktivitäten hinsichtlich der Buchführung (Records)
- Aktivitäten hinsichtlich der Berichte (Reports)

Alle denkbaren Aktionen in diesen Stufen werden jeweils in einem Aktionsbaum entwickelt und diese Teilbäume später durch eine logische UND-Verknüpfung wieder zu einem Gesamtbaum verbunden.

Wichtige Kenngrößen eines Fehlerbaumes, die seine logische Struktur beschreiben, sind die Minimalschnitte. Dabei wird unter einem Minimalschnitt die minimale Gruppierung von Komponenten verstanden, deren gleichzeitig vorliegender Ausfall den Systemausfall bewirkt.

Minimalschnitte bei den Aktionsbäumen von PASE sind die minimalen Gruppierungen von Elementaraktionen, die zusammen zum TOP-Ereignis führen. Bei einfachen Bäumen, wie sie bei der Anwendung von PASE in der Regel vorliegen sollten, können die Minimalschnitte leicht von Hand ermittelt werden. Für komplexe Fehlerbäume existieren rechnergestützte Verfahren zur Ermittlung der Minimalschnitte. Diese basieren in der Regel auf dem logischen Ausmultiplizieren des Baumes von oben nach unten.

Als Beispiel für einen Aktionsbaum ist in Abb. 1 der Teilbaum für die Freilegung bereits eingelagerter und versetzter Gebinden aus dem Endlager mit ihren Minimalschnitten dargestellt.

PASE kombiniert jeden Minimalschnitt eines Teilbaumes mit jedem Minimalschnitt der anderen Teilbäume. Jede dieser Kombinationen stellt dann einen theoretischen Abzweigungspfad dar. Bei komplexeren Modellen führt dies bald zu einer kombinatorischen Explosion bei der Anzahl der theoretischen Abzweigungspfade, z.B. ergeben sich bei 5 Teilbäumen mit je 10 Minimalschnitten bereits 10*10*10*10*10 = 100 000 verschiedene Kombinationen.

Festlegung des Statusvektors und der Indikatoren

Dadurch, daß die einzelnen möglichen Stufen einer Abzweigung in den einzelnen Aktionsbäumen völlig losgelöst voneinander betrachtet werden, hat die Anzahl der möglichen Kombinationen noch keinen praktischen Aussagewert. In der Regel stellt nur ein Bruchteil der Kombination für einen Abzweiger einen sinnvollen Pfad dar. So ist z. B. nicht sinnvoll, C/S zu manipulieren, wenn die Abzweigung nicht von diesem C/S-Gerät registriert werden konnte, etc.. Es stellt sich damit das Problem, die Anzahl der theoretisch möglichen Pfade auf "rationale" Abzweigungspfade zu reduzieren. Dies erfordert eine Bewertung der einzelnen Pfade.

Dazu wird in PASE das Konzept des Statusvektors und der Indikatoren eingeführt. Der Statusvektor ist eine Menge von verschiedenen Parametern, die das System beschreiben sollen und durch die einzelnen Aktionen des Abzweigers verändert werden. Als Parameter sind nur Zählgrößen zugelassen. Diese Zählgrößen können beispielsweise die Materialverteilung in der Anlage, d. h. die Anzahl von Items an den verschiedenen Orten, den Zustand der Anlage, z. B. die Anzahl der zerstörten Streckenabschlußbauwerke, und sonstige Ereignisse, z.

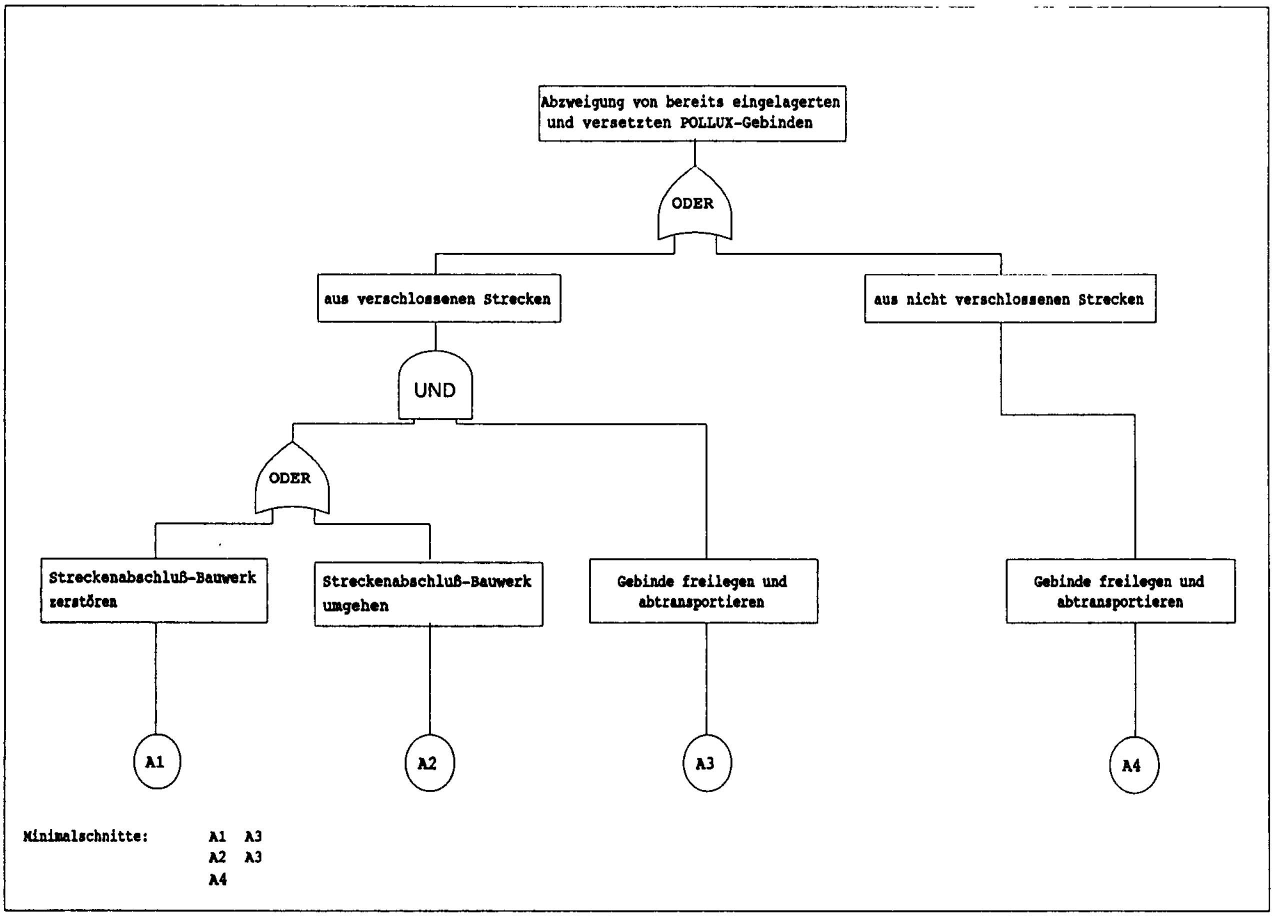

Abbildung 1: Aktionsbaum für die Freilegung des Kernmaterials

B. Kamera-Records, Siegelverletzungen, etc. beschreiben. Der Benutzer von PASE muß festlegen, welche Parameter für sein Modell von Bedeutung sind.

Für jede in den Aktionsbäumen aufgeführte Elementaraktion wird spezifiziert, wie sie welche Statusparameter verändert. So erhöht z. B. die Freilegung eines Gebindes in einer bereits verschlossenen Strecke den Zähler für zerstörte bzw. umgangene Streckenabschlußbauwerke und den Zähler für freigelegte Gebinde.

Anschließend muß der Benutzer Indikatoren festlegen, die Unstimmigkeiten im System anzeigen sollen. Sie sind als logische Daten mit den Zustandswerten "Wahr" und "Falsch" definiert und können gebildet werden durch einen Vergleich verschiedener Elemente des Statusvektors und ihrer Veränderungen. Diese Indikatoren sollen ohne Rücksicht darauf festgelegt werden, ob sie durch eine bestimmte Safeguardsmaßnahme entdeckt werden können oder nicht.

Das Konzept des Statusvektors und der Indikatoren erlaubt es nun, die einzelnen Pfade zu bewerten und Kriterien zu definieren, nach denen "irrationale" Abzweigungspfade aus der Menge der theoretisch möglichen Pfade eliminiert werden (Screening process). Ausgesondert werden z. B. alle Pfade

- die nur zusätzliche Indikatoren erzeugen, ohne andere zu verdecken,
- die keine Abzweigung darstellen,
- die nicht bestimmten, vom Benutzer definierten Bedingungen genügen (z. B. Pfade, die negative Bestände erzeugen)
- die die gleichen Statusparameterwerte aufweisen wie ein bereits berücksichtigter Pfad,
- die die gleichen Indikatoren erzeugen wie ein bereits berücksichtigter Pfad.

Das Ergebnis von PASE ist zunächst eine Liste der Pfade, die nach diesem Filterprozeß übrigbleiben. Diese Pfade können in einem weiteren Schritt noch gruppiert werden nach der Anzahl der Indikatoren, die sie erzeugen. Dabei werden die Pfade, die die wenigsten Indikatoren erzeugen als "hardest to detect" eingestuft.

Erfahrungen bei der Anwendung von PASE auf das Endlager

Die uns zur Verfügung stehenden Unterlagen zu PASE umfassen eine Darstellung des Verfahrens, mehrere PC Programme und drei Anwendungsbeispiele. Die Programmunterstützung setzt bei PASE erst in einer relativ späten Phase ein. Die Strukturierung des Problems, die Erarbeitung der Aktionsbäume, die Ermittlung der Minimalschnitte, die Festlegung des Statusvektors, die

Festlegung des Indikatorvektors und teilweise die Definition der Filterkriterien müssen vorab manuell erfolgen. PASE erwartet als Inputdateien den Statusvektor, den Vektor der Indikatoren, die Filterkriterien und für jeden Aktionsbaum eine Liste der Elementaraktionen und der Minimalschnitte. Diese Eingabedaten müssen in FORTRAN-Syntax erstellt werden. Sie werden von PASE nach einer formalen syntaktischen Prüfung in ein als Gerüst vorhandenes FORTRAN-Quellprogramm eingebaut. Das FORTRAN-Programm wird anschließend kompiliert und gebunden und berechnet dann die Kombinationen der Minimalschnitte. PASE arbeitet in diesem Bereich als reines Batchverarbeitungsprogramm. Eine interaktive Unterstützung für die Erzeugung der Eingabedateien steht nicht zur Verfügung.

Der Output des Programmes ist eine Liste der Pfade, die nach dem Filterprozeß als relevante Pfade übrig bleiben und Listen von Pfaden, die bei der Filterung eliminiert wurden. Diese Listen werden in einem internen Format als Dateien abgespeichert und können mit weiteren PASE-Programmen gelesen und nach bestimmten Kriterien sortiert werden.

Entsprechend der Verfahrensdarstellung soll im ersten Schritt der Analyse die Anlage beschrieben und der Umfang der Analyse festgelegt werden. Die Beschreibung der Anlage umfaßt dabei:

- die räumlich und technische Auslegung der Anlage,
- die Beschreibung des Materials, daß in der Anlage gehandhabt wird,
- Inventare und Bewegungen des Materials,
- eine Beschreibung der Prozesse, die am Material ausgeführt werden.

Das Mischkonzept für die Endlagerung sieht vor, daß ausgediente HTR- und LWR-Brennelemente und wärmeentwicklende radioaktive Abfälle sowie auch relativ schwach radioaktive Abfälle im gleichen Endlagerbergwerk eingelagert werden. Für die Einlagerung dieser Materialien werden verschiedene Konzepte untersucht, die teilweise weder eine zeitliche noch eine räumliche Trennung von überwachungspflichtigem und nicht überwachungspflichtigem Material im Einlagerungsablauf vorsehen. Wir müssen uns hier bei der Beschreibung der Anlage und des Modells notwendigerweise auf eine grobe Skizze beschränken. Für unsere ersten Anwendungen von PASE wurde ein vereinfachtes Modell zugrunde gelegt. Darin wird zunächst nur die Streckenlagerung von LWR-BE in POLLUX-Behältern während der Betriebsphase des Lagers betrachtet.

Gebindehandhabung im Endlager

Für die Gebindehandhabung wird von folgendem Ablauf ausgegangen (vergl. Abb. 2):

Der An- und Abtransport der POLLUX-Behälter erfolgt wahlweise über LKW oder Bahn über einen durchgehenden Fahrweg für beide Transportmittel. Nach der Eingangskontrolle werden die Fahrzeuge in die Entladeposition gefahren und die POLLUX-Behälter auf einen vorher bereitgestellten Plateauwagen in der Umladehalle umgesetzt. Bei einer Störung des Einlagerungsbetriebes werden die POLLUX-Behälter mit dem Kran in die Pufferhalle gesetzt und nach der Behebung der Störung auf den Plateauwagen geladen.

Vom Beladegleis werden die Plateauwagen zum Schachtbeschickungsgleis transportiert und auf den Förderkorb geschoben. Dann erfolgt der Transport nach Untertage bis auf die Einlagerungssohle.

Im Füllort werden die Plateauwagen durch eine Aufzieh- und Abschiebevorrichtung aus dem Förderkorb verfahren und auf das Bereitstellungsgleis gebracht. Mit einer Grubenlokomotive werden sie dann in den Einlagerungsbereich gezogen und auf dem Ausweichgleis für beladene Plateauwagen abgestellt. Hier werden sie von einer Rangierlokomotive übernommen, die jeweils einen beladenen Plateauwagen in die Einlagerungsstrecke schiebt.

Dort wird der Plateauwagen unter die Einlagerungsvorrichtung positioniert und der Behälter angehoben. Der Plateauwagen wird vorgezogen und der Behälter auf dem Gleis abgelegt. Um den Behälter versetzen zu können, muß zunächst die Einlagerungsvorrichtung aus der Strecke geholt werden. Dazu fährt sie zunächst mit ihrem Fahrwerk in eine Ablegeposition vor und wird dort auf der entladenen Plateauwagen aufgesetzt. Die Rangierlok transportiert die Einlagerungsvorrichtung in die benachbarte Einlagerungsstrecke, wo sie erneut aufgestellt und eingerichtet wird. Bevor die Versatzfahrzeuge den Streckenabschnitt versetzen können, sind die vorhandenen Einbauten und Einrichtungen, z.B. für die Bewetterung, zurückzusetzen.

Ist eine Einlagerungsstrecke vollständig mit Gebinden belegt, so werden die maschinellen Einrichtungen in die folgende Einlagerungsstrecke umgesetzt. Die geräumte Strecke wird vollständig mit Versatzgut verfüllt. Als Verschluß wird ein Abschlußbauwerk errichtet, daß als Abgrenzung des nicht mehr in Betrieb befindlichen Bereiches dient und als weitere Barriere gegen eine Nuklidausbreitung wirkt.

Betrachtete Abzweigungszenarien

Im Modell werden folgende Abzweigungszenarien betrachtet (vgl. Abbildung 2):

- Abzweigung nach dem Eingang in die Entladestation,
- Abzweigung aus dem Pufferlager und
- Abzweigung nach der Umladung auf dem Plateauwagen auf dem Weg zum Schacht.

Die Abzweigung kann dabei alternativ folgende Aktionen umfassen:

1. Wegnahme des Behälters,
2. Wegnahme des Behälters und Ersatz durch einen Dummy,
3. Wegnahme des Behälters, Öffnen des Behälters in einer heißen Zelle, Entnahme von Kernmaterial, Verschließen des Behälters und dessen Rücktransport (Entnahme von Kernmaterial).

Untertage werden folgende Abzweigungszenarien berücksichtigt:

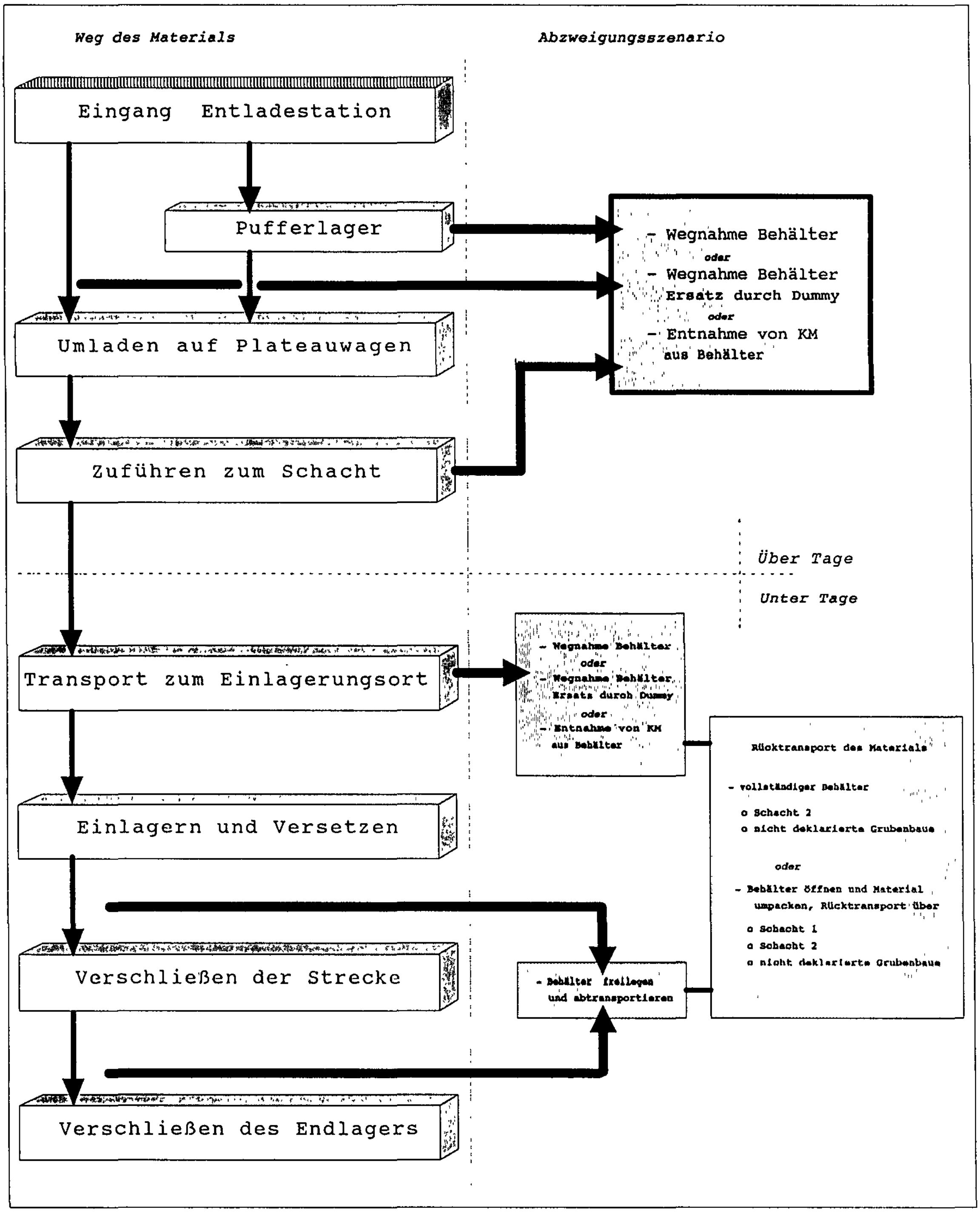

Abbildung 2: Gebindefluß und betrachtete Abzweigungsszenarien

- Abzweigung während des Transportes im Grubengebäude zum Einlagerungsort,
- Abzweigung nach dem Einlagern und Versetzen des Behälters und
- Abzweigung nach dem Verschließen der Strecke.

Die beiden letzten Möglichkeiten erfordern das Freilegen des Behälters und seinen Abtransport. Bei einer Abzweigung Untertage muß das Material nach Übertage zurücktransportiert werden. Für den Rücktransport werden folgende Möglichkeiten berücksichtigt:

1. Rücktransport vollständiger Behälter über:
   - Schacht 2
   - nicht deklarierte Grubenbaue,
2. Das Öffnen des Behälters, das Umpacken des Materials und Rücktransport über:
   - Schacht 1
   - Schacht 2
   - nicht deklarierte Grubenbaue.

Erfahrungen beim Aufbau des Modelles

Die Aktionsbäume für die betrachteten Abzweigungsaktivitäten konnten ohne große Schwierigkeiten erstellt werden. Probleme ergaben sich dann jedoch bei der Definition des Statusvektors, und zwar bei der Frage, wie die zeitlichen Abläufe, d.h. der Gebindefluß, im Modell effizient abgebildet werden können.

Für PASE gibt es kein Tutorial und die Verfahrensbeschreibung ist in diesen Punkten relativ allgemein gehalten. Konkrete Regeln, wie in welchen Fällen vorzugehen ist, sind in der Dokumentation nicht aufgeführt.

Bei den zur Verfügung stehenden Beispielen wurden der Statusvektor und die Indikatoren im Prinzip aus der Detaillierung der Materialbilanzgleichung und den Maßnahmen zur Überprüfung des realen Bestandes abgeleitet. Es wird ausgegangen von einem statischen bzw. stationären Zustand, der durch die Aktionen des Abzweigers verändert wird und nach der Aktion überprüft werden kann. Die Indikatoren werden im wesentlichen gewonnen durch den Vergleich des Ausgangszustandes mit dem Zustand nach der Abzweigungsaktion.

In Endlager muß jedoch auch der Fluß der Gebinde über mehrere Stationen betrachtet werden. Das System kann dabei eine Vielzahl verschiedener Zustände annehmen, je nachdem wo und wie die Abzweigung erfolgt (Wegnahme eines Gebindes ohne Ersatz, Ersatz durch Dummy, Öffnen des Gebindes und Entnahme des Materials, etc.) und und dieser Zustand ändert sich mit jedem Weitertransport des Gebindes (von Station A an Station B wird ein reguläres Gebinde weitergegeben, es wird kein Gebinde weitergeben, es wird ein Dummy weitergeben, es wird ein leeres Gebinde weitergegeben, etc.). Um alle diese verschiedenen Zustände in einem Modell zu erfassen, ist die Aufstellung eines relativ komplexen Statusvektors erforderlich, da der Statusvektor nur Zählgrößen enthalten darf.

Wegen des Fehlens von konkreten Verfahrensregeln ist der Benutzer von PASE auf eigene Experimente angewiesen. Diese werden jedoch von PASE in keiner Weise unterstützt:

- Der Benutzer erhält erst in einem relativ späten Stadium, nachdem er sein Modell konstruiert und die Eingabedateien zusammengestellt hat, eine Rückkopplung vom Programm. Er muß im Vorlaufbereich schon sehr viel Aufwand erbringen, ohne zu wissen, ob er sich in die richtige Richtung bewegt.
- Die Fehlermeldungen und die vorhandenen Möglichkeiten, die Arbeitsweise des Programmes nachvollziehen zu können, sind sehr dürftig. Bei den ersten Läufen wurden z. B. alle Abzweigungspfade als irrational verworfen, die Ergebnisliste enthielt keine Pfade mehr. Zur Fehlersuche sahen wir uns gezwungen, den FORTRAN-Quellcode zu ergänzen, um auch Zwischenergebnisse anzeigen zu lassen.
- Erstellung und Änderungen der Eingabedaten sind extrem aufwendig und fehleranfällig. Die Statusparameter und die Indikatoren müssen in FORTRAN-Syntax als Elemente von Feldern mit fortlaufendem Index angegeben werden. Statusparameter und Indikatoren haben dabei z. B. die Form S(17) und I(58). Entfällt ein Parameter oder wird einer hinzugefügt, so müssen alle nachfolgenden Indizes entsprechend geändert werden. Da bei der Definition der Indikatoren und der Beschreibung der Aktionen auf die Statusparameter zugegriffen wird, müssen auch diese Referenzen entsprechend angepaßt werden. Dies muß alles manuell in einem Editor ohne jegliche Programmunterstützung durch PASE erfolgen.
- Bei komplexeren Modellen steigen aufgrund der kombinatorischen Explosion der zu betrachtenden Anzahl von Pfaden die Programmlaufzeiten erheblich an. Sie betragen teilweise mehrere Stunden, auch auf einem leistungsfähigen 386-PC. Dies ist für den Benutzer kaum zumutbar.

Der Output von PASE sind Listen von Minimalschnitt-Kombinationen, die nur mit den dafür angebotenen Programmen analysiert werden können. Dabei kann für diese Abzweigungspfade eine Rangordnung erstellt werden. Diese Rangordnung basiert auf die Anzahl der Indikatoren, die auf dem einzelnen Pfad erzeugt werden. Die Indikatoren werden dabei nur anzahlmäßig und nicht qualitativ betrachtet. So wird z. B. die Wegnahme eines Gebindes aus dem Pufferlager ohne weitere Verschleierungsaktionen als "hardest to detect" eingestuft, da hier nur ein Indikator erzeugt wird, der eine Unstimmigkeit zwischen Buchbestand und realem Bestand im Pufferlager anzeigt.

In einer weiteren Option der Analyse können die Abzweigungspfade einem Safeguardssystem gegenübergestellt werden. Dies erfolgt dadurch, daß für das Safeguardssystem angegeben wird, welche der Indikatoren vom ihm erfaßt werden können. Dann erfolgt die Gruppierung im Hinblick auf die Entdeckungsfähigkeit des Safeguardssystems, d. h. nach der Anzahl der Indikatoren, die vom Safeguardssystem erkannt werden können.

Zusammenfassung und Empfehlungen

Gegenüber der konventionellen Vorgehensweise bei der Analyse von Abzweigungspfaden bietet PASE zunächst den Vorteil eines strukturierten und systematischen Ansatzes. Dadurch, daß alle möglichen Stadien von Abzweigungsaktionen unabhängig voneinander bis ins Detail aufgeschlüsselt und dann per Programm kombiniert werden, ergibt sich eine gewisse Gewähr dafür, daß keine relevanten Abzweigungsmöglichkeiten übersehen werden.

Der Einsatz von PASE erscheint lohnenswert insbesondere für die Untersuchung von komplexen Zusammenhängen, die manuell kaum noch überschaubar sind, und für die Analyse von alternativen Safeguardsmodellen. Bei einem gegebenen Satz von Abzweigungspfaden können mit PASE Safeguardsmodelle leicht daraufhin überprüft werden, inwieweit sie diese Abzweigungspfade erfassen.

PASE kann auch als ein Hilfsmittel eingesetzt werden, um Abzweigungspfade in einer standardisierten Form verbal zu beschreiben und graphisch darzustellen. Eine übersichtliche Darstellung erleichtert das Verständnis und die Kommunikation.

Der Umgang mit PASE erfordert allerdings einen hohen Lernaufwand vom Benutzer. Die PC-Programme sind zu starr und nicht benutzerfreundlich. Es ist zwar zu berücksichtigen, daß PASE noch ein sehr junges Verfahren ist und sich noch in der Entwicklung befindet, jedoch entspricht die Handhabung nicht heutigen Maßstäben. Die Arbeit mit PASE ist derzeit zu vergleichen mit der Programmentwicklung mit einem Compiler ohne eine Entwicklungsumgebung und mit nur minimalen Debugging-Möglichkeiten.

Für PASE sollte ein Tutorial erarbeitet werden, das den Anwender bei der Erstellung des Modells unterstützt. Insbesondere ist eine Hilfestellung für die Abbildung von Prozessen, z. B. von Materialflüssen, erforderlich. Dieses Tutorial könnte auch zur Standardisierung des Modellentwurfes beitragen.

Im Bereich der Dateneingabe für PASE sind unbedingt Verbesserungen erforderlich. Die derzeitige Form entspricht nicht dem Stand der Technik. Hier sollte eine Modell-Entwicklungsumgebung erarbeitet werden, die den Benutzer bei der Erstellung, Änderung und Konsistenzprüfung des Modelles unterstützt. Der Output von PASE sollte in einem Format zur Verfügung gestellt werden, das dem Anwender eine eigene Weiterverarbeitung über den von PASE angebotenen Rahmen hinaus ermöglicht.

Die Programmlaufzeiten sind auf PC-Maschinen unzumutbar lang. Wenn nicht softwareseitig, z. B. durch eine Optimierung der Berechnungsalgorithmen, eine erhebliche Verringerung erzielt werden kann, ist eine Portierung auf leistungsfähigere Hardware erforderlich.

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EVALUATION OF MEASUREMENT DATA FROM CONTINUOUS CALIBRATION PROCEDURES AT AN INPUT ACCOUNTANCY TANK

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Abstract

One of the essential aims of the *CALDEX* project [1] is the comparative evaluation of the conventional tank calibration procedure using an incremental addition of liquor and of the calibration with a continuous liquid feed.

The three calibration exercises performed with water in continuous mode have been evaluated. Estimates of volume determination uncertainty are given for all manometers which can be compared with the evaluations from other calibration procedures.

1 Introduction

The *CALDEX* project [1] the experimental part of which was carried out and concluded during the year 1989 was aimed for comparing various calibration methods for accountancy tanks. Apart from a comparison of measurement devices which were tested simultaneously and from an investigation of influencing factors which occur during plant operation, particular emphasis was placed on the analysis of a calibration procedure with a continuous liquid feed. It was to be determined wether continuous calibration — which reduces the time needed by the factor 6 compared to the conventional procedure — meets the requirements as to precision and to reproducibility.

A first examination of data did not yield any reliable result on the comparison between conventional, i.e. incremental; and continuous calibration. Therefore a detailed investigation of the continuous calibration procedure was carried out.

2 Experimental setup

In the *TEKO* facility (Karlsruhe) a 12500ℓ annular vessel has been equipped to allow studies on the degree of accuracy of measurements achievable in conditions of operation.

The whole setup consists of three main components: the accountancy tank including instruments, the dosing station and storage tanks (see fig. 1).

For the continuous calibration experiments the calibration liquid was pumped from the storage tanks into the overflow feed tank and passed, over the rotary piston meter, into the accountancy tank continuously.

Calibration data have been recorded with the following instruments:

- Volume measurements
  - Rotary piston meter
- Level measurements
  - *Crouzet* electro-mechanical gauge
  - *Ruska* electro-mechanical gauge
  - *Wallace & Tiernan* electro-mechanical gauge, type Diptron 3
  - *Hartmann & Braun* standard pressure transmitter (*RUF*)
  - Capacitive level measurement probe
  - Sonic ranger (UKAEA Harwell)
- Tank weight
  - *ASEA* weighing system (load cells)

3 Screening of measurement data

Measurement data have been stored in two datasets for each calibration run. A high speed recording dataset contains all readings from level instruments and the rotary piston meter. The time resolution between readings is 2 seconds. The second dataset with a 30 second recording interval contains data about the environmental conditions of the calibration: Temperature

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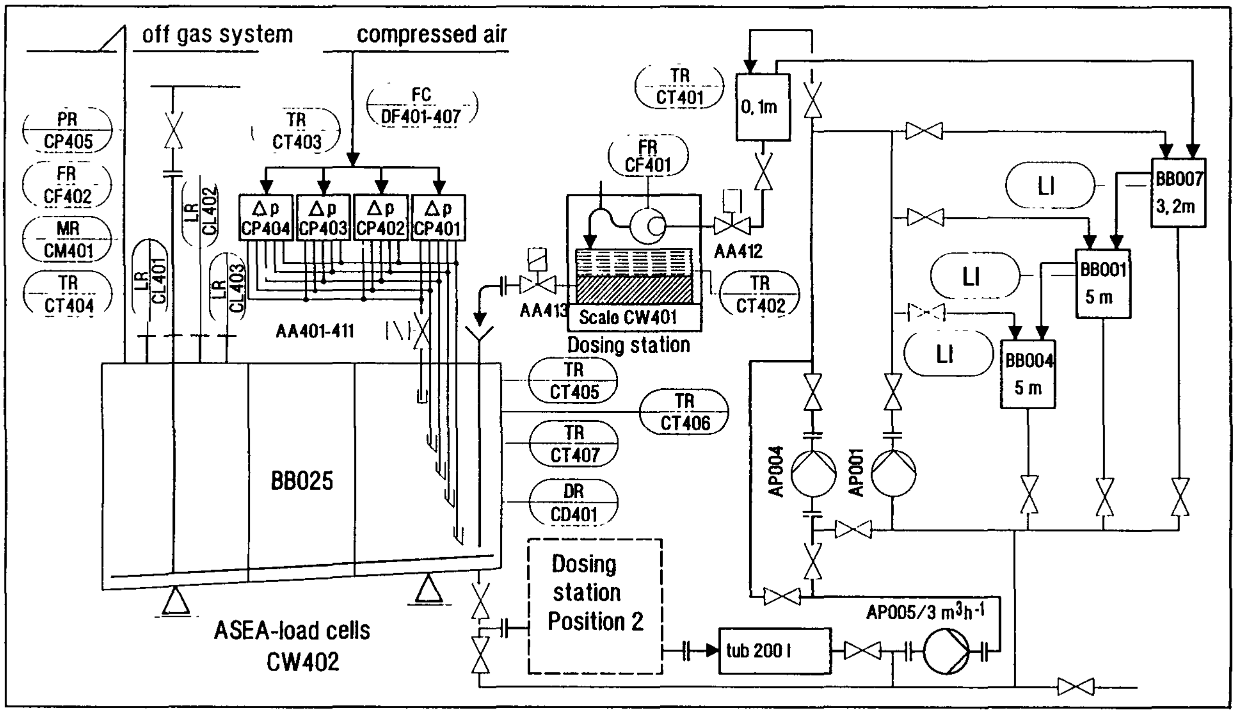

Figure 1: CALDEX setup

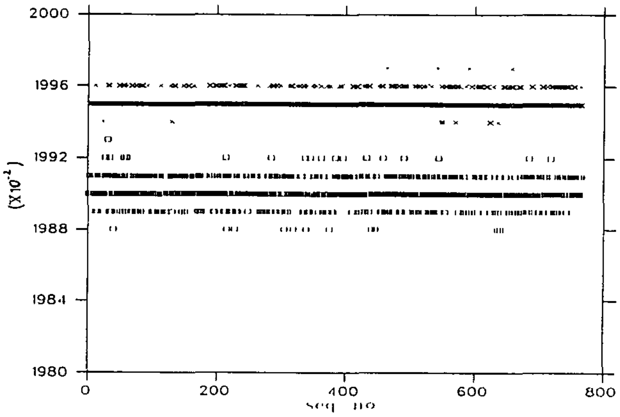

Figure 2: Constancy of air flow rate

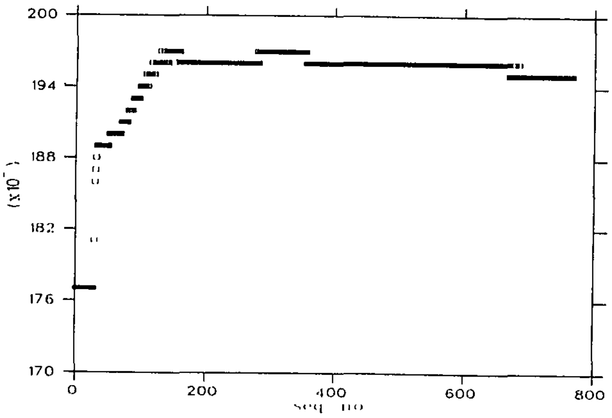

Figure 3: Temperature measured at the bottom of the accountancy tank (run 1)

readings within the feed and calibration tanks and the air flow rates in the dip–tube system.

The constancy of the air flow rate was within two steps of the resolution of measurement($0.01\ell/h$; see fig. 2). The temperature within the tank was stable within ±0.1 °C (resolution of measurement) after the thermo cell location was flooded with water (see fig. 3). Therefore no correction within one calibration run is necessary. Unfortunately all temperature measurements for the last calibration run failed, there are constant readings in the dataset over the whole period of time including the starting phase where none of the thermo cells was in contact with the calibration liquid. This lack of information precludes a temperature correction for this calibration run — all the following evaluation has been made under the assumption that the temperature for all these calibration runs was identical but unknown.

Time correlation of measurements Volume measurement with the rotary piston meter and the corresponding liquid level measurement are done at different times, because the rotary piston meter is located atop of the accountancy tank and the water needs a little time to pour down into the tank. The filling line was vented before each calibration run; it is running down to the bottom of the tank. During the calibration run water is flowing at a nearly constant speed. For this setup the time correlation is only a constant bias [2], which can be determined at the starting time of the calibration from the delay of the weight measurements of the whole tank against the first none–zero recording from the rotary piston meter. The start of differential pressure readings is not suitable for this purpose because the level of liquid is below the outlet of the lower dip tube at the beginning of the calibration run.

This kind of time correlation requires no within–run corrections and has no influence for the estimation of volume determination uncertainty.

Detection of outliers A first graphical analysis of the measurement data showed two kinds of outliers:

- The differential pressure measurement failed for some times because the dip tubes have been vented.
- there are a number of constant readings — usually on all data lines — so that the next couple of data is only a replicate of the former one and no true data.

A data averaging program handed over together with the original data did not take account for these outliers and could therefore not be applied.

The latter kind of outliers was easily removed from the data by searching for constant readings in pairs of data. About one third of all data have been rejected this way. The other outliers required individual identification by picking them interactively from the residual plots.

Correction of zero readings At the beginning and the end of each calibration run zero readings have been taken from all level recording instruments. The rotary piston meter is a counting device which is reset to zero at the beginning of each calibration run.

The zero readings for each instrument have been averaged separately for the starting and the ending phase. Differences between these both averages have been equally distributed over the time range of calibration.

Evaluation of diagnostic plots Profile plots of the reduced calibration data are used for diagnostics. A profile plot is a plot of the residuals obtained by fitting a linear function to the calibration data. The profile plots shown here (figure 4 to 6) are constructed from all runs. A profile plot shows graphically the change in the cross–sectional area of the tank. Abrupt changes in the tank's profile help to identify transition regions.

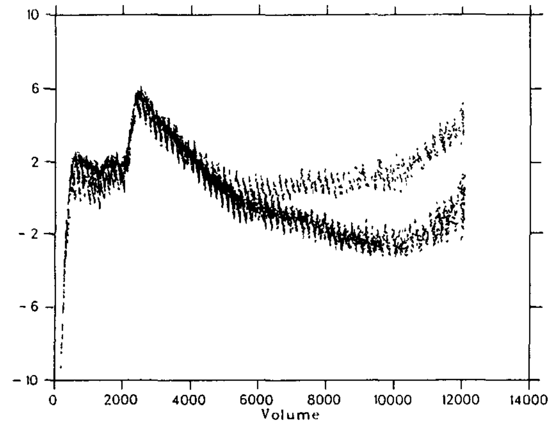

Figure 4: profile plots *Ruska*

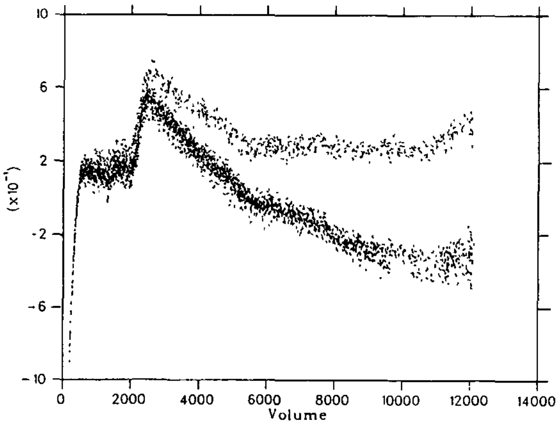

Figure 5: profile plots *Diptron*

Looking at the profile plots the instruments can be separated into two distinct groups: all differential pressure recording instruments are one group, the sonic ranger, capacitive probe and the load cells are the other one. For the differential pressure instruments the profiles from run 1 and run 2 match very well, the profile of run 3 is significantly different. The increasing difference to the other profiles assumedly is a scaling error — there are no valid temperature measurements for run 3 — or a malfunction of the data transmission equipment. Under these circumstances it is at least doubtful if one should include the data from run 3 into the data evaluation. We decided to reject the whole calibration run.

With only two calibration runs it is not useful to

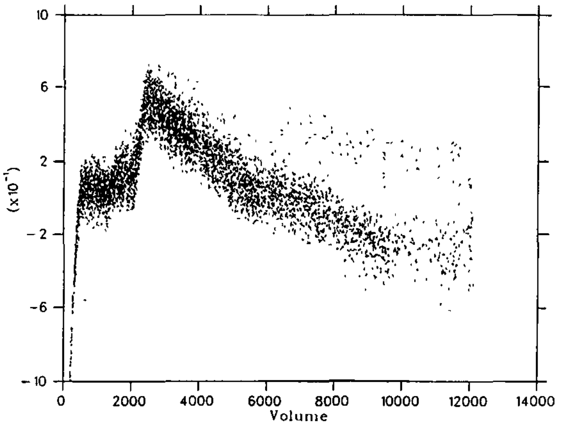

Figure 6: profile plots *Hartmann & Braun*

compare different error models; this should be done later including the results of future calibration runs performed with the *CALDEX* setup at Ispra.

For the second group of instruments the profiles of the three calibration runs differ widely in slope. These instruments should not be used for accounting purposes.

4 A measure for comparison of calibration procedures

The purpose of tank calibration is to determine the liquid contained within the tank as precise as possible. Comparative evaluation of different calibration procedures should therefore give an estimation about the uncertainty of volume determination. Such uncertainties can be derived from variance estimates of the liquid level measurements.

The variance estimates should include a component for run-to-run variances. Using high-precision instruments for volume and liquid height measurements the within-run measurement errors are smaller than the run-to-run differences resulting presumably from uncontrolled calibration conditions.

These two variance estimates — within-run variance σ_M (related to the calibration function) and run-to-run variance σ_R — depend on the regression model which is adequate to the calibration data — a single function describing all calibration runs, parallel functions or individual functions for each run. A detailed description of the component variances for the three possible regression models (restricted to the linear case) is given in Liebetrau et al. [3].

The adequacy of a regression model can be determined by the significance of the model's lack-of-fit against the next more individual model: the single function against parallel functions and the parallel functions against individual functions. A lack-of-fit test for the individual functions cannot be done, because due to the design of the calibration experiment there are no repeated measurements at exactly the same volumes to determine the pure error of measurement.

From the variance estimates confidence intervals can be derived to show where the true curve may lie relative to the fitted curve within a specified confidence $(1 - \alpha)$. For this purpose Scheffé-type confidence intervals are used.[4]

$$W(Y)_{\text{Scheffé}} = \pm\sqrt{2F^{\alpha/2}_{2,\nu}}\,\sigma(Y|X) \qquad (1)$$

In the classical tank calibration procedure the response variable Y is the liquid level height, $\sigma(Y|X)$ is the variance of liquid level measurement at a given volume X. The parameters of the F-distribution are the number of parameters of the regression function — 2 for linear functions — and the degrees of freedom of the overall variance of liquid height measurement.

The procedure of volume uncertainty depends on the selection of the response variable. Choosing the liquid level as the response variable, the regression function must be inverted for volume determinations. Liebetrau et al. [3] developed a procedure to obtain uncertainty estimates for volume determination. Prediction intervals that hold simultaneously with specified propability for an indeterminate number of volume determinations can be calculated by combining the confidence intervals for the calibration functions with the tolerance interval for liquid level measurements (see fig. 7). If liquid level measurements for input accounting are done the same way as calibration measurements, the variance of these measurements could be estimated from the calibration data by using

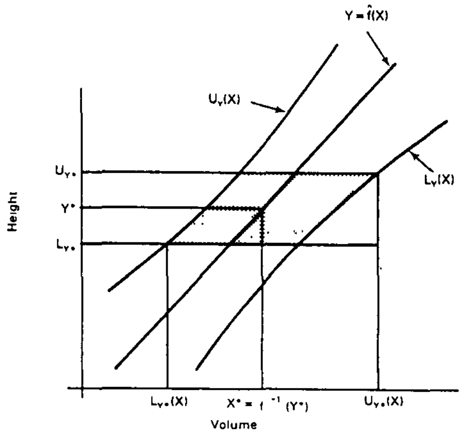

Figure 7: Determination of discrimination intervals

Crouzet

region	confidence interval for the cal. fct.	tolerance interval for level measurement	volume uncertainty
	[mm]	[mm]	[ℓ]
1	3.70	32.60	87.90
2	0.35	0.67	3.23
3	0.16	0.65	2.56
4	0.17	1.12	3.98
5	0.29	0.66	3.03
6	0.32	0.59	2.89
7	0.26	0.68	2.98
8	0.53	1.00	4.83

Diptron

region	confidence interval for the cal. fct.	tolerance interval for level measurement	volume uncertainty
	[bar]	[bar]	[ℓ]
1	0.380	2.97	78.5
2	0.010	0.11	3.9
3	0.022	0.12	4.4
4	0.021	0.14	4.9
5	0.018	0.12	4.4
6	0.032	0.12	4.6
7	0.093	0.13	7.0
8	0.110	0.23	10.6

Ruska

region	confidence interval for the cal. fct.	tolerance interval for level measurement	volume uncertainty
	[mm]	[mm]	[ℓ]
1	3.40	32.40	84.6
2	0.98	0.87	5.9
3	0.76	0.88	5.2
4	0.58	1.06	5.1
5	0.54	0.84	4.4
6	0.36	0.85	3.9
7	0.32	1.02	4.2
8	0.88	2.03	9.2

RUF

region	confidence interval for the cal. fct.	tolerance interval for level measurement	volume uncertainty
	[bar]	[bar]	[ℓ]
1	0.410	3.24	84.5
2	0.031	0.21	7.8
3	0.034	0.22	8.1
4	0.035	0.25	8.6
5	0.010	0.24	7.9
6	0.034	0.25	9.0
7	0.150	0.31	14.6

the within-run variations. This does not work for continuous calibrations. On the other hand the variances of the repeated readings at each increment during the classical incremental calibration runs — which is comparable to accountancy measurements — are not known to the authors. Therefore we use the within-run variances of the continuous calibration runs as an upper estimate for the tolerance intervals of liquid level measurement. During accountancy measurement there are no disturbances from liquid streaming into the tank — measurement variances will be smaller.

5 Results

Evaluation of the continuous calibration exercise has been restricted to the first two runs, because the runs environmental conditions are unknown due to a data recording failure.

The volume uncertainty estimates for the differential pressure measuring instruments are shown in the tables above. The selection of calibration regions can be seen exemplarily for the *Crouzet* manometer in figure 8. From the results it can easily be seen that the first region cannot be fitted sufficiently with a linear regression function. For all other regions the volume determination uncertainties are in the range from $3\,\ell$ to $15\,\ell$ with the smaller intervals for the *Crouzet* and *Ruska* instruments. These volume determination uncertainties are about 0.1 % or less of the tanks nominal volume ($12.5\,m^3$).

As stated above the tolerance intervals for level measurements have been estimated from the continuous calibrations within-run variations. The variance of accountancy measurements using repeated instrument readings at constant volume might be significantly smaller. This would also reduce the uncertainty of the volume determination.

During run 2 and run 3 the readings from the *Ruska* showed periodic variations of about $2\,mm$ with a periodicity of 75 seconds approximately. Therefore the volume uncertainties obtained for the *Ruska* are not as good as one would expect of the instruments precision.

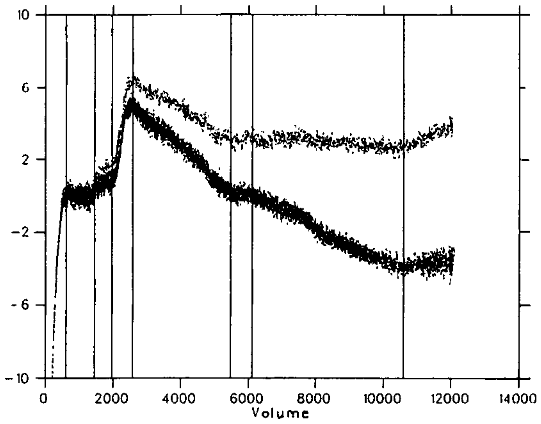

Figure 8: Calibration regions *Crouzet*

6 Concluding remarks

- The complete failure of the third calibration run shows the necessity of preliminary evaluation of the data recorded simultaneously with the calibration exercise. Even for a relatively fast calibration procedure like the continuous calibration 6 to 8 hours are needed to perform one calibration run for accountancy tanks of such great size — lost time if one instrument or data line would fail.
- To obtain more realistic estimates for the tolerance of liquid level measurements during accounting the results from the incremental calibration exercises with the *CALDEX* tank (Goldman et al., [5]) should be used upon availability.
- Calibration exercises to be performed with the *CALDEX* tank at its new location at Ispra can possibly be evaluated together with the calibration runs evaluated in this paper to see the influence of large time intervals betweeen calibrations — or a different tank profile due to the displacement.

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SAFEGUARDS CONCEPTS FOR SPENT FUEL CONDITIONING FACILITIES

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Abstract

Spent fuel conditioning is a likely option prior to disposal in geological repositories. During conditioning, spent fuel assemblies will be repacked with possible dismantling or even chopping in a final disposal container. Loss of identity of fuel assemblies will have a major impact on safeguards. The inventory of final disposal containers has to be re-established. This would require the development of an advanced accountancy verification and containment and surveillance technique in order to maintain effective safeguards. This paper identifies the system requirements for conditioning plants which would provide adequate assurance of non-diversion of nuclear material.

1. Introduction

Direct disposal of spent fuel has developed an important option for the nuclear fuel cycle. The two overlying objectives of underground disposal are[1]:

- to isolate spent fuel from the human environment over long time-periods without relying on future generations to maintain the integrity of the disposal system or imposing significant constraints on them due to the existence of a repository; and
- to ensure the long-term radiological protection of humans and the environment in accordance with current internationally agreed radiation protection principles.

To achieve these objectives, spent fuel has to be immobilized and conditioned in accordance with specifications dictated by regulatory authorities. The goal of conditioning is to immobilize and package spent fuel so that it is suitable for all stages of handling, storage and disposal.

The conditioning provides:

- Safe handling during the pre-disposal and disposal operation, i.e. storage, transportation and emplacement in a repository to ensure no release of radioactivity under normal conditions; release under accidental conditions needs to be acceptable to the regulatory authorities;
- Assurance that releases of radionuclides into the biosphere must be as low as possible, and within limits established by the regulatory authorities.

To fulfil these requirements spent fuel will be processed and packaged in final disposal containers making it inaccessible for verification purposes using the existing material verification procedure. These containers will contain a large amount of nuclear material and an advanced verification technique will be needed to provide adequate assurances of non-diversion as demanded by the international community. Development of advanced safeguards technology is essential to reduce facility intrusion, radiation exposure to inspector/operator personnel and inspector personpower costs. To realize this goal, development of (a) redundant-automated verification and surveillance equipment, (b) new techniques and equipment for verification of spent fuel assemblies and pins, and (c) techniques to monitor nuclear material content of the final disposal package, will be required.[2]

One may point out that a documentation of radioactive inventory including contents of the nuclide and its chemical composition will be required by the national regulatory authority. A verification procedure would have to be employed, not only for the requirements of the regulatory body, but also for addressing public concern. Verification may be carried out either at the conditioning plant or at the repository site to confirm the contents. Whatever verification procedure is adopted for regulatory purposes, safeguards related activities could be performed simultaneously with other operational practices.

2. Safeguards System.

The purpose[3,4] of IAEA safeguards is to assure that nuclear material is not diverted from peaceful purposes. The objectives are "the timely detection of diversion of significant quantities of nuclear material from peaceful nuclear activities to the manufacture of nuclear weapons or of other nuclear explosive devices or for purposes unknown, and deterrence of such diversion by the risk of early detection". A set of parameters called detection goals are used to design a safeguards system; these are: the significant quantity, the detection time, the detection probability and the false alarm probability. In the case of spent fuel, the detection goals are based on plutonium as it is a more desirable material for proliferation. The detection goal for plutonium in spent fuel is to detect a missing amount of 8 kg within 3 months with a detection probability of 90 - 95%[5].

Using the detection goals as guidelines, the IAEA draws up the inspection goals for every facility, taking the design and operational practice of the facility, the provisions of the safeguards agreement and the state-of-the-art technical capabilities of safeguards measures. Appropriate safeguards approaches are developed, designed and implemented accordingly.

A system of accountancy for, and control of, nuclear material is required to assure that the safeguards system and measures are effective to demonstrate continued presence of the material within the designated boundaries. The plant operator is obliged[3,4] to provide the Agency with its description of a procedure for nuclear material accountancy and control including a system of measurements on which such accountancy is based.

3. Safeguards for Spent Fuel.

The Agency's safeguards for irradiated reactor fuel assemblies is one of item accountancy. Nuclear material content is based on fabrication measurement data and adjusted based on calculation of production and loss during irradiation. Nuclear material content is assumed to be traceable as long as item integrity is maintained. The success of item accountancy as a safeguards measure is critically dependent on the ability to implement measures that provide an acceptable assurance of continued item integrity. Safeguards measures include the use of a combination of containment and surveillance measures with periodic verification by item counting, serial number identification and fingerprint radiation emissions.

Existing safeguards measurement techniques have been challenged by recent technological developments for better management of reactor fuel. Recent fuel design permits easy disassembly. This makes it more cost-effective to repair and reconstitute assemblies for reuse. During such operations fuel assemblies lose their original constituents and identity. This introduces new problems for material accountancy in addition to complications in the application of containment and surveillance methods. Advanced fuel designs incorporating different initial enrichments, axial variations in enrichment, use of mixed-oxide fuel and greater use of burnable poison rods have introduced additional difficulties for NDA measurements.

In addition, expansion of reactor storage pool capacities with high density storage racks, consolidation of fuel rods, double stacking of fuel assemblies and the use of special storage baskets or multi-element bottles to accommodate an increasing number of spent fuel assemblies have added further complications in the application of safeguards measures. Currently used safeguards methods have difficulty in verifying the loading of irradiated fuel shipping containers, fuel assembly dismantling and reconstitution, or the removal of individual spent fuel rods.

These problems have raised questions about the basic assumption and hence the adequacy of safeguards measures based on item accountancy. More recently the alternative of reverifying integrity by NDA measurements has been given increased attention. Considerable progress has been achieved in non-destructive-analysis (NDA) verification instruments and technique. Further improvements can be achieved by constructing facility specific NDA instruments. Operator understanding and cooperation for adopting are necessary for successful application of these advanced instruments.

4. Conditioning of Spent Fuel

As mentioned earlier, the goal of Conditioning is to immobilize and package spent fuel so that it is suitable for all stages of handling storage and disposal and to minimize release of radionuclides into the biosphere to be as low as possible. Conditioning[2,6,7] is normally performed under dry conditions. At a conditioning facility the spent fuel would be transferred to a hot cell for further processing. A conditioning facility receives (a) intact fuel assemblies (b) defective fuel assemblies (c) containers of consolidated fuel rods in case rod consolidation has been performed at the spent fuel pond and (d) containers of damaged/defective fuel rods. These will be processed in the following manner:

(a) Encapsulation of intact fuel assemblies or containers consolidated spent fuel rods in the final disposal cask; this involves operations without any further disassembling;

(b) Encapsulation of disassembled components: In this case fuel assemblies/containers of consolidated rods are disassembled and the disassembled components are packed into canisters and subsequently into final disposal casks. Depending on operational requirements, fuel and non-fuel bearing components will be separated;

(c) Encapsulation of divided component: In some cases, depending on operational requirements, disassembled fuel components will be divided into pieces before placing into final disposal casks;

(d) Encapsulation of damaged fuel assemblies/rods will be processed by using methods described above;

(e) Encapsulation of waste generated by the process operation which will also be packed in a final disposal cask of same specification.

A flow diagram of nuclear material in a conditioning plant is given in Figure 1. The shaded part shows the places where the spent fuel processing will take place in the hot cell. It may be mentioned here that any processing facility which causes MUF (material unaccounted for) will need verification techniques to determine the amounts involved.

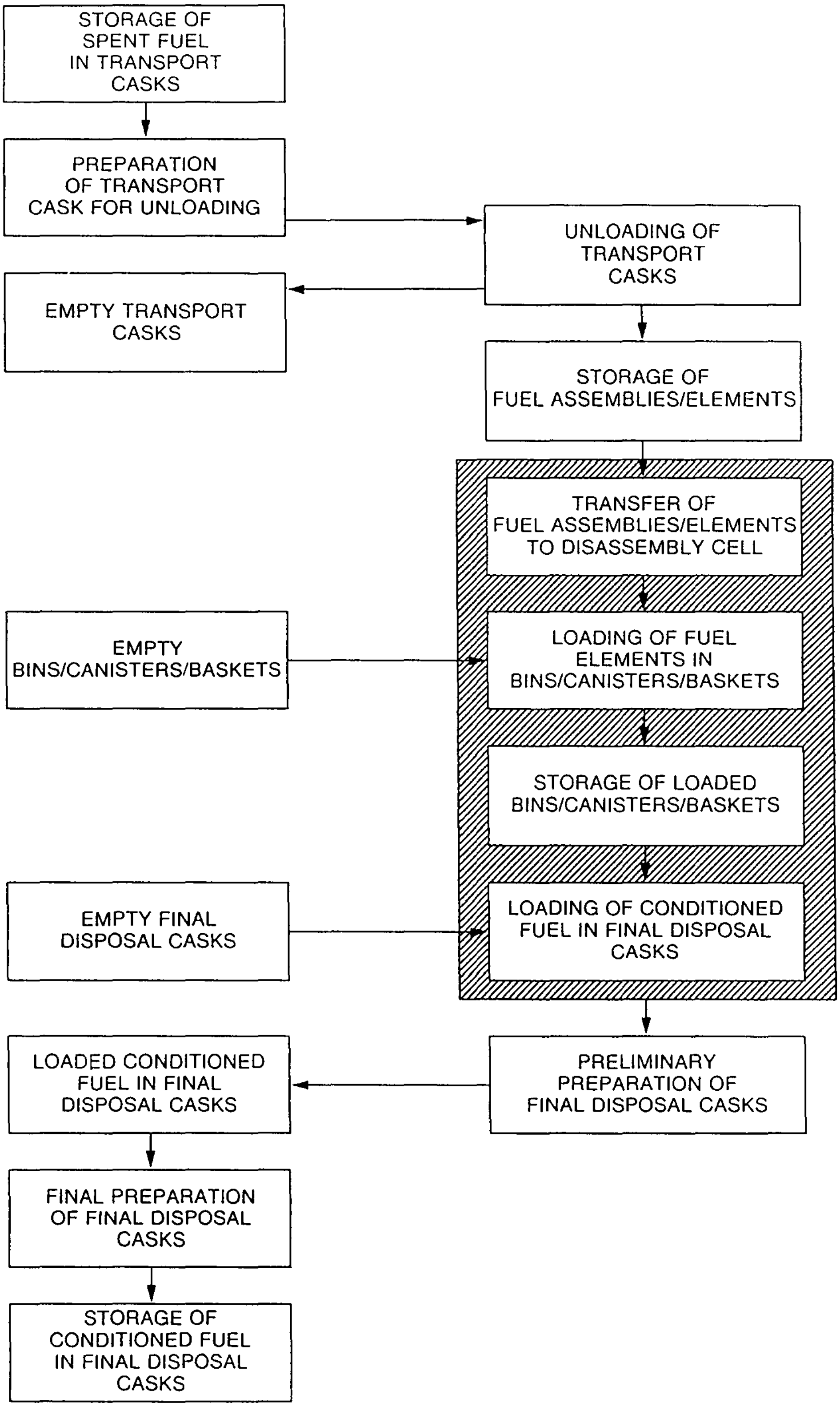

Figure 1 FLOW DIAGRAM OF NUCLEAR MATERIAL IN A CONDITIONING PLANT

5. Safeguards concern

Fuel assemblies arrive at the conditioning plant with one very important assumption that their integrity has not been breached. Assurance must be provided that all nuclear material has actually been received at the conditioning facility. Many reactors currently reconstitute fuel assemblies to obtain additional burn up. The safeguards accounting concern for these reconstituted assemblies need to be addressed. The safeguards concern for diversion of material by understating the nuclear material content of the fuel assemblies must also be taken into account.

During conditioning a transition from one type of the items, i.e. fuel assemblies/rods into another type of item, i.e. final disposal container (mentioned as item-item transition later in the text) will occur. The content of this new item should be verified in respect of nuclear material content according to the prescribed detection goal. This reinforces the need for material accountancy verification procedures to maintain continuity of knowledge during this item-item transition in order to draw Safeguards conclusions. Effective safeguards depends on the accounting practice and its verification techniques.

Possibility of diversion of nuclear material during conditioning operation needs to be addressed by applying appropriate verification measurements. If falsification is not detected through such verification, then it will never be detected because the quantity represented by the falsification will be buried along with the spent fuel in the geological repositories.

Implementation of effective safeguards will require highly reliable safeguards systems, since reverification of the nuclear material inventory in the event of any anomaly or discrepancy will be extremely difficult. A sound quality assurance programme is required to ensure the effectiveness of the applied safeguards measures.

6. System requirements.

Considering these factors, the following systems requirements for implementing effective safeguards deserves consideration. To facilitate the application of safeguards, integration of these requirements needs to be incorporated during the early stages of facility design. If operator systems are used, it will be required that Agency specifications are fulfilled for authentication.

a) NDA System

i) The system should be able to verify the nuclear material of all fuel assemblies/rods in unattended mode during unloading from the shipping cask.

ii) The system should be able to verify the nuclear material of all fuel assemblies/rods/other waste material during loading into the bins/canisters after disassembly or other processing stages, as appropriate.

iii) The system should be able to verify the nuclear material during loading of final disposal cask.

iv) The system should be able to transmit measurement data over a reasonable distance to the inspectors' room.

v) The system data review capabilities should not interfere with the system when operating in data acquisition mode.

vi) The system should incorporate diagnostic checks with an alarm to indicate when it is not functioning within acceptable bounds.

vii) The system should be fully automatic and operate in an unattended mode.

b) C/S System

- i) The system should be able to count and identify fuel assemblies/rods and other material which enters the facility.
- ii) The system should be able to distinguish various types of material which will be handled by the facility.
- iii) The system should provide surveillance of the storage hall in order to cover all cask movements into and out of the area.
- iv) The system should be able to verify that all casks leaving the area are consistent with the operator's declaration.
- v) The system should be operable in an unattended verification mode by surveillance and pattern recognition of a neutron and/or gamma monitoring system to confirm that all movements are as recorded by the operator.
- vi) The system should be able to transmit C/S signals and measurement data over a reasonable distance to the inspectors' room.
- vii) The system should provide remote live-time observation of surveillance, of identification and measurement data, as well as recording of all data for subsequent review.
- viii) The system should provide optical-electrical methods for checking integrity and identification of weld for final disposal cask.
- ix) The system data review capability should not interfere with the system when operating in data acquisition mode.
- x) The system should incorporate diagnostic checks with alarms to indicate when the system is not functioning within acceptable bounds.
- xi) The system should be fully automatic and operate unattended for a reasonable time interval between scheduled servicing.

7. Conclusion

Implementation of effective safeguards will require highly reliable safeguards systems, as reverificaiton of the nuclear material inventory in the event of any anomaly or discrepancy will be extremely difficult. A sound quality assurance programme is required to ensure the effectiveness of the applied safeguards measures.

A high degree of operator/inspector cooperation will be required to implement such a complex and advanced safeguards system. It is high time to define system specifications and to initiate development work before such facilities are built.

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SAFEGUARDS-RELEVANT DESIGN CHARACTERISTICS OF SPENT FUEL AND RADIOACTIVE WASTES IN THE PLANNED GORLEBEN REPOSITORY

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Abstract

Safeguards-relevant design characteristics of the planned Gorleben repository have been identified by a team established by the "Bundesminister für Forschung und Technologie" - Federal Minister for Research and Technology - (BMFT). These characteristics are the advanced conditioning, the reloading cell, the above-ground buffer stores for the bridging of short-term disturbances, the hoisting facilities, the mine (here especially the permanent structural change, the retreating, the exploratory level which is later to be used as return air horizon and the duration of shaft sinking) as well as the mixed emplacement of radioactive wastes and spent fuels.

Introduction

In September 1989, the Federal Minister for Research and Technology, BMFT, established a team which is supposed to elaborate suggestions for a safeguards draft concept for the direct disposal of spent nuclear fuels. Five members of the following institutions belong to this team:

- BfS (Bundesamt für Strahlenschutz, Federal Office for Radiation Protection), D-W 3320 Salzgitter
- DBE (Deutsche Gesellschaft zum Bau und Betrieb von Endlagern für Abfallstoffe mbH, German Company for the Construction and Operation of Waste Repositories), D-W 3150 Peine
- GNS (Gesellschaft für Nuklear-Service mbH, Company for Nuclear Service) D-W 3000 Hannover
- KFA (Forschungszentrum Jülich GmbH, Jülich Research Centre) D-W 5170 Jülich
- KfK (Kernforschungszentrum Karlsruhe GmbH, Karlsruhe Nuclear Research Centre) D-W 7500 Karlsruhe

BMFT has been elaborating this basic concept for IAEA (International Atomic Energy Agency) in Vienna within the framework of the Task A.14 "Safeguardss for Final Disposal of Nuclear Material in a Geological Repository" of the German supporting programme.

Safeguards-Relevant Design Characteristics

Members of the team have identified the following safeguards-relevant design characteristics for the direct disposal of spent nuclear fuels:

1. Conditioning facility

In the conditioning facility, spent fuel elements are packaged in thick-walled containers which are supposed to function as long-term barriers in the repository. This presupposes that the containers are welded with a deep-split welding seam (approximately 5 cm) around the cap. Another composite welding is applied with CASTOR-containers with HTR-fuel elements. In the conditioning facility, the nuclear material can be seen and measured for the last time, before it is packaged into the containers. From this time on, the material is regarded as inaccessible. It is not planned to open the containers later on which is mechanically not possible in the area of the repository anyhow.

2. Reloading cell

In the reloading station of the repository there is a reloading cell in which heat-generating wastes from reprocessing (HLW canisters, BT drums) are reloaded from collecting transport containers to single shielding containers. This reloading cell can be designed in such a way that the opening of the POLLUX containers and the removal of LWR fuel elements can be excluded for mechanical reasons. Furthermore, mechanical monitoring systems are possible.

3. Buffer store

An above-ground buffer store is necessary for all types of waste and spent fuel elements in order to guarantee an optimum operation, respectively to bring under control operational breakdowns. Collecting transport containers and single shielding containers with heat-generating wastes from reprocessing as well as POLLUX and CASTOR containers with spent fuel assemblies to be disposed of directly can be buffer stored there to a limited extent (for approx. 1 week). A check of identity and integrity of the buffer stored POLLUX and CASTOR containers is possible at any time (seal, surveillance by camera or TV, inspection by the inspectorates).

4. Hoisting facility

For mechanical reasons (max. hauling load), POLLUX and CASTOR containers can only be hauled in shaft 2, perhaps (investigation is being carried out) only if an additional weight is attached to the counterbalance. For mechanical reasons, the haulage of POLLUX containers without additional weight is probably excluded because of the ropes slipping through on the Koepe sheave.

In comparison to the haulage of other radioactive materials, the haulage of POLLUX containers is relatively rare: If, for example, all spent fuel assemblies arising in the Federal Republic of Germany amounting to approx. 500 t/a were disposed of directly, each year only approx. 115 POLLUX containers would have to be hauled, i. e. a little bit more than 2 containers per week.

Shaft 1 serves for the transportation of personnel (man-riding), of equipment and of salt to the surface. Possible C/S measures are control of identity and integrity at the charge of shaft 2, monitors to measure the dose output in order to control possible material backflow at the two shaft charges in case an opening of the containers below ground cannot be excluded.

With the measures taken at the hoisting facility the safeguards measures in the repository could stop.

The Federal Republic of Germany, however, is very interested in demonstrating that no nuclear material is removed. Therefore, the following, confidence-creating measures might additionally be taken on a voluntary basis without this being necessary from a safeguards-relevant viewpoint.

5. Mine

The mine shows four safeguards-relevant design characteristics: as there are the permanent structural change of the mine caused by the drivage of new and the abandonment of old emplacement fields, the emplacement by retreating, i. e. from the periphery of the emplacement horizon to the shaft, and the existence of the exploration/return air horizon. Further, the long duration of shaft-sinking is safeguards-relevant.

5.1 Verification and re-examination of the mine

Due to the permanent structural

change of the mine, the design information (physical structure of the mine) changes permanently which forms an essential part of a safeguards concept. Verification and re-examination of the mine are thus essential elements of a safeguards concept for the final disposal of spent fuel assemblies. The goal of the measures is to check periodically the correspondence of the actual physical structure of the mine with the details given by the operators to the surveillance organizations in official reports.

The construction of a repository and the emplacement of wastes and spent fuel elements presuppose the permission according to Atomic Energy Law of the competent government of the federal states (Länder) and the permission according to Mining Law of the competent mining authorities.

The plan-approval decision determines which waste packages with which characteristics and which content may be emplaced.

All works, like the drivage of drifts and rooms, the sinking of boreholes, the emplacement of wastes, the stowing of drifts and the construction of closing-off structures, are carried out according to operational plannings (general plan of operation), which are to be determined beforehand and to be individually authorized by the mining authorities, which may be announced to the inspectors of the surveillance organizations before starting the works.

The drivage of a repository mine in relation to structure and outline of the salt dome is always accompanied by geotechnical investigations temporally immediately running ahead. To a limited extent, this is also valid for the emplacement rooms, i. e. for the drivage of emplacement drifts, rooms and other subsurface openings including borehole sinking. Form and shape of the drifts, rooms and boreholes are not known exactly before drivage or sinking. They are then surveyed by the mine survey department and entered in plannings, the so-called mine map, and certified. The changes in the mine caused by the emplacement of the waste packages, the placing of stowage material and the construction of closing-off structures are also entered in the mine map and certified by the mine survey department as soon as these works are carried out. With that a relevant, certified mine map (sketch plan according to art. 63 Mining Law) is always availabe which can be used by the inspectors of the surveillance organizations for the purpose of verification and re-examination.

A verification and re-examination of the mine could then be carried out with the help of periodical inspections of the accessible parts of the mine by inspectors of the surveillance organizations, in the course of which the correspondence of the actual physical structure with the one according to the relevant mine map is checked.

This measure could possibly be supported by processes using technical equipment which have primarily been developed for geological investigations, like, for example, borehole radar or drift radar. Hereby openings including emplaced containers etc. can be determined as irregularities in the salt dome. It must be checked to what extent a representation of the current mine map can be gained with such methods.

5.2 Emplacement by retreating

The establishment of emplacement fields and the emplacement of repository waste packages is started in parts of the mine being farthest away from the shaft. Filled emplacement areas including access drifts are stowed with salt sinter and separated from the operational part of the mine. The emplacement is carried out by retreating, i. e. to the shafts. Retreating will probably be carried out regularly to the shaft in the northeastern and southwestern part of the elder rock salt, in order to gain a loading of the shafts which is symmetric to a large extent. Emplacement by retreating means that there is no access to abandoned districts and that the mine gets smaller in the course of time. Possibilities to control this: design information re-examination.

5.3 Investigation/return air horizon

Approx. 30 m above the emplacement horizon there is the exploration level which is to be used as return air horizon. Like the emplacement area, this area must also be included in the measures for the verification and re-examination of the mine. Relevant mine maps have been drawn up for this area too. The return air horizon is also operated by retreating and abandoned.

5.4 Time of shaft sinking

Because of the water bearing and the low stability of the cap rock the shaft sinking takes several years. A quick and secret sinking must be excluded. Possibilities to control this: above-ground supervision of the ground, seismic methods.

6. Mixed emplacement

Wastes from reprocessing, spent fuel elements to be disposed of directly, and other radioactive wastes are to be emplaced in the repository, i. e. a mixed emplacement of material which must be supervised and of material which needs not be supervised occurs. Material which must be supervised as well as material which needs not be supervised is disposed of in separate emplacement fields. The type of waste package and the emplacement technique used allow a clear statement whether the material must be supervised or not.

Transport containers for spent fuels are disposed of with their content. Single shielding containers for the internal transport of the wastes from the reloading cell to the emplacement field (borehole) are transported back when they are empty. Whether they are empty, can be controlled by opening them or by weighing.

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USE OF PROCESS MONITORING FOR VERIFYING FACILITY DESIGN FOR LARGE SCALE REPROCESSING PLANTS

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Abstract

During the decade of the 1990s, the International Atomic Energy Agency faces the challenge of implementing safeguards in large, new reprocessing facilities. The Agency will be involved in the design, construction, checkout, and initial operation of these new facilities to ensure effective safeguards are implemented. One aspect of the Agency involvement is in the area of design verification. The U.S. Support Program has initiated a task to develop methods for applying process data collection and validation during the cold commissioning phase of plant construction. This paper summarizes the results of this task.

1. Introduction

Design information verification is a required element for international safeguards as practiced under INFCIRC-153 /1/ for signatories to the Non-Proliferation Treaty (NPT) /2/. Paragraph 43 specifies the design information to be made available.

Proposed procedures for design verification of large reprocessing plants are reviewed in Refs. 3 and 4 and include:

1. examination and checks required for installed safeguards-relevant components [dissolver(s), feed clarification devices, plutonium evaporator];
2. identification and checks of input accountancy, plutonium product, and all other tanks in the plutonium process line, including input and output piping connections and "all other details";
3. identify recalibration procedures;
4. follow up and check the procedures and results of calibration and homogenization/sampling of the input and output tanks and all tanks where plutonium is to be measured;
5. follow up and check the procedures and results for testing and calibrating the measurement instruments, the transfer systems (including dilution and "dead volumes" involved), and the auxiliary services provided for every tank;
6. follow up and check the homogenization and sampling procedures and the calibration of measuring and monitoring instruments foreseen for each key measurement point (KMP);
7. follow up and check the homogenization and sample transfer equipment and analytical procedures for the laboratories;
8. verify the calibration of nondestructive assay instruments to Agency standards; and
9. verify the proper location, installation, and calibration of surveillance equipment and systems, and check the false-alarm rate.

Also, the inspector should experimentally test and validate process models used to assess the quantity of nuclear material present in process equipment at predetermined process conditions.

Although this represents only a partial list of suggested inspector verification activities, it is apparent that design verification is a formidable task and could require a team of knowledgeable engineers to do the job effectively and efficiently.

Most reprocessing plants in operation or under construction will have some form of process monitoring to aid the operator in reading and assessing process status for safety and process control purposes. We propose that some aspects of the process monitoring system can be used for facility design verification to reduce the inspector resources required to meet the design verification objectives and, as such, to reduce the impact of the verifications on the operator's work load.

2. Cold Commissioning Activities

The full construction project typically lasts more than 10 years, from design to hot operation. Agency interest related to design verification is concentrated in the final phases of the project. As the construction of the facility is complete, the operator enters the acceptance testing phase, where system operability is demonstrated by the constructor and the operator assumes responsibility. This starts the cold commissioning phase.

Cold commissioning requires 2–3 years and typically consists of four phases: (1) system acceptance, (2) water runs, (3) acid runs, and (4) cold uranium runs. The Agency has a role throughout this period to develop a confidence in the operator's ability to supply reliable and accurate information.

During the system acceptance phase, Agency-supplied instruments and computers are installed and checked. The Agency also has an interest in acceptance testing of operator systems related to safeguards measurements. During the water runs, the operator usually performs volume calibrations of interest to the Agency. During the acid runs, there is an opportunity to verify plant design information supplied to the Agency. Sample system checkout usually occurs during this time. The uranium runs allow evaluation of measurement systems, procedures, and inventory estimates during cold commissioning. The Agency should have a role throughout all of these activities. Process monitoring allows for the unattended collection of information to support Agency evaluation.

3. Process Monitoring and the Process Control System

Process control in modern plants involves interface of computers to instruments and control equipment while an operator sits at a console in a control room environment. Hardware and software consolidate information on the control room console to allow the operator to monitor performance and effect control over processes and equipment. To varying degrees, the computer systems contains logic structures to automatically control and adjust process parameters. Modern process control systems are really process control and information systems, often involving hierarchies of computers that consolidate information from plant processes with other information such as analytical results, and pass this information to plant support organizations and management.

Process monitoring is a term normally associated with operation and control of plant operations and the use of these modern information systems. Process monitoring has also evolved a definition associated with safeguards. In the safeguards context, process monitoring is "the use of a broad range of process data and analysis tools to make timely and sensitive judgements on the location and movement of nuclear materials," and as a corollary, "to make timely and sensitive judgements on the status and performance of measurement systems used for nuclear material accountancy" /5/.

There is need to draw a distinction between process monitoring in the process control sense and in the safeguards application. In process control, the operator monitors conditions and makes adjustments such that the control of plant processes is implied. The operator has free access to information and returns control signals based on his analysis or some level of automated response. In safeguards process monitoring, a prescribed subset of process information is automatically transferred on a routine basis, likely to a safeguards data collection and analysis computer system /6-9/. This is a one-way transmittal of information with no free access to process information. It is a limited set of information, negotiated and agreed to, to provide the required minimum safeguards information. This structure presents several advantages to both parties:

- error-free data collection and recording;
- less intrusion time on the operator;
- less time consuming;
- limited topics agreed to by the Agency and operator; and
- a reduction in the Agency and operator work load.

To develop the safeguards applications, there is a need to understand the architecture and operation of the distributed control system. Safeguards applications should make use of process control measurements and minimize the need for special, dedicated measurement systems. Safeguards process monitoring can be developed to be transparent to operations and make use of the information available within the operators' process control system.

Process monitoring should be viewed as an advanced technique to collect and validate safeguards information in support of conventional accounting and NRTA; however, it is not proposed as an advanced safeguards technique.

3.1 Distributed Control Systems.

The distributed control system comprises a number of process control modules (PCMs) that interface directly to the plant instrumentation and controls. The PCMs are usually linked to a communications module (CCM) that coordinates the flow of information from the PCMs to other modules. A central host computer is used for program development and for downloading operating software to other components including PCMs.

Modern nuclear facilities take the distributed control system used for process control and integrate it into the plant information system. A number of computer systems, for example, dedicated to destructive and nondestructive assay, are interfaced to a central laboratory system that maintains all analytical information and results. This, in turn, is interfaced to the process control system.

Safeguards becomes another node on the information system.

3.2 Agency Computer System.

An agency-owned computer (or operator-supplied computer for Agency use) can be interfaced to the host. High level communications packages are available to make the interface between Agency and host computers a rather simple task. Typical system software permits establishing the link as a "slave", allowing only structured communications. In this application, the Agency passes the message to the host to return a pre-defined minimum data set. The software package within the host puts the request on the data highway. The PCMs return the current information which is arranged by the host in the structured message for passing back to the Agency computer. The Agency computer receives and archives the "snapshot" of plant information.

The operator has control of the communication package at the host end of the safeguards link. The list of data and frequency of acquisition is negotiated. This arrangement does not allow the Agency to access additional information or in any way influence process control.

The Agency is responsible for the data reduction and analysis software in the Agency computer system. Any Agency-owned measurement equipment should also be interfaced to this machine. It is important for the Agency to maintain contact with the operator through the design and construction of the facility to ensure compatibility of systems in the final installation.

3.3 Process Description

The process considered for this study is based on equipment and instrumentation designed, installed, and tested at Oak Ridge in the Integrated Engineering Test (IET) facility /5/ and at the Barnwell facility /10/. The input accountancy tank and associated equipment including the process monitoring capability are shown in Fig. 1. The accountancy tank is fed from the dissolver surge tank. Typical process monitoring instrumentation is discussed in Section 7 of this paper.

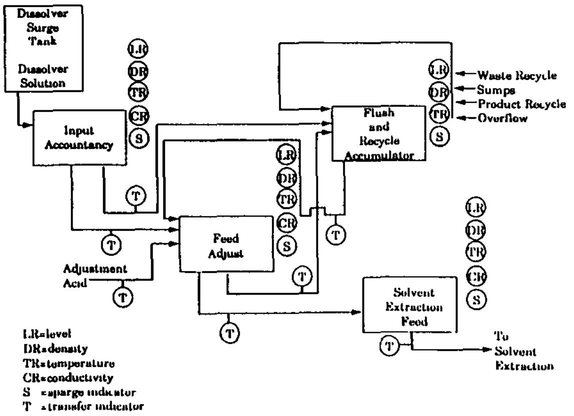

Fig. 1. Accountancy/feed adjust system.

4. Data Authentication

Agency use of process monitoring information requires some means of authenticating the data because process monitoring relies on use of information obtained from the operator's systems. Authentication has been defined in one IAEA paper /11/ as "the technique used to assure that valid results have been obtained for safeguards purposes using operator-provided equipment." Process monitoring, and potential applications of NRTA, involve the use of data supplied to the agency through the operator's computerized process control system. However, scenarios develop that involve falsification of data, either through data tampering, instrument tampering, or materials tampering.

Concerns on data authentication, when considering safeguards applications for process monitoring and the use of data from the operator's process control system, include authentication and maintaining continuity of knowledge on:

- pneumatic instrument lines;
- instruments (calibration settings);
- instrument (electrical) lines to computer interface;
- computer software;
- computer to computer communications; and
- off-site transmission of information.

Process monitoring provides the continuity of knowledge which has application for authentication of other measurements. At several locations in a reprocessing plant, increases or decreases in tank volumes can be related to a particular flow measurement or other flow indicator. Undeclared additions or falsification of data within a continuous recording system will be reflected in discrete changes without corresponding indicators.

The cold commissioning phase of plant operation is the time for the Agency to establish authentication mechanisms. During the early stages of this phase, the Agency will deliver computer equipment and other measurement systems for installation and interface to the operator's control and information system.

Part of the normal installation process is to develop an acceptance test procedure that tests and documents performance of the final system to prescribed standards. During the testing, each piece of information delivered through the software is checked to the actual instrument output, whether read from the operator console or directly from the instrument. These tests form the basis for routine checks during later operations that can be random in a typical inspection mode.

During the later phases of the cold commissioning, specific tests conducted in conjunction with the operator's facility acceptance tests establish many of the other comparative techniques that provide authentication.

In summary, plant process information is, to an extent, "self authenticating." In a complex chemical operation like a reprocessing plant, the processes are interactive and virtually all measurements are related to some other activity indicated by measurements. These comparisons form the basis for authentication to allow use of process information without the burden of many of the computer security techniques and concerns applied to other computer information systems. Checkout and demonstration of these techniques is an important part of the cold commissioning activities.

5. Diversion Scenarios

Scenarios for diverting plutonium from the process area that are of concern during design verification include the following /12/:

1. transferral of undeclared dissolved fuel solution to the process area bypassing the input accountancy tank and removal from the process area;
2. removal of plutonium nitrate from the input accountancy tank or from other points in the process area;
3. removal of concentrated plutonium nitrate solution from the product accountancy tank;
4. transferral of undeclared dissolved fuel solution to the process area, bypassing the input accountancy tank, and transferral to the nitrate storage area, bypassing the output accountancy tank;
5. transferral of plutonium nitrate solution from the process area to the nitrate storage area, bypassing the product accountancy tank; and
6. removal of liquid waste containing plutonium.

Scenarios 1, 4, 5, and 6 require bypass of one or more key measurement points (KMPs) (the waste accountancy tank is considered a KMP). Scenarios 2 and 3 require undeclared pipes from the process area or making use of abnormal routings (e.g., sampling lines).

Therefore, design verification activities should address bypass of KMPs, detection of undeclared pipework, and use of abnormal routings.

6. Description of Inspection Activities

Inspection activities during cold commissioning should address design verification and recognize the need to collect information to support safeguards needs during hot operation of the facility.

Specific Agency activities using process monitoring proposed for the period of cold commissioning address the following:

1. transfer routes,
2. in-process inventory,
3. in-process holdup,
4. volume calibration and recalibration, and
5. mixing and sampler tests.

A minimum data set negotiated for automatic transfer to an Agency computer system includes level, density, temperature, and flow measurements as well as status indicators on pump, steam jet, and air lift actuators. Part of the design verification activities during cold commissioning is verification and validation of measurements and information provided in this minimum data set.

The use of process data for Agency safeguards is always a sensitive issue. From the operator's perspective, process activities can have a broad and complex effect on measurement system performance. The operator recognizes the opportunity to misinterpret process information and offers that intrusiveness is an issue in providing such information to the Agency. It is extremely important to establish procedures for analysis of process information and implement these in software within the Agency System. Cold commissioning offers the opportunity to implement and demonstrate these procedures outside of concerns during processing of plutonium during routine plant operations.

6.1 Transfer Routes.

Bypass of key measurement points is a concern expressed in several of the diversion scenarios presented in Section 5. Processing equipment and transfer routes are identified in design information typically requested and provided to the Agency. Process monitoring information transmitted to the Agency computer system can be used to identify transfer routes for verification of design information and to maintain continuity of knowledge over those routings which were subject to physical design information verification (DIV). Data analysis routines developed and implemented during cold commissioning can be the basis for a program to analyze a similar set of information provided during routine operation for continued assurances against activities that bypass accountancy measurement points.

Tank transfer devices are typically sized during design to accomplish a full tank transfer over a period of 15-30 minutes. Process monitoring information transmitted to an Agency computer system at frequencies on the order of 5 minutes provide a continuity of information and several points during a transfer for analysis.

6.2 In-Process Inventory

In-process inventory resides in tanks, contactors, concentrators, and associated piping. For NRTA applications during plant operation, it will be necessary for the inspector to measure (or estimate) this inventory. During cold commissioning activities, it may be possible to use process monitoring to verify transfer of nuclear material between adjacent tanks as described in Section 6.1 and to use the information to validate techniques to provide information for NRTA. A number of techniques have been proposed for estimation of holdup in contactors. Past efforts in application of NRTA for international safeguards have been primarily on a plant that uses a batch evaporator for plutonium product concentration. Inventories are typically timed to concentrator dumps. Modern plants will have continuous concentrators and activities during cold commissioning will have to address estimator methods for these concentrators.

6.3 In-Process Holdup.

In-process holdup is the material that remains in process vessels, pipework, and equipment after the process material and stored materials have been removed. During the cold commissioning of the facility, the quantity of material retained can be estimated using by-difference principles once the proper transfer of material through the facility has been verified. Material measured in one vessel should equal that determined in the next vessel if no material is held up. Differences between the two assay results can then be attributed to material retained in that phase of the process in order to complete the mass flow balance. Upon the completion of the commissioning activities, a model can be developed using this information to identify and aid estimations of material that would be retained during normal processing operation.

6.4 Tank Volume Calibration.

Tank calibration is a significant part of the cold commissioning activities because the operator needs volume measurements for operational control as well as to satisfy safeguards requirements.

Inspector resources are not adequate to witness calibration activities for every vessel important to safeguards, which includes all tanks and equipment measured for inventory. However, the operator must go through the procedure for each tank and must record the information and develop the relationships for each tank as a calibration package. As a minimum, the calibration relationships must be available to the Agency, together with the calibration package that includes the basic information for review.

6.5 Solution Mixing in Tanks

Adequate mixing of solutions in accountancy tanks is necessary to assure that homogeneous solutions are available in the tanks for sampling to assure that the quantity of material contained in the tanks is accounted for. Adequate mixing also is important to facility operations to ensure process control information is representative and the homogeneous solutions are presented to process systems. During cold commissioning, the facility operator will conduct a series of tests of mixing equipment and will establish mixing procedures for equipment. The Agency is concerned to verify mixing tests to establish optimum mixing procedures to assure proper accountancy measurements for key measurement points. In addition there is an interest in mixing requirements for most process equipment in support of inventory measurements for conventional accounting and NRTA.

It can be expected that the operator will provide the results of mixing tests to the Agency for tanks that are important for safeguards applications. The actual testing will be carried out over a period of time during cold commissioning. The Agency likely cannot devote inspector resources to validate all of the tests, but can use process monitoring to validate the test information provided by the operator.

6.6 Sampler Tests.

The operator is interested in establishing the minimum recirculation time/volume through the sampler feed line to ensure that the sample is representative of the solution in the tank, and to ensure that the sampling method does not induce any bias in the sample.

A test that is effective is derived from the mixing test. Assuming a homogeneous solution in the tank or process line (after the mixing test), the operator backflushes the sampler system with a solution of significantly different density. The operator then initiates recirculation and pulls a sequence of samples. The samples are analyzed for density (density being used for the reasons given in Section 6.5) until sequential samples agree within the accuracy of the method. As a "rule of thumb", the total volume of the sampler feed line should be replaced at least five times.

As with the mixing tests, the Agency can validate the information on the tests provided by the operator by comparison to process monitoring information. The Agency again reduces inspector resource requirements to validate the tests by using the comparisons to information automatically recorded by the process monitoring system.

7. Minimum Data Set

International safeguards applied to the large scale reprocessing plants of the future place new demands on information transfer and management. For large scale reprocessing plants, conventional material balance accounting will likely be supplemented by some form of NRTA.

Activities during cold commissioning should be directed at validation of measurement systems associated with safeguards applications, including verification that design information is complete and accurate. Process monitoring is offered as a technique to accomplish this validation. Process monitoring information includes the same set of information required for the conventional accounting, NRTA, and RBI applications, but records that data set at frequencies that provide a continuity of information for assessment of measurements.

The Input Accountancy Tank is one of the most important key measurement points associated with conventional accounting. Verification of measurements at this point is a focus of Agency inspection activities. Tanks adjacent to the accountancy tank are major surge locations and are important to inventories in conventional accounting and to in-process inventories under NRTA. Process monitoring applications proposed during cold commissioning are described in Section 6. These provide validation of the measurement systems to support conventional accounting and NRTA and are directed at design verification in response to diversion scenarios associated with by-pass of the accountancy measurement point.

Table I lists the measurements available within the process control system that are of potential interest to safeguards. The table shows the minimum data set needed to meet the requirements of process monitoring during cold commissioning, and relates that set to the measurements needed to support conventional accounting and NRTA. The measurements needed during cold commissioning include those necessary for all the activities identified in Section 6.

TABLE I. Process Control Measurements of Interest to Safeguards

		Conventional Accounting		NRTA	
	Cold Commissioning Process Monitoring	I/O Accounting	Inventory	I/O Accounting	Inventory
Surge Tank					
Level	X		X		
Density	X		X		
Temperature	X		X		
Sparge	X		X		
TI (to acc)	X		–		
TI (to recycle)	X		–		
Accountancy					
Level	X	X	X	X	X
Density	X	X	X	X	X
Temperature	X	X	X	X	X
Conductivity	X	–	–	–	–
Sparge	X	X	X	X	X
TI (to adjust)	X	–	–	–	–
TI (to accum)	X	–	–	–	–
Sampling indicator	X	X	X	X	X
Feed Adjust					
Level	X		X		X
Density	X		X		X
Temperature	X		X		X
Conductivity	X		–		–
Sparge	X		X		X
Acid add indicator	X		–		–
TI (to feed)	X		–		–
TI (to accum)	X		–		–
Sampling indicator	X	X	X	X	X
Accumulator					
Level	X		X		X
Density	X		X		X
Temperature	X		X		X
Sparge	X		X		X
TI (to adjust)	X		–		–
Sampling indicator	X	X	X	X	X
Feed					
Level	X		X		X
Density	X		X		X
Temperature	X		X		X
Conductivity	X		–		–
Sparge	X		X		X
TI (to sol. ext.)	X		–		–
TI (to adjust)	X		–		–
Sampling indicator	X	X	X	X	X

X = needed; – = useful

For both conventional accounting and NRTA, each accountancy tank batch must be measured for input/output accounting. The chart indicates that status indicators are "useful", but not necessary for accountancy measurements since part of an effective measurement procedure is to verify that solutions are not moving into or out of a tank during measurement. These measurements must be transmitted and recorded for each accountancy batch, which is typically once per day in a large scale facility.

Inventory requirements for both conventional accounting and NRTA add the necessity of volume measurements for the additional tanks in the feed preparation area of the plant. The table shows that measurements for the surge tank are necessary for conventional accounting but not NRTA. This is because the annual shutdown inventory generally tries to close the balance to the pool storage area, including the dissolver/head end area. There is a necessity to ensure that the head end area has been flushed prior to inventory. In NRTA, the balance is closed from input to output accountancy tanks and the surge tank is not in the area.

The in-process inventories to support NRTA will likely occur on intervals of a month or less. As indicated in the table, essentially the same information required for conventional accounting inventories is required for these NRTA inventories. Thus, the same basic data set required for conventional accounting is required on frequencies of a month or less to support NRTA.

The data sets for conventional and in-process inventories show transfer indicators and conductivity measurements as "useful". Conductivity is the direct measurement of the electrical conductivity of the process solution and an indirect measure of acid concentration. In concert with the density measurement, this offers the operator a process control estimate of heavy metal concentration. In the safeguards applications, these measurements can help validate sample information. For the cold commissioning process monitoring applications, during the uranium run phase, the density-conductivity relationships are accurate measures of heavy metal and acid concentrations that are useful in the transfer tests and validation exercises discussed in Section 6.

The transfer indicators are indicated as necessary for process monitoring applications and useful for inventory measurements. In all cases, the transfer indicators only provide backup of indications of transfers. As shown in the data evaluations presented as part of the discussion in Section 6, these indicators also require some interpretation and are useful based on the interval between data set transmissions. Even in the process monitoring applications during cold commissioning, these indicators add additional credibility to the decisions based on the volume changes alone. There has been some work /13/ on evaluation of a data set similar to that which would be recorded for the feed preparation area, without the transfer indicators. This work attempted to evaluate transfer routes without any design information. Presumably, the Agency will start from the position of having design information and its verification as a rule base for evaluation of transfers based on declared routes. The transfer indicators are also not absolutely necessary for process monitoring.

The discussion in this section presents an assessment of the minimum data set required for process monitoring applications applied to the feed preparation area during cold commissioning. The data set is essentially the same as that required for conventional accounting and NRTA with the differences in the frequencies that the data set must be transmitted. The data is required frequently, on intervals of 5-15 minutes, for process monitoring. The data collection activities during cold commissioning can help establish the link to be used during routine plant operation.

8. Conclusions and Recommendations

Process monitoring is a term that describes the use of information collected from the operator's process control system for safeguards purposes. It should be viewed as an advanced technique to collect and validate safeguards information in support of conventional accounting and NRTA; however, process monitoring is not proposed as an advanced safeguards technique.

Process monitoring has been defined as the broad use of process data to provide a continuity of information on the location and movement of nuclear materials and a continuity of information on the performance of safeguards-related measurement systems. Process monitoring implies a routine and automated collection of a specified data set from the control system and transmittal to a safeguards computer system for storage and evaluation. Process monitoring has a number of applications during the cold commissioning phase of reprocessing plant operation that can substantially reduce the Agency inspector resources that must be devoted to design verification. It provides the inspectorate with the continuity of knowledge of design information during plant operation and reduces efforts to validate measurement systems to meet the international safeguards requirements during hot plant operation.

Process monitoring can be used to collect and evaluate data on tank transfers that can verify the design information provided to the Agency. This is an important step to ensure that all equipment typically containing significant quantities of plutonium are considered in the design of conventional accounting and NRTA. Procedures that consider all equipment must be established to record inventory information for both safeguards activities. In addition, there is a need to identify all routine transfer routes for nuclear materials to ensure that accountancy points are not by-passed, that material has not been measured more than once, or that borrowing of material during inventories would be detected.

Additionally, there are a number of activities during cold commissioning that are directed at setup and testing of measurement systems that will be used by safeguards during hot plant operation. These include vessel volume calibration activities and tests to qualify mixing and sampling systems. These tests and procedures are typically scheduled throughout the cold commissioning operations. Direct validation by the Agency requires considerable inspector resources. Process monitoring offers the opportunity to automatically collect information to validate these test results without direct inspector involvement, thus saving Agency and operator resources, and avoiding data transcription errors.

Many of the process monitoring tests and activities were developed and tested to a limited degree during activities at the Barnwell Nuclear Fuel Plant /10/ and more recently in tests at the Integrated Equipment Test facility at Oak Ridge National Laboratory /7,8/. Initial results were favorable. However, additional testing and development involving Agency personnel is desirable to prepare specific methods to be transported to future plants.

Since the Barnwell and Oak Ridge activities, there have been advances in application of intelligent software systems to the data analysis portion of the evaluations. Preliminary work /14/ was done in this area by Los Alamos National Laboratory using historical data sets from one of the United States Department of Energy reprocessing facilities. Additional testing and development of this concept should be undertaken in conjunction with Agency personnel, potentially at the Oak Ridge facility during upcoming test runs, or at a suitable Agency test facility.

As the Agency progresses towards application of international safeguards in modern large scale reprocessing facilities, there should be consideration given to methods of automated data collection and handling to support safeguards. This may or may not include process monitoring during cold commissioning. However, the Agency will have to consider incorporating their need for data processing and computers in the facility design. The Agency will have to provide specifications for hardware and software to be incorporated in the facility. Recognizing the similarity of the basic data sets required by conventional accounting, NRTA and process monitoring applications for cold commissioning, there should be an effort to refine this set as facility design information becomes available and pursue definition of appropriate specifications for the facilities involved.

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EVALUATION OF INPUT ACCOUNTANCY TANK CALIBRATIONS AT WAK SINCE 1982

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Abstract

For reverification purposes the input accountancy tank at WAK has to be recalibrated every second year. Since 1982 a very precise electromanometer is used in addition to a conventional analog manometer. Six recalibration runs have been evaluated — differences between runs are analyzed and the uncertainty of subsequent volume determinations is calculated.

1 Introduction

At any nuclear power plant the contents of Plutonium in spent fuel elements can only be calculated. For the first time in the fuel cycle this contents will be measured after dissolution in a reprocessing plant. Therefore this input measurement is of great relevance to safeguards. Input measurement at WAK pilot reprocessing plant is done by volume and concentration measurement. The concentrations of U and Pu as well as their isotopes are verified to 100% by taking samples of each input batch and independant determination by EURATOM and IAEA. In contrast to this situation a verification of volume measurements is only possible by observing calibrations and input volume readings. For verification purposes every second year a recalibration of the Input Accountancy Tank is performed. Since 1982 WAK has installed an identical set of precision manometers for level and density measurements. This paper will describe a statistical evaluation of all recalibrations between 1982 and 1990.

2 Input Accountancy Tank at WAK and its environment

The accountancy tank of WAK is a 800ℓ annular vessel with cooling shell. Fig. 1 shows a schematic drawing. The tank is equipped with pneumercators for level and density measurements, a thermocouple for temperature measurements, and a sampling line. The differential pressures in the pneumercators are measured by a *Wallace & Tiernan* analog pressure gauge and — in parallel — a *Ruska RT6000* electromanometer.

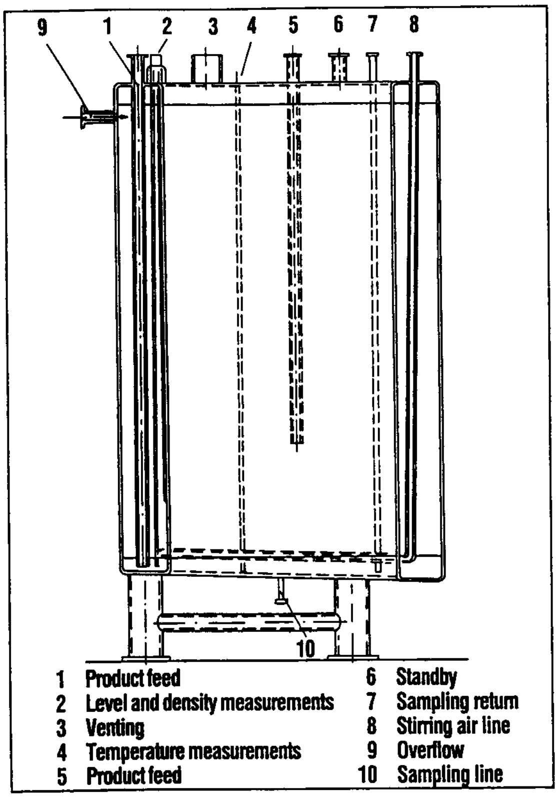

Figure 1: WAK Input Accountancy Tank

The chemical headend area (from the dissolver to the feed tank) is shown in fig. 2. Here the environment of the input accountancy tank can be seen.

For calibrations the input tank is rinsed with water and emptied. Then fully deionized water of about room temperature is weighed and poured directly into the tank using increments of approximatly 8 kg, therefore a regression function is derived from more than 100 measuring points. All calibration runs are normalized to 20 degrees (Celsius).

During hot operation pieces of the fuel rods are dis-

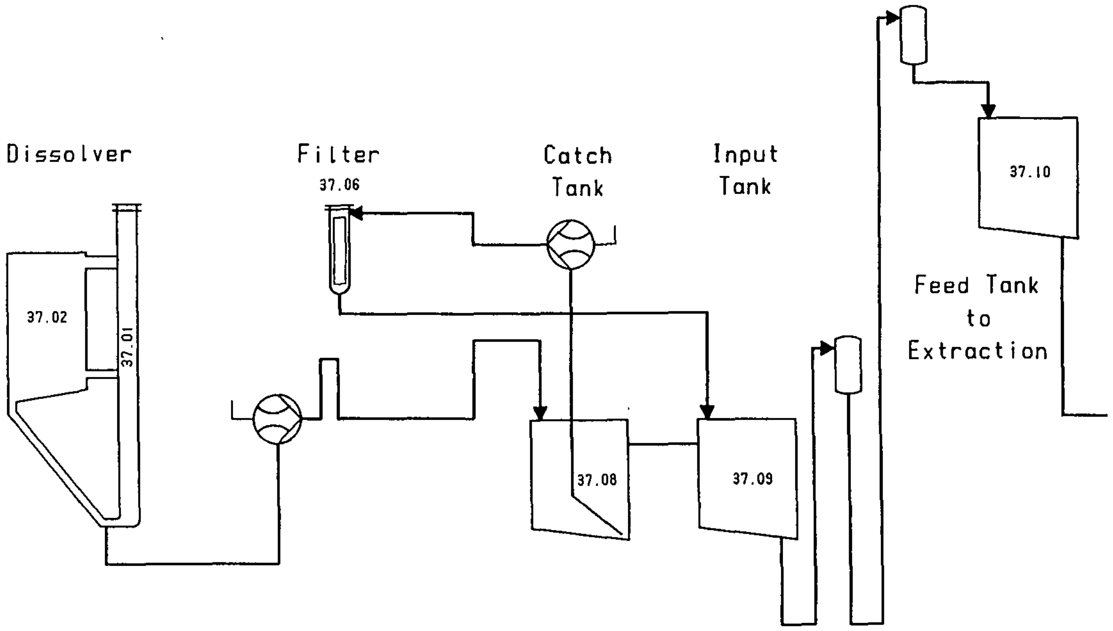

Figure 2: simplified schematics of WAK headend

solved in the dissolver (37.01), transferred by steam jet to a catch tank (37.08), and transferred by steam jet over the filter (37.06) to the input accountancy tank (37.09). Here the volume is determined and samples are taken. Then the feed solution is adjusted to extraction conditions and transferred subsequently by airlift to the feed tank (37.10), so that this vessel — in normal operation — will never become empty. Rework solutions enter the first extraction cycle on the same way. Therefore it is necessary to empty the input accountancy tank completely. This means that the incremental calibration with water must be representative for the feed volume transferred to extraction. Not only the slope but also the intercept of the calibration curve must be constant during a recalibration interval.

3 Observations during calibrations

Graphical data analysis shows that even very smooth calibration curves differ in intercept and slightly in slope. To point out this situation we plot the residuals of these curves in relation to a common regression line. In fig. 3 three subsequent calibrations, monitored with the Ruska manometer, can be seen. Two of them are performed within one week, the third one approximately two years later. We assumed that the major effect should be a difference in the heel volume.

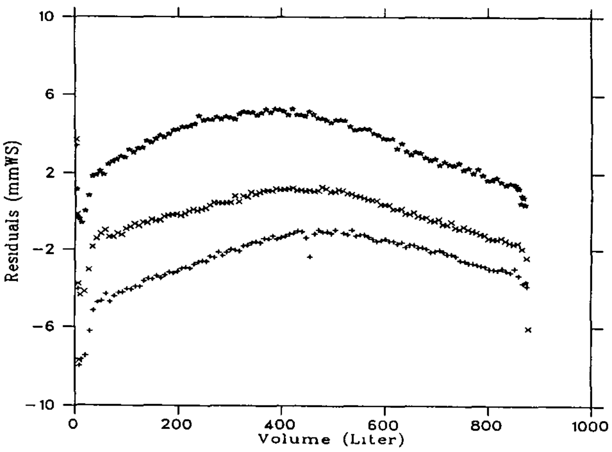

Figure 3: Residuals for a common linear regression function of *Ruska*-readings from last three calibration runs

Because it was impossible to confirm this suspicion at our hot input tank, we built a mock-up. These experiments showed [1], that the remaining heel in the transfer line from 37.09 to 37.10 is nearly constant (±2.5%) and influences the intercept variations only by $\pm 0.12\ell$. This value is small compared to the variations observed between different calibration runs.

Furthermore the different intercept variations between the two manometers at two calibrations gives indication for an instrumental reason. Up to now we also could not identify reasons for variation of slope, but — as the slopes differ systematically between the manometers at each calibration — they should be caused by the instruments and/or their specific installation.

All these effects are quite small (< 1%) and their reasons might be never detected. Therefore we decided to treat all calibration data of each manometer as one unit and to evaluate them statistically.

4 The data evaluation procedure

The experiments with the mock-up of the input accountancy tank described above give a strong indication that starting conditions are identical for all calibration runs. Therefore no alignment of calibration data can be performed, the whole run-to-run differences must be treated statistically.

It can obviously be seen (cf. fig. 3) that the run-to-run differences are significant. Standard statistical procedures usually ignore these differences and would lead to a serious underestimation of measurement variability. Liebetrau et al. [2] worked out a procedure that takes run-to-run differences into account for variance estimates of the calibration function. Together with tolerance intervals for liquid height measurements uncertainty intervals can be given for volume determination during input accounting. This evaluation procedure has been applied to the calibration runs of WAK input accountancy tank.

Selection of a calibration function We assume — as is done mostly for calibration exercises of this kind — that the error of volume determination is small compared with the error of liquid height measurement. Therefore we use the volume as the independent and the liquid height determination as the dependent variable for calculating the calibration functions. These calibration functions must be inverted for volume determination from observed liquid height measurements.

For calibration purposes the tank has been divided into five regions and linear regression functions have been applied to each region. The region boundaries have been selected from calibration profile plots, which show the residuals against a linear regression over the whole tank. The calibration profiles indicate a deformation of the cylindrical tank walls, but this deformation seems to be stable since the first calibration runs in the early 1970's.

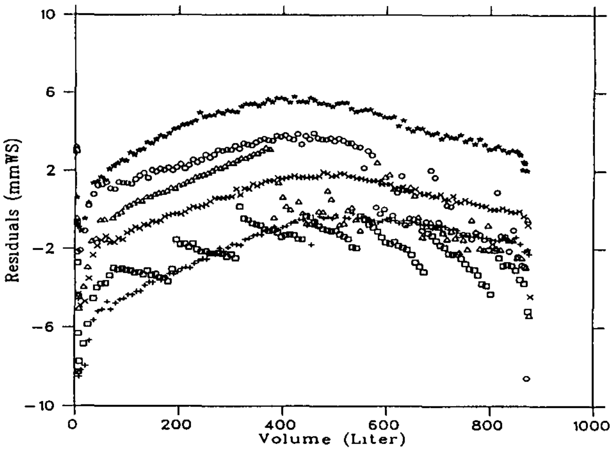

Figure 4: profile plots *Ruska*

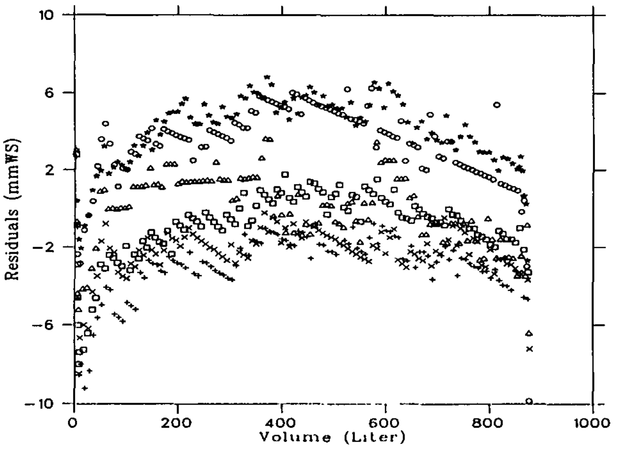

Figure 5: profile plots *W&T*

Unfortunately some calibration measurements evaluated here show disturbances (see fig. 4, 5). Some disturbances are common to both manometers (calibration □), others are specific to one instrument or show a different pattern of disturbances. Former calibrations (before 1982) have been performed with a less precise manometer. Comparing the residuals sum of squares with previous calibrations therefore gave no indication for distorted measurements. As will be seen later in this paper the effect on the overall uncertainty of volume determination is only a minor one.

We have tested, if a group of linear functions with constant slope for all calibration runs but different intercepts is an adequate calibration function. A lack-of-fit test against individual linear functions showed that the slope differences between calibration runs are significant. The different slopes are significant for all five regions.

Variance estimates An estimate of liquid height measurements includes two components: variance σ_M due to the functional form of the calibration function and variance σ_R due to measurement differences between calibration runs. The total variance for linear regression functions is

$$\sigma^2(Y|X_0) = \sigma_M^2 \left[\frac{1}{nr} + \frac{(X_0 - \bar{X})^2}{\sum_{i,j}(X_{ij} - \bar{X})^2}\right] + \sigma_R^2 \quad (1)$$

Here $\sigma^2(Y|X_0)$ denotes the variance of the level measurement Y taken at volume X_0, nr is the total number of measurement points of all runs and X_{ij} is the volume filled in at the i^{th} measurement point of the j^{th} calibration run.

The variance estimators depend on the regression model which is adequate to the calibration data — a single function, parallel functions or individual functions for all calibration runs. A detailed description of the component variances for the three possible regression models is given in Liebetrau et al. [2]. Because a lack of parallelism contributes significantly to the run-to-run differences, individual regression functions and their appropriate variance estimators have been used.

From the variance estimates confidence intervals can be derived to show where the true curve may lie relative to the fitted curve within a specified confidence $(1 - \alpha)$. For this purpose Scheffé-type confidence intervals are used.[3]

$$W(X)_{\text{Scheffé}} = \pm\sqrt{2F_{2,\nu}^{\alpha/2}}\,\sigma(Y|X) \quad (2)$$

The parameters of the F-distribution are the number of parameters of the regression function — 2 for linear functions — and the degrees of freedom of the overall variance of liquid height measurement.

Uncertainty intervals for volume determination Prediction intervals that hold simultaneously with specified propability for an indeterminate number of volume determinations can be calculated by combining the confidence intervals for the calibration functions with the tolerance interval for liquid level measurements (see fig. 6).

The liquid level measurements for input accounting are done the same way as calibration measurements. Therefore the variance of these measurements can be estimated from the calibration data by using the within-run differences.

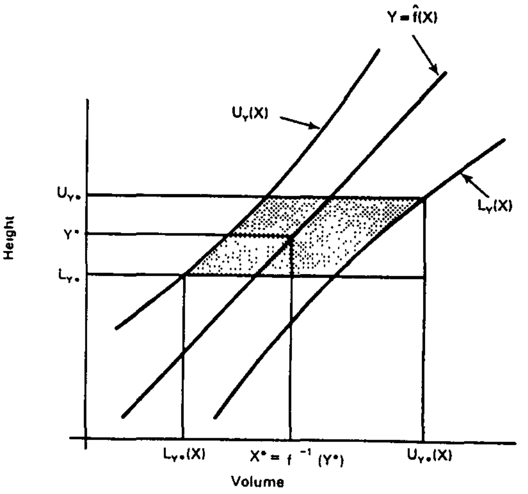

Figure 6: Determination of discrimination intervals

In the case of linear regression functions the equation

$$\begin{aligned} b_0 + b_1 X \quad &\mp \quad \sqrt{2F_{2,\nu}^{\alpha/2}} \\ &\times \quad \sqrt{S_M^2\left(\frac{1}{nr} + \frac{(X - \bar{X})^2}{\sum\limits_{i,j}(X_{ij} - \bar{X})^2}\right) + S_R^2} \\ = Y^\star \quad &\pm \quad z_{1-p/2}\sqrt{\frac{\nu'}{{}^{\alpha/2}\chi_{\nu'}^2}}\,S' \qquad (3) \end{aligned}$$

must be solved to obtain the upper and lower limits of the discrimination interval for Volume X at a given liquid height measurement $Y^\star$. The left hand side of the equation is the confidence interval for the used set of calibration runs, the right hand side is the tolerance interval for liquid level measurements — z and χ^2 being the normal and chi-square distribution, respectively. The variance estimates S' for liquid level measurements are set equal with the within-run variance estimates from the calibration runs $(S' = S_M)$. With this setting the degree of freedom ν' of liquid level measurements is set equal to the degree of freedom ν_M of the regression function.

5 Results

The evaluation of calibration data has been performed in parallel for the *Wallace & Tiernan* analog pressure gauge and the *Ruska* electromanometer, because the precisions of both instruments differ about one order of magnitude.

Evaluation of calibration profiles The six calibration runs evaluated here can be separated into

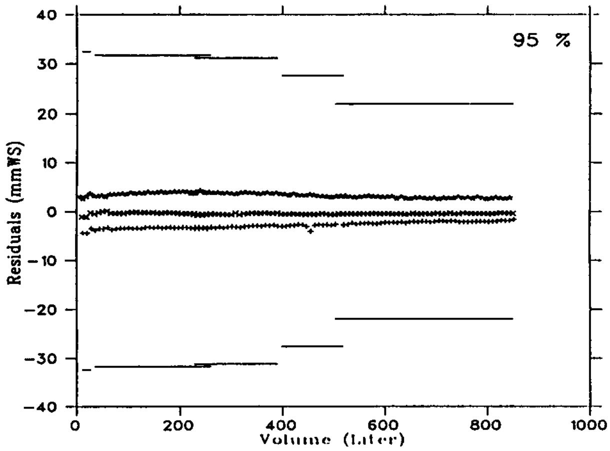

Figure 7: 95% confidence regions for three "good" calibration runs (*Ruska*)

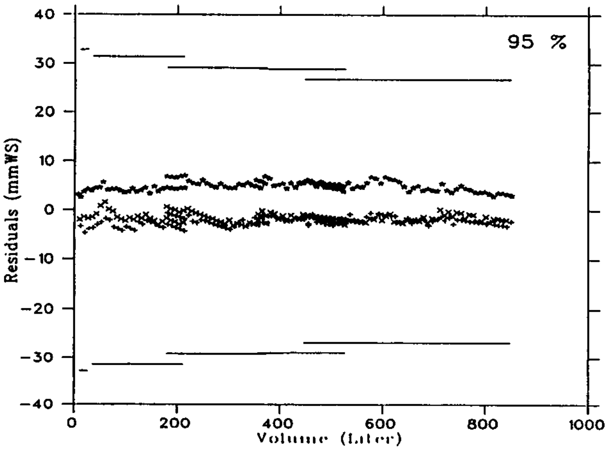

Figure 9: 95% confidence regions for three "good" calibration runs (*W & T*)

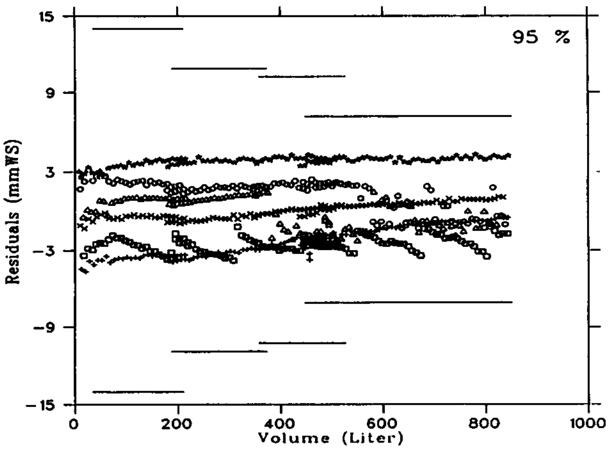

Figure 8: 95% confidence regions for all calibration runs (*Ruska*)

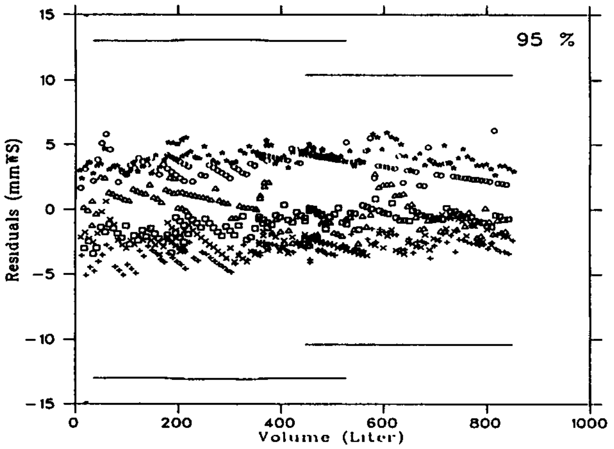

Figure 10: 95% confidence regions for all calibration runs (*W & T*)

three runs with only small within-run variances (see fig. 3) and three other ones with greater variances at least over a part of the calibration range. For this reason we investigated the three "good" calibration runs separately.

The three calibrations with small within-run variances show the greatest intercept differences (see figs. 3, 4). Therefore the effect of between-run differences is dominating the overall variance and leads to much wider confidence regions. The figures 7 and 8 show the 95% confidence regions together with the residuals of region-specific linear regressions common to all investigated calibration runs. In the most interesting range of typical input batch volumes (> 500ℓ) the 95% confidence region for the *Ruska*-manometer is about $\pm 7mm$ for all calibration runs and $\pm 22mm$ for the three runs with small within-run variations. For the less precise *Wallace & Tiernan* pressure gauge confidence intervals of $\pm 11mm$ and $\pm 28mm$ have been achieved (see figs. 9, 10).

Tolerance intervals for liquid level measurements The 95% tolerance intervals for liquid level measurements are given in table 2. It can be seen, that one can take advantage from the higher precision of the *Ruska*-manometer only when the calibration measurements are undistorted. For the *Wallace & Tiernan* pressure gauge the tolerance intervals are only slightly increased when measurements are distorted.

As the source of measurement distortion could not be identified, it seems adequate to assume that also

	W & T	%	*Ruska*	%
all measurements	±6.7ℓ	±0.95%	±4.7ℓ	±0.67%
"good" measurements	±15.2ℓ	±1.90%	±11.8ℓ	±1.48%
all measurements assumed to be good ones	±6.5ℓ	±0.92%	±3.9ℓ	±0.56%

Table 1: Uncertainties for an indeterminate number of volume determinations

	W & T	*Ruska*
"good" measurements	1.7mm	0.3mm
all measurements	2.1mm	1.65mm

Table 2: Tolerance intervals for liquid level measurements

some level measurements for input accounting have been disturbed. Therefore the wider tolerance intervals are a good approximation for typical plant operation conditions.

Uncertainty of volume determinations Solving eq. 3 for the calibration confidence limits and tolerance intervals for level measurements presented above, we obtain the uncertainties for volume determinations based on the investigated calibrations. Table 1 lists these uncertainties for the volume range of typical input batch sizes (> 500ℓ). Strictly spoken, there is a propability of 95%, that for 98% of the volume determinations made with these regressions the true volume lies within the discrimination interval. Despite their ragged profile the calibration runs with great within-run variances are valuable because the uncertainty of volume determination can be reduced by more than 50%.

The last row of table 1 shows hypothetical values. These uncertainties for volume determination would have been obtained, if all calibration runs had as small within-run variations as the last three ones. From these figures it can be seen that the uncertainty of volume determination is dominated by the large intercept variations. For improvement of this uncertainty most effort should be made to get an explanation for the intercept variations. Only with mostly reduced intercept variations the use of the *Ruska* electromanometer for accounting purposes would be justified by very small uncertainties of volume determination.

6 Conclusion and outlook

Derived from our observations and calculations we can state that a routine volume determination at the input accountancy tank of WAK had an uncertainty of less than 1% with only a small advantage for the *Ruska* electromanometer. This result influences the uncertainty of the accountancy system noticeably. In contrast to analytical procedures an *ESARDA* target value for volume determinations is missing. Therefore we propose a compilation and evaluation of data (from experiments like *RITCEX* and *CALDEX* as well as from investigations like this one) according to the method described by Liebetrau et al. [2]. This work will lead to a realistic and achievable target value.

On the other hand we will try to clarify the WAK situation by doing some additional experiments in the near future.

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EVALUATION OF TANK THERMAL EXPANSION DATA IN CALDEX

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ABSTRACT:

A thermal expansion test involving a large annular input reprocessing tank was carried out as part of the CALDEX Project at the TEKO test facility in Karlsruhe, FRG. The objective of this test was to investigate thermal expansion properties of the tank and effects on various pressure and level measurement instruments used in the determination of liquid volume. In the thermal expansion test, a weak nitric acid solution was heated internally to a temperature of 60°C by means of steam injection through the sparge ring. After heating, the annular tank took about one hour to thermally equilibrate, and it took another hour for the sparge ring and pulsator pipes to fill before thermal effects could be followed. The temperature at the end of the test, after tank and its contents had cooled undisturbed for fifty hours, was 29.9°C. Thirteen instrument readings were obtained during each measurement cycle of roughly 70 seconds for a total of over 2800 readings per instrument. Thermal expansion effects for the CALDEX annular tank were consistent with that reported for cylindrical tanks. Temperature variations effect each type of probe in a way that depends on the properties of the probe and the characteristics of the measurement system.

1. INTRODUCTION

In the framework of the ESARDA Working Group for Reprocessing Input Verification, member and observer organizations were invited to participate in the Calibration Demonstration Exercise (CALDEX) organized by DWK, Hannover, Germany /1/. Included among the specialized tests proposed was a test to investigate thermal expansion properties of the tank and the effects on various pressure and level measurement instruments /2/. The test was designed so that correlated data involving the observed liquid height, the tank weight, the dimensional expansion of the tank, and the density and liquid height, would be used to examine the application of temperature correction factors and methodology. Experimental studies involving bubble-probe manometry in cylindrical tanks have been carried out previously /3/, but experimental data for annular tanks and acoustic and capacitance probes are lacking.

2. TANK

A schematic diagram of the CALDEX tank is shown in Figure 1. The tank is 4.2 meters high and has a diameter of 2.88 meters. The annulus of the tank has a radius of 400 mm and a capacity of 12500 liters. The change in tank height due to the slope of the bottom of the tank is 150 mm.

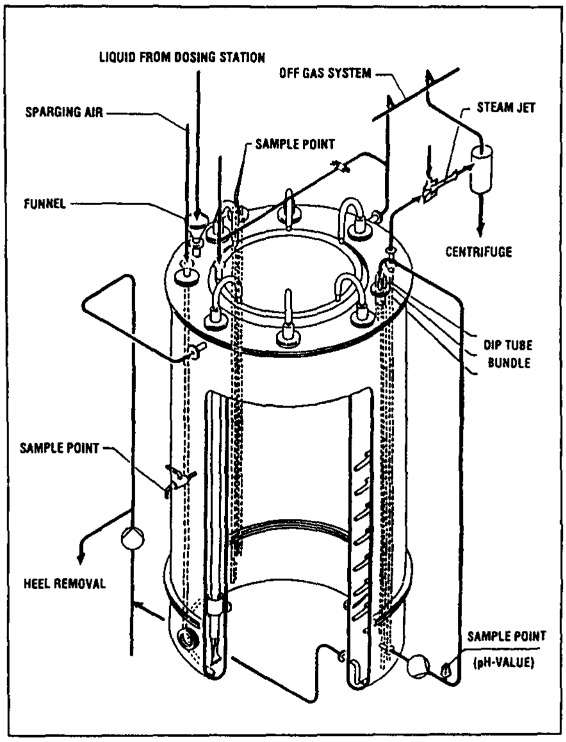


Figure 1. CALDEX Accountancy Tank

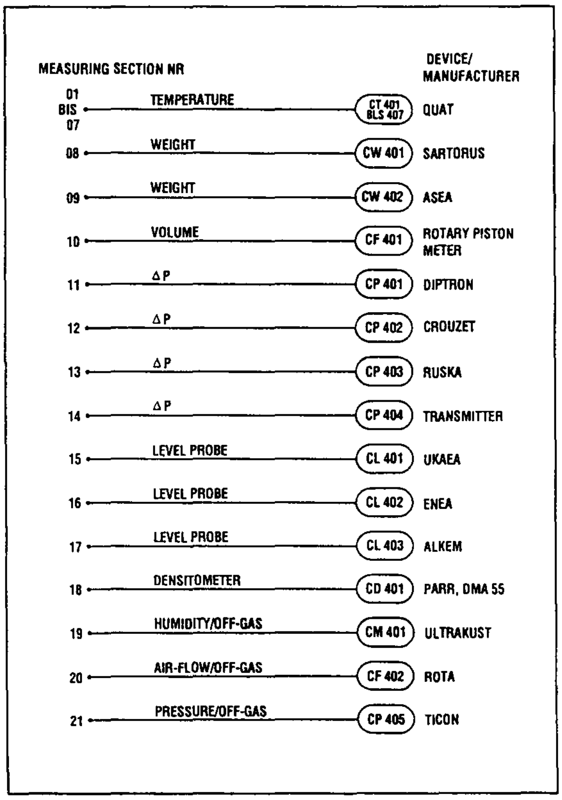
3. TEST CONDITIONS

The test solution was 0.5 molar nitric acid with a density of 1.017 g/cm^3 at 20°C. The off-gas air-flow rate was 9 standard liters/hour. The solution was preheated internally by means of steam injection through the sparge ring to about 50°C the day before the test. Additional heating on the morning of the test raised the temperature to 60°C. The annular tank took about one hour to thermally equilibrate, and it took another hour for the sparge ring and pulsator pipes to fill before thermal effects could be followed. The temperature at the end of the test, after the tank and solution had cooled undisturbed for 47.5 hours was 29.9°C.

4. INSTRUMENTATION

In the organization of the CALDEX project, DWK made a deliberate effort to assure that all types of instruments currently in use to determine tank volumes were part of the test apparatus. A list of the CALDEX tank instrumentation is contained in Figure 2. To the extent possible, instrument control and data acquisition were automated by means of an industrial PC computer-controller, including the reading and setting of the mass gas-flow meters. The types of measurements made in this test were:

i) actual height of liquid in tank as viewed through the upper port window and recorded on a strip of graph paper
ii) pneumatic pressures (height) for three different bubbler-probe lengths
iii) liquid level based on acoustic probe data
iv) liquid level based on capacitance probe data
v) liquid weight based on load cell data
vi) liquid density based on bubbler-probe data
vii) temperature measurements at four locations
viii) tank dimension changes measured at four locations
ix) off-gas flow rate and absolute humidity



MEASURING SECTION NR			DEVICE/ MANUFACTURER
01 BIS 07	TEMPERATURE	CT 401 BLS 407	QUAT
08	WEIGHT	CW 401	SARTORUS
09	WEIGHT	CW 402	ASEA
10	VOLUME	CF 401	ROTARY PISTON METER
11	ΔP	CP 401	DIPTRON
12	ΔP	CP 402	CROUZET
13	ΔP	CP 403	RUSKA
14	ΔP	CP 404	TRANSMITTER
15	LEVEL PROBE	CL 401	UKAEA
16	LEVEL PROBE	CL 402	ENEA
17	LEVEL PROBE	CL 403	ALKEM
18	DENSITOMETER	CD 401	PARR, DMA 55
19	HUMIDITY/OFF-GAS	CM 401	ULTRAKUST
20	AIR-FLOW/OFF-GAS	CF 402	ROTA
21	PRESSURE/OFF-GAS	CP 405	TICON

Figure 2. CALDEX Measuring Instruments

Figure 3 depicts the location of liquid-level measurement systems. Of particular importance for the thermal expansion test was the upper viewing window through which the height of the liquid could be observed directly.

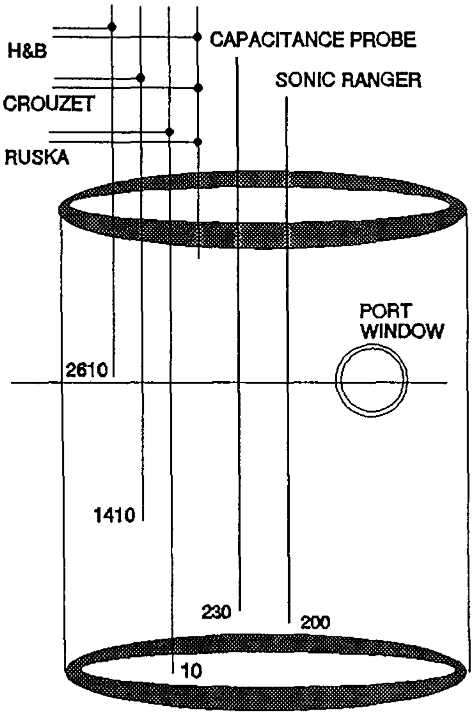


FIGURE 3. LIQUID HEIGHT MEASUREMENTS

The drawing in Figure 4 shows the position of feeler gages used to make the dimensional measurements. Not shown are the locations of the temperature gages which were located near the bottom, at 1500 mm, at 3000 mm (above the level of the test solution), and in the off-gas piping.

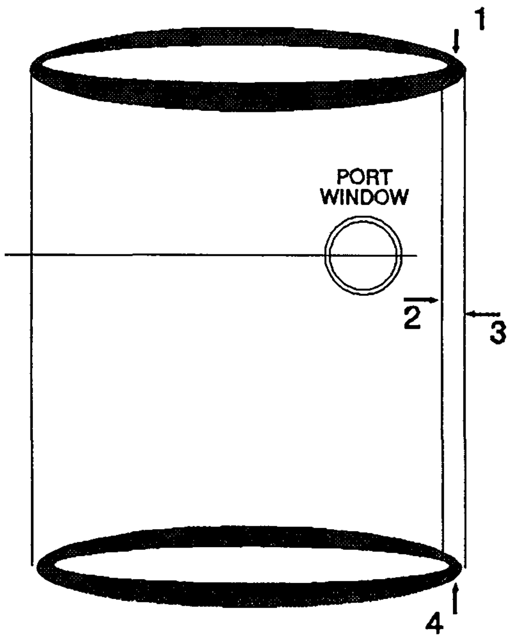


FIGURE 4. DIMENSIONAL MEASUREMENTS

5. TANK TEMPERATURE EFFECTS

Thirteen instrument readings were obtained during each measurement cycle of roughly 70 seconds for a total of 2810 readings per instrument. The large quantity of data obtained during the 47.5 hours of tank monitoring created some data handling problems. In addition to the automated data acquisition, dimensional and sight window data were recorded manually.

The thermal expansion data are summarized in Table 1. The readings have been corrected to account for the calculated loss of liquid due to evaporation. Thermal expansion effects for the CALDEX annular tank were similar to that reported for cylindrical tanks. Each probe has its own thermal expansion coefficient that depends on the properties of the probe and the characteristics of the measurement system.

TABLE 1. THERMAL EXPANSION EFFECTS

Instrument (units)	READINGS at 57.9°C (start)	READINGS at 29.9°C (end)	Difference in Readings	After Correction* for Evaporation
Sight Port (mm)	0	-27	-27	-24
Ruska (mm)	2606	2610	4	7
Crouzet (mm)	2633	2621	-12	-9
H&B (mm)	2616	2610	-6	-
Sonic Ranger (mm)	2698	2632	-66	-63
Capacitance probe (mm)	2601	2627	26	26
Diptron (g/cm^3)	1.01	1.02	0.01	-

* The 3 mm estimate of change in liquid level due to evaporation losses is based on absolute humidity, temperature, and air flow-rate off-gas readings.

The Ruska bubbler probe and the capacitance probe showed an increase in liquid level readings as the solution cooled. The sonic ranger showed the largest change in liquid level readings.

Sight Window The liquid level, observed through the sight window and recorded on a strip of graph paper, decreased by 27 mm during the 47.5 hour cooling time.

Ruska The Ruska data showed a very small increase in pressure as the tank cooled. The Ruska probe was located at 10 mm above the bottom of the CALDEX tank.

Crouzet The Crouzet data showed a 12 mm decrease in pressure level as the tank cooled. The Crouzet probe was located 1410 mm above the tank bottom.

H & B The Hartmann and Braun probe, at 2610 mm above the bottom of the tank, was covered only during the first few hours of the test, and, therefore could not be used to estimate thermal effect.

Sonic Ranger At 63 mm, the decrease in the sonic ranger readings was the largest observed change in liquid level. The sonic probe was located 2400 mm below the ceiling of the tank.

Capacitance Probe The 33 mm increase in the capacitance probe readings was unexpected. The capacitance probe was located 2370 mm below the ceiling of the tank.

Diptron A Diptron pressure gage was assigned to measure changes in the liquid density directly as function of the distances between two probes (probe separation of 200 mm). The 200 mm distance corresponded to a pressure of about 21 mbar (less than 0.3 psi), and was below the certified range of the pressure gage.

6. ESTIMATES OF LIQUID EVAPORATION

Data on the amount of liquid lost via the off-gas system during the cooling period were estimated on the basis changes in tank weight and off-gas flow measurements (Table 2).

Off-Gas Data The absolute humidity, gas-flow rate, and temperature in the off-gas system were measured and used to establish the rate of evaporation. The calculated loss of test solution based during the cooling period on off-gas data was 9 liters which corresponded to a change of 3 mm in liquid height.

TABLE 2. ESTIMATES OF LIQUID EVAPORATION

Instrument (units)	READINGS at 57.9°C (start)	READINGS at 29.9°C (end)	Difference in Readings
ASEA Load Cell (kg)	8107	7895	109 kg 107 liters
Off-gas Flow Rate and Temperature	Reference	9 kg	9 liters

Load Cell Data The loss of test solution due to evaporation based on ASEA load cell system weight data was 107.5 liters. This corresponded to a change of 38 mm in liquid height.

7. CHANGE IS TANK DIMENSIONS

As shown in Figure 3, feeler gages were positioned at four points to monitor the dimensional changes in the tank.

Tank Capacity The increase, shown in Table 3, in the capacity of the tank calculated on the basis of the coefficient of observed height expansion was 11 liters.

Solution Volume The change in the volume of test solution, which occupied 60% of the tank's capacity, was 7 liters.

TABLE 3. CHANGES IN TANK DIMENSIONS

Gage Location (units)	READINGS at 57.9°C (start)	READINGS at 29.9°C (end)	Change in Dimension
Top (mm)	Reference	0.781	1.211
Bottom (mm)	Reference	0.430	
Inner (mm)	Reference	0.400	0.035 mm
Outer (mm)	Reference	0.365	
Calculated change in annulus	Based on coefficient of height expansion		0.115 mm

8. FUNDAMENTAL PRINCIPALS

The volume of liquid in the tank is a function of the probe heel and the change in cross-sectional area.

The volume equation for a linear region of a regular tank is

$$V_i = a + b*h_i \qquad (1)$$

where V = volume; h = height; and a and b are coefficients determined by least squares analysis.

8.1 EXPANSION OF THE LIQUID

Fundamental relationships between the mass (M), volume (V), density (ρ), pressure (P), and height (h) are

$$M_i = \rho_i * V_i , \qquad (2)$$

$$P_i = \rho_i * g * h_i . \qquad (3)$$

Substituting values of **V** and **h** in equation (1)

$$\frac{M_i}{\rho_i} = a + b*\left[\frac{P_i}{\rho_i\, g}\right] . \qquad (4)$$

Multiplying equation (4) by the density.

$$M_i = (a*\rho_i) + \left[\frac{b}{g}\right]*P_i . \qquad (5)$$

Equation (5) states that a change in the solution density, which changes with temperature, only effects the intercept (location) term of the equation. In practice, the value of the intercept is related to the distance of the pressure measuring probe above the bottom of the tank.

8.2 THERMAL EXPANSION OF THE TANK

The thermal expansion of the vessel is independent of the solution it contains. The terms in the volume equation (except for the intercept term) express the change in cross-sectional area as a function of height. For a linear region in a regular tank, the change in volume (V) can be expressed in terms of the changes in the vessel's height (h), circumference (C), and cross-sectional area (A) as

$$\partial V = h\partial A + A\partial h \qquad (6)$$

where $\partial A = 2\pi r\partial r$, *and* $\partial r = \partial C/2\pi$.

8.3 FUNDAMENTAL RELATIONSHIPS FOR THERMAL EXPANSION

If L_0 is the length at 0°C and α is the coefficient of linear expansion, the length at t°C is,

$$L_t = L_o (1 + \alpha \Delta t) \qquad (7)$$

where Δt *is the change in temperature.*

Equation (7) is applicable to changes in the length of the probes and the height of the tank.

If A_0 is the area at 0°C and α is the coefficient of linear expansion, the area at t°C is,

$$A_t = A_o (1 + 2\alpha \Delta t) \qquad (8)$$

where Δt *is the change in temperature.*

Equation (8) is applied to the changes in the volume of the tank.

8.4 Determination of Temperature Corrections for Volume

Temperature correction factors must be determined individually for each tank, type of probe, and measurement system. The change in tank capacity due to thermal expansion is a function of the thermal properties of the material used in its fabrication. The coefficient of thermal expansion for a common type of stainless steel is 0.00001872, which is about one percent that of water at 40°C.

9. DATA REDUCTION AND ANALYSIS

Although the results of the CALDEX thermal expansion test are in general agreement with previous experiments, spurious data, and small inconsistencies or unexpected results were observed. Examples include the increase in capacitance probe liquid height and the difference between the off-gas estimate of evaporation losses and that measured as change in weight by the load cells.

The Ruska data displayed the characteristic increase in pressure for a probe whose tip is located near the bottom of the tank. The Crouzet probe showed thermal expansion effects of both the tank and the test solution. In practice, a correction should also be made for the change (which was not made) in the length of this probe relative to the tank.

For bubble probes fabricated from the same material as the tank and terminating near the bottom of the tank, the change due to the vertical expansion is negligible. Since the pressure gages sense the weight of the column of liquid which does not change with temperature, bottom probe readings are effected only by the expansion of the tank. For bubble probes not near the bottom of the tank, the expansion of the liquid must be taken into account as shown in equation (5).

Temperature correction algorithms for the sonic ranger and the capacitance probe, which measure distance from the top of the tank to the liquid level, are more complicated since the temperature and conditions in the vaporhead above the liquid level must also be taken into account. The sonic probe level reading is influenced by the expansion of the tank, the expansion of the liquid and the temperature effects on the air in the vaporhead of the tank. The capacitance probe temperature effect is larger than that which can be attributed to the tank and solution combined.

Additional testing and data analysis will be needed to formalize the sonic ranger and capacitance probe temperature correction equations.

The Diptron pressure gage measured the differential pressure over a distance of 200 mm, as opposed to measuring both the probe pressures relative to the vapor head. Although in theory very accurate determinations of the solution density should be possible using the Ruska and Crouzet probe readings as a back-up estimate, the fast bubble method used in collecting these data rendered them unusable for this purpose.

Estimates of the loss of liquid due to evaporation were made on the basis of the ASEA load cell weight data and absolute humidity, temperature and the off-gas flow rate calculations. The estimate based on the weight data was much larger than the off-gas calculations.

On the surface, the load cell data appeared to be relatively error free. However, analysis of other CALDEX test data showed that true vertical alignment of the load on the load cells was not being maintained. This was due to 1) the four load cell suspension arrangement which was not self correcting, and 2) the fact that the tank had a sloped bottom so that the tank center of gravity shifted as it was filled creating a shear force that effected the readings. Therefore, the evaporation loss calculated on the basis of the off-gas data was considered to provide a better estimate. For future reference, a three point load cell suspension, which is intrinsically self balancing, is recommended.

The measured change in tank height was 1.211 mm. Two methods were used to calculate changes in the radii: 1) inner and outer wall measurements, and 2) the coefficient of expansion based on the observed height increase. Because of the inconsistency of the inner and outer feeler gage data with other measurements, the increase in the capacity of the tank was calculated on the basis of the coefficient of observed height increase. The calculated value was 11 liters. Determination of the calculated capacity and volume are given in Appendix A.

ACKNOWLEDGMENTS

We thank H. Dratschmidt, GNS (now with The EURATOM Safeguards Directorate, Luxembourg), for getting us started in the analysis, and B. Keisch, BNL, for helpful comments and suggestions that improved the paper. Research by the first author was supported by the U.S.DOE under contract DOE-AC02-76CH00016.

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APPENDIX A: CHANGES IN TANK DIMENSIONS

The formula for the volume of an annular tank with height h and inner and outer radius r. and r.., respectively, is pi * (r.^{2} - r..^{2}) * h.

	Tank capacity at reference conditions	
Reference height	h. = 4200 mm	
Annulus = 400 mm		
inner radius:	r. = 1040	
outer radius:	r.. = 1440 mm	
pi * h.	= 13,194.7	Published tank capacity:
(r..^{2} - r.^{2})	= 992,000	= 12,500 liters.
mm^3 / 10^6	= 13,089	
	Tank capacity at 55.9°C	
Change in height = 1.211 mm		
New height	h, = 4201.211	
Measured change in annulus	= 0.035 mm	Calculated change is based on
Calculated change in annulus	= 0.115 mm	coefficient of observed height
New calculated radii	r, = 1040.415	expansion:
	r,, = 1440.300	(1.211/4200 = 0.000288)
pi * h,	= 13,198.5	
(r,,^{2} - r,^{2})	= 992,571	
mm^3 / 10^6	= 13,100	
	Change from reference	
Reference capacity	= 13,089 liters	
Tank heated capacity	= 13,100 liters	
Capacity	= 11 liter	Test solution volume was 60% of
Solution	= 7 liter	capacity
	Comparison with Ruska data	
Level	= 3 mm	
Volume	= 8 liters	

INTEGRATED MATERIAL ACCOUNTANCY SYSTEM

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Abstract

In this paper we present the system that we are actually using for Nuclear Material Accounting and Manufacturing Management in our UO_2 Fuel Fabrication Plant located at Juzbado, Salamanca, Spain.

The system is based mainly on a real time data base which gather data for all the operations performed in our factory from UO_2 powder reception to fuel assemblies shipment to the customers.

The accountancy is just an important part of the whole integrated system covering all the aspects related to manufacturing: planning, traceability, Q. C. analysis, production control and accounting data.

1. Introduction

Since 1987 we have been developping at Juzbado a computer based system with the target of performing the automatization of the capture, treatment and final presentation of all information related to the different operations in a Fuel Fabrication Plant.

In the fabrication of the UO_2 fuel assemblies, every year it is necessary to handle hundred of thousands of minor components which are subjected to different processes in order to get a final fuel assembly. The generated data, are processed and suffer mathematical and logical calculations to obtain the traceability and necessary accounting data required for both international authorities and customers. This tasks makes necesssary the use of a complete and complex computer system to avoid the thousands of man-hours used during the reloads before 1987.

This computer system has been called "PATMAN":

PROYECTO DE
AUTOMATIZACION DE L
TRAZABILIDAD Y CONTROL DEL
MATERIAL
NUCLEAR

At the beginning the system was conceived from the difficulties that appeared on years 1985 and 1986 in making the annual nuclear material inventory, but after that it merged as a "CAM"(Computer Aided Manufacturing) System which integrates all management aspect of the fuel production. Today the system is being improved and developed to incorporate the manufacturing process computer automation, and so, become a "CIM" (Computer Integrated Manufacturing) System in the next future.

It is interesting to say that the sotware work has been completely developed by our own computerization personnel who practically have been only dedicated to this job from year 1985.

2- Brief Description of the Areas Included in the Project

Due to the complexity of the project it has been divided into modules (or areas) which have been chosen the most possible isolated among them and, at the same time, in such a way that the relations among the areas are simple.

With the above considerations the following areas were defined:

- Project Planning and Programming
- Non Nuclear Component Control
- Powder Storage Control
- Chemical Laboratory Control
- Ceramic Process Control
- Mechanical Process Control
- Residues Control
- Traceability and Shipping Documentation
- Accountancy Inventory and Official Reports.

These areas (or modules) are themselves divided into operations in order to develop the system.

3. Nuclear Material Control

Characteristic Description

Under the point of view of Internal Nuclear Material Control our instalation is divided into seven accounting areas.

This division is due, among other reasons, to the different types of products we handle. The areas are:

1.- Storage of clean UO_2 and U_3O_8
2.- Storage of other N. Materials
3.- Materials in Ceramic Fabrication Process
4.- Other Materials in Ceramic Area
5.- Materials in Mechanical Fabrication Process
6.- Materials in Chemical Lab.
7.- Storage of Non-Recoverable Residues.

The identification of all items handled in the shop (except for area # 5) has a codified number as explained hear on:

a			b
999	XX	XX	999
1ª	2ª	3ª	4ª

a) Fabrication Lot Identification

1ª. These three first digits show the nominal enrichment in the U-235 isotope.

2ª. These two digits identify the lots in alphabetical order to avoid repetitions.

3ª. These two digits identify the product of the material lot in such a way that the first one indicates the type of product and the second one the phase of the process where it is located.

b) Item identification

4ª. These are three sequential numbers to indicate the different items belonging to the same lot.

There are material transfers among different areas, which generate an internal accounting of the nuclear material managed by the system.

It also manages the division of an item to generate two or more items, or the blending of several items to generate just one.

In the accounting of the Nuclear Material there are two main characteristic: the chemical analysis (to determine U and U-235) and the weighing of the material. Based on that, there are three modules to calculate the accountancy:

- Chemical Laboratory Management
- Nuclear Material Handling
- Not Measured Material Evaluation

Chemical Laboratory Management. The analysis of representative samples of the lots used in the process are performed in the Chem. Lab.

The system controls not only the samples and essays but also the "standards" used to calibrate each laboratory equipment and the status of equipments regarding recalibration. The results of the essays are also storaged in the system associated with the samples they cover.

The system also takes into account the essays still not made belonging to a determined lot. When all analysis have been done, the system makes the calculation of the porcentage of U and U-235 as well as other requeriments, checking that all of them meet the product specification.

Nuclear Material Handling. In all the operations associated with a physical process in which Nuclear Material is handled, the system obtains the Uranium weight either by direct weighing (difference) or by substraction of tare and gross weight.

The percentages of element and isotope are transferred or recalcu-lated when blending of items is performed.

The weight data is trasferred automatically from the measuring instrument (scale, etc.) to the system which evaluates if it is inside a determined range. If several items are placed together or are assembled the system calculates the weight of the final component or resulting assembly and also gives the percen-tage of U and U-235.

Not Measured Material Evaluation. In the Ceramic Process we handle material that it is not "sealed" (handled under containment), and is submitted to changes in his physical or chemical properties. (In these changes weight variations are produced because of either chemical or physical changes as for instance humidity loss). Also there is some amount of material that remains inside the equipment. In these cases the system performs a "Material Difference" (MD) per each lot of material managed in the area.

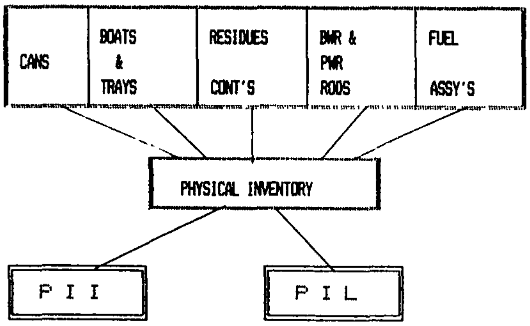
Lot by lot, along the time, we have several "M.D." that can be "counteracted" (positives and negatives) because of the remaining material that can be recovered during the equipment cleaning operations.

These accumulations of M.D.'s would give us the "MUF" of the area for a pre-determined period.

Inventory Process

The way of preparing the inventory is by getting a report of items or groups of items in the whole factory taken from the physical situation of the items of each system module /1/.

So, we may have the following situations:



In each report there is an item identification with more than 18 atributes related to characteristics about location as well as physical, chemical properties, type of product and even an error code associated to each. From this report we can obtain (as wished), the following two other reports:

PIL.-Physical Inventory Listing
PII.-Physical Inventory Inspection Program

The information given by "PIL" is codified and sent to EURATOM through the IBERPAC line.

The information given by "PII" may also be transmited in Juzbado to the personal computer brought at the time of the inventory by Euratom inspectors and constitutes the starting information for the "JUZBADO INVENTORY INSPECTION PROGRAM", which is a computer program developped by EURATOM that allows their inspectors to perform the Physical Inventory in our Instalation.

The system allows us to use at any time all data for "Internal Material Accountancy", as well as to perform an Accounting Inventory of about 40000 items in about 20 minutes.

One of the improvements now being developed in the system is an ICR (Inventory Change Report) that allows us, at any time, to know the Inventory changes happenned in a pre-determined period of time. Once the Report is obtained and CODIFIED, it can be sent to EURATOM by means of the Comunication System already explained.

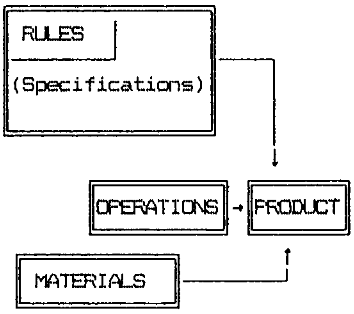
4.Project Development

Methodology.

The Project started with the basic idea of integrating all the informations taken from the nuclear material and from the process they have been submitted to. For this reason we have used, in the Project conception, a Methodology of the type Subjet Orientated (MERISE)/2/,/3/,/4/.

System operation.

The following graphic gives an idea of how the system operates:



Materials are physical items that are described by logical items, both with the same identification.

Each logic item is associated to the identification of those atributes corresponding to the characteristic of the physical item.

The identification of the items depends on the fabrication phase and the type of material as it was described before.

At the phases where the characte-ristics of the product (both Physical and Chemical) are evaluated, the system selects the specifications to be used by just checking the identi-fication of the evaluated items and the type of Product to be manufac-tured.

At the phases where the physical state of the product changes, the new product and consequently its new atributes are also calculated by the system.

Operators (users or shop workers).

Many operations can be performed with the system and the operators (users) are also a great number. We have designed a relational system so that the operations and inspections can only be made by authorized people.

OP_1 —— ID_A OP_2 —— ID_B

OP_3 —— ID_A OP_4 —— ID_B

OP_5 —— ID_A OP_6 —— ID_B

....

OP_{n+1} —— ID_C

OP_i means operators (users)
ID_i means operator identification

The operators (users) are identified by their user-name and each one accedes to the system by their secret word (password); each one has an "identification" that allows him to perform only the authorized operations in the system. Several operators may have the same identification if they perform the same activity in the shop. In this way we can guarantee the use of the appropriate operations by only the authorized people.

5.SYSTEM SOFTWARE SUPPORT.

The basis of the project is a group of inter-related data bases that containt the manufacturing process information referred above. There are means to guarantee the information integrity and withdrawal in case any hardware trouble can happen.

There is also implemented in the system a QUERY language (considered a fourth generation language), develop in Spanish, to which has been added several statistical functions, in this way the system can be managed by the Plant operators who are not familiarized with computers. The indicated language, called DTRSP, has been developped from the "datatrieve" (DTR) commercial package.

The commercial software packages used to develop the system are:

VMS	Operating System
LAVC	Local Area Vax Cluster
CDD	Command Data Dictionary
RDB	Relational Data Base
TDMS	Terminal Data Management System
COBOL	Programming Language
FORTRAN	Programming Language
DECNET	Decnet Network Management
FCSA	Personnel Computing System Architecture
POLYCOM-240	VT 240 Terminal Emulator
SMP	Symbol Manipulation Program
DTR	Data Terminal Report

6.PROJECT HARDWARE SUPPORT

The Hardware means which support the system are:

a.- Computers
    MICROVAX 3500
      (CPU 2,7 MIPS, I/O 3,3 MB/S)
    VAX 8250
      (CPU 1,2 MIPS, I/O 13,3 MB/S)
b.- Mass Storage Disks
    2 RA-82
      (622 MB , 32,3 ms access time)
    2 RD-54
      (159 MB, 38,3 ms access time)
c.- Magnetic Tape Unit
    TSV05
      (1600 BPI, 45 MB)
d.-Computer Terminals
      40 units
      VT-220,VT-320,VT-340,
      VT-330 and VT-420
e.-Personal Computers
      35 units
      IBM,EPSON,etc.
f.-Local Area Network(LAN)
    ETHERNET
      1000m, 3 sectors, 20 nodes
      with 3 network branches PCSA.
g.-Private communication network
    from Salamanca to Madrid
h.-Public connection
    with IBERPAC national network
i.-Process Computers
    Digital PDP-1173
    Digital Vax Station 3100
j.-Computer System Configuration
    At the moment Local Area Vax
    Cluster, eventually Dual Host

7.Conclusions

With this system we have accomplished the main aim planned which consisted in implementing an integrated system able to cover all different purposes in our Manufacturing Plant which signifies an increase in QUALITY in the fuel manufactured by ENUSA.

Our objective was also to provide our customers, and the national and international safeguards authorities,with the necessary information required and in a near real time mode, allowing a way of nuclear material control according to the most modern systems used nowadays in the world.

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ORGANISATION ET DEVELOPPEMENT DU SYSTEME FRANCAIS DE CENTRALISATION DE LA COMPTABILITE DES MATIERES NUCLEAIRES

B. DUFER CEA/IPSN
F. LECOMTE CEA/IPSN

Introduction

Le CEA a mis en place au début de 1965 une centralisation de la comptabilité des matières nucléaires détenues dans les installations nucléaires françaises dont il était propriétaire. Ce système a été utilisé également pour adresser à Euratom les rapports comptables périodiques.

Au plan national, la loi du 25 juillet 1980 sur la protection et le contrôle des matières nucléaires a donné lieu à la parution de l'arrêté du 24 juin 1982, confiant au CEA (Institut de Protection et de Sûreté Nucléaire - Département de Sécurité des Matières Radioactives) la charge d'effectuer la centralisation de la comptabilité des matières nucléaires pour la totalité des installations nucléaires françaises.

Si, dans une première étape cette centralisation ne fut qu'une quasi généralisation au plan national du système utilisé déjà depuis de nombreuses années au CEA, elle a, depuis lors, fortement évolué afin de servir au mieux la sécurité des matières nucléaires tant au plan national qu'international.

Ainsi aujourd'hui le système français de centralisation de la comptabilité des matières nucléaires a plusieurs objectifs :

- rassembler toutes les données permettant à la France de remplir ses obligations vis-à-vis des contrôles internationaux et principalement fournir à l'AIEA et à Euratom des rapports périodiques,
- dans le cadre du contrôle national :
  - . connaître en permanence les stocks des matières nucléaires détenues dans chaque installation, y compris dans celles intéressant la Défense,
  - . conserver l'historique de ces stocks et des flux de matières nucléaires ayant transité dans ces installations.

Compte tenu de cet historique, une comparaison du système national français avec les dispositions du Règlement Euratom et les Arrangements subsidiaires de l'Accord conclu entre la France, Euratom et l'AIEA fait apparaître certaines différences de conception. Elles portent notamment sur le découpage des installations en zones de bilan matières ou ensembles techniques et sur le regroupement des données élémentaires à partir desquelles les déclarations sont effectuées et les stocks sont calculés. Certaines opérations techniques, telles que les mélanges, ou administratives, telles que les transferts de propriété, donnent lieu à des enregistrements et à des traitements différents selon qu'il s'agit du système national ou des systèmes internationaux. Ces disparités ne s'opposent pas cependant à une parfaite cohérence des résultats comptables et ne créent aucune divergence, comme cela va être montré dans ce qui suit.

1. Origine, type de fréquence de transmission des informations centralisées par l'IPSN

1.1. Entités à l'origine des déclarations

La réglementation nationale française prévoit que les établissements ou installations nucléaires peuvent être divisés en ensembles techniques et sous-ensembles techniques. Si cette notion est voisine de celle de zone de bilan matières utilisée pour les contrôles internationaux, il convient néanmoins de percevoir les relations existant entre ces deux notions.

Aujourd'hui, l'ensemble des détenteurs français de matières nucléaires soumis au régime d'autorisation représente 30 sociétés exerçant leurs activités dans 63 établissements, au sein desquels sont distingués 284 ensembles techniques représentant 371 sous-ensembles techniques.

Le nombre de détenteurs soumis au régime de la déclaration annuelle (stocks inférieurs à 500 Kg d'uranium naturel ou appauvri ou de thorium ne nécessitant pas d'autorisation) est supérieur à 600.

C'est au niveau de ces 371 sous-ensembles techniques que sont rédigées les déclarations comptables élémentaires transmises, par les exploitants au DSMR, au titre du contrôle national.

Ces ensembles techniques étant de taille et d'importance variables, il en résulte une certaine hétérogénéité dans la relation entre sous-ensemble technique et zone de bilan matières distinguée pour les besoins des contrôles internationaux. Ces zones de bilan matières peuvent donc selon les cas représenter au sein d'un même établissement, une ou plusieurs installations, ou un ou plusieurs ensembles techniques ou sous-ensembles techniques, gérés distinctement dans le cadre du contrôle national.

Cependant hétérogénéité ne signifie pas incompatibilité et les découpages entre sous-ensembles techniques restent cohérents avec ceux constituant les zones de bilan matières et ainsi permet d'éviter tout chevauchement.

1.2. Type de déclarations effectuées au titre du contrôle national

Outre la différence décrite précédemment, et pour ne se borner qu'au seul aspect des rapports comptables, le système de comptabilité national prévoit que seules les variations de stocks doivent faire l'objet de déclarations, alors que le système international impose d'effectuer des déclarations de stock en fin de mois, même s'il n'y a eu aucune variation. Si cette différence ne paraît pas fondamentale, dans la mesure où le système informatique national détermine en permanence la valeur du nouveau stock à partir de chaque variation enregistrée, on verra dans le paragraphe relatif aux procédures de codification que les systèmes national et international sont malgré tout très différents pour ce qui concerne les stocks élémentaires gérés distinctement par chacun d'eux.

1.3. Fréquence de transmission des déclarations

Une des autres particularités du système national de centralisation de la comptabilité des matières nucléaires réside dans la fréquence de transmission des déclarations comptables par rapport à la périodicité mensuelle demandée au titre des contrôles internationaux.

En effet, les opérateurs sont tenus de transmettre au DSMR toute déclaration de variation de stock intervenue au sein d'un ensemble technique, le jour même où cette variation de stock a eu lieu ou a été déterminée.

La réglementation nationale prévoit par ailleurs que les mouvements de matières nucléaires entre deux sous-ensembles techniques appartenant au même ensemble technique peuvent être regroupés pour être déclarés à la fin du mois au cours duquel ils ont eu lieu.

En résumé le système national de centralisation de la comptabilité des matières nucléaires, bien qu'il soit tout à fait compatible et cohérent avec les contrôles internationaux, diffère donc de celui mis en oeuvre pour les besoins de ces contrôles dans ses grands principes de base et notamment au niveau :

- des entités à l'origine des déclarations,
- du type de déclarations transmises,
- de la fréquence de transmission de ces déclarations.

De telles différences pourraient être considérées comme mineures, si par ailleurs les systèmes national et international utilisaient les mêmes codifications ou des codifications distinctes entre lesquelles pourraient être établies de véritables relations bi-univoques ; mais tel n'est pas le cas.

2. Principes de codification

Pour ne citer que les principales et sans prétendre les examiner en détail, ces codifications différentes portent essentiellement sur les catégories de matières distinguées et sur les différentes variations de stock suceptibles d'être déclarées.

2.1. Codification des catégories de matières

Sous le terme catégorie de matières le système national regroupe diverses informations élémentaires relatives à l'entité à comptabiliser, ce sont : l'élément, sa teneur, son code d'engagement de contrôle, son irradiation (ou absence d'irradiation) et le produit (forme physicochimique) qui le contient.

2.2. Codification de l'élément

Outre le fait que le système national prévoit la comptabilisation de certains éléments non suivis au titre des contrôles internationaux (tritium, lithium 6 ...), on notera d'une part que l'uranium enrichi, quelle que soit sa teneur en uranium 235, est caractérisé par un code élément unique, là où le règlement d'Euratom en prévoit deux (légèrement enrichi, hautement enrichi) et d'autre part que l'uranium 233 fait l'objet d'un code élément spécifique, alors que le système international le distingue par le biais du code isotope.

2.1.2. Codification de la teneur

Si le système international prévoit pour l'uranium enrichi deux grandes classes de teneur : uranium faiblement enrichi et uranium hautement enrichi, le système national gère de façon spécifique chaque teneur isotopique distincte. Ainsi sont comptabilisées, principalement pour les besoins de la gestion, environ 400 teneurs isotopiques distinctes pour l'uranium enrichi.

2.1.3. Codification de l'engagement de contrôle

Bien que la notion d'engagement de contrôle ne soit pas une notion prévue explicitement par la réglementation nationale, le système national de comptabilité des matières nucléaires a dû en tenir compte au niveau de son organisation pour répondre aux exigences des contrôles d'Euratom et aux responsabilités de l'Etat français dans ce domaine. Cette organisation permet également :

- d'une part à la France de suivre distinctement des matières entrant dans le cadre d'engagements bilatéraux souscrits auprès de pays tiers,
- d'autre part, et historiquement, de distinguer les matières dont le groupe CEA est propriétaire.

Ce sont ces divers paramètres qui conduisent aujourd'hui le système national de centralisation à gérer distinctement 59 "engagements d'utilisation ou de codes pool" différents, là où Euratom ne distingue que 14 engagements particuliers de contrôle ou codes pool.

2.1.4. Codification de l'irradiation

Outre les matières fraîches et les matières irradiées, le système national distingue l'uranium issu du retraitement auquel a été affecté un code d'identification spécifique pour des motifs de gestion.

2.1.5. Codification du produit

En dernier lieu et comme cela a été indiqué en première partie, le système national comptabilise par ailleurs 551 formes physicochimiques différentes appelées "code produit", là où la réglementation internationale n'en gère qu'une trentaine.

2.1.6. Codification des stocks élémentaires

Ces principales disparités entre le système national et international de centralisation de la comptabilité des matières nucléaires conduisent donc au suivi de stock élémentaires fondamentalement différents dans les deux systèmes.

Ainsi, si dans le système de contrôle international les stocks sont caractérisés par la combinaison des paramètres ci-après :

- zone de bilan matière,
- élément,
- engagement de contrôle,

dans le système national un stock élémentaire résulte de la corrélation de chacun des paramètres suivants :

- sous-ensemble technique,
- élément,
- teneur,
- code d'engagement de contrôle,
- irradiation,
- produit.

A titre indicatif, étaient comptabilisés au plan national, au 1^{er} mars 1991, 7147 stocks élémentaires soumis au contrôle d'Euratom, à comparer aux 614 que le système international conduisait à distinguer.

Encore une fois, il convient de souligner que ces disparités de traitement des données ne font nullement obstacle à une parfaite cohérence des résultats comptables et ne créent aucune divergence entre les systèmes.

2.2. Codification des opérations

Plusieurs grandes catégories d'opération donnant lieu à variation de stock et à déclaration de cette variation sont différenciées au titre du contrôle national. Elles regroupent notamment :

- des transferts externes de matières nucléaires de ou vers un ensemble technique ou une zone de bilan matière,
- des transferts internes de matières nucléaires au sein d'un même ensemble technique - échange entre deux sous-ensembles,
- des modifications des caractéristiques techniques de la matière,
- des entrées ou sorties de matières nucléaires des comptabilités nationale où internationale,
- des écarts constatés entre des quantités de matières nucléaires attendues ou provisoires et des quantités reconnues,
- des modifications des conditions d'utilisation ou de comptabilisation des matières,
- des modifications des données patrimoniales, douanières ou commerciales,
- des déclarations purement comptables.

Compte tenu de la différence notable entre les stocks élémentaires gérés par chacun des systèmes, on imagine aisément que certaines variations de stocks se rapportant à l'une quelconque de ces catégories puissent faire l'objet de déclarations au titre du contrôle national et n'avoir aucune incidence au niveau de l'évolution des stocks différenciés pour les besoins des contrôles internationaux, c'est le cas notamment :

- des mouvements de matière entre sous-ensembles techniques relevant de la même zone de bilan matière,
- des changements de teneurs ne conduisant pas à un changement de catégorie de matière pour Euratom.

A l'inverse certaines variations de stock ne faisant l'objet que d'une déclaration unique au plan du contrôle national peuvent dans certains cas conduire à plusieurs déclarations au titre du contrôle international. Nous ne citerons à titre d'exemple pour illustrer ces propos que :

- les mélanges de matières nucléaires.

A ces équivalences partielles viennent par ailleurs s'ajouter des variations de stocks qui ne sont générées que pour l'un ou l'autre des deux systèmes, compte tenu de leur organisation et de leur mode de fonctionnement propre ; c'est le cas par exemple :

- du changement d'utilisation ou de propriétaire sur le plan national, sans changement de l'engagement de contrôle international,
- des mouvements internes à un même ensemble technique.

Cet examen succinct a permis de mettre en évidence les réelles singularités de chacun des systèmes. Comme on le constatera ensuite, l'importance et le nombre de ces disparités ne pouvaient conduire à envisager l'utilisation, par les exploitants, des mêmes procédures pour effectuer leurs déclarations de variations de stocks au titre des contrôles national et international.

3. Support permettant d'effectuer les déclarations de variations de stock et procédures de validation de ces déclarations

3.1. Le support

L'exploitant français dispose d'un document unique, le "bordereau de déclaration d'opération sur les matières nucléaires (BDOMN)" pour effectuer ses déclarations de variation de stock.

Outre le fait que ce document permet de déclarer tout type de variation de stock il conduit à :

- une meilleure séparation des informations fournies au titre du contrôle national et des contrôles internationaux, voire de la gestion patrimoniale,
- un chaînage strict des déclarations annulées et remplacées avec les déclarations remplaçantes.

Il autorisera par ailleurs :

- un traitement informatisé des comparaisons entre :
  - . d'une part les déclarations comptables et les autorisations d'exécution de transport et,
  - . d'autre part les déclarations comptables et les notifications effectuées dans le cadre de l'application des INFCIRC 290 et 207 et des articles 24 et 25 du règlement Euratom,
- l'entrée en vigueur d'un système d'accusé de réception traité informatiquement et consistant à corréler les déclarations faites par le destinataire et par l'expéditeur.

Ces deux derniers points seront développés plus loin dans le cadre de l'évolution programmée pour l'année 1991.

3.2. Procédure de validation des déclarations

La France étant un Etat de droit écrit, chaque variation de stock déclarée au moyen du formulaire précédent, doit faire l'objet d'une authentification par l'autorité qui en est l'origine. C'est pour cette raison que chaque BDOMN comporte une zone particulière dans laquelle l'autorité responsable au niveau de l'unité émettrice doit apposer sa signature manuscrite afin de valider les informations qu'il contient. Ceci est à rapprocher du système de signature prévu au bas de chacun des feuillets de rapport de variation de stocks adressé à Euratom pour les besoins des contrôles internationaux.

Environ 3200 déclarations de ce type sont adressées mensuellement au DSMR/BIC (Bureau Informatique et Comptabilité) au moyen de divers modes d'acheminement qui vont être examinés maintenant.

3.3. Les moyens de transmission

Compte tenu de la présence de cette signature induisant un statut de preuve, au sens juridique français, aux documents sur lesquels sont rédigées les déclarations comptables, celles-ci doivent réglementairement être adressées sur un document papier au DSMR.

Toutefois ce mode d'acheminement pouvant conduire à des délais de transmission relativement importants (de l'ordre d'une semaine), il ne permet pas de connaître en temps quasi réel les variations de stock de matières nucléaires détenues dans les différentes installations. La recherche de cet objectif, associé au développement chez l'exploitant de moyens informatiques permettant d'assurer le suivi et la comptabilité des matières nucléaires, ont conduit le DSMR à proposer en parallèle aux unités effectuant le plus grand nombre de déclarations, différents modes de transmission de leur variation de stock par voies télématiques.

3.3.1. Les transmissions de type BSC 2780

Ces transmissions qui utilisent le réseau commuté (réseau téléphonique des P et T) s'établissent par l'intermédiaire d'une procédure de communication développée par IBM, la procédure BSC 2780 (binarysynchroneous communication) qui permet, avec une bonne fiabilité, l'échange de fichiers relativement importants à une vitesse élevée.

Pour des raisons évidentes de sécurité ce mode de transmission demeure cependant réservé aux seules informations non classifiées pour les besoins de la défense.

Aujourd'hui les correspondants les plus importants utilisent ce mode de transmission, il s'agit :

- de COGEMA : par le biais de son organe concentrateur de données localisé à Pierrelatte,
- d'EURODIF,
- de COMURHEX : pour les Etablissements de Malvési et de Pierrelatte.

Près de 75 % des déclarations reçues mensuellement sont transmises de cette façon.

3.3.2. Les transmissions de type "Minitel"

Ce type de liaison s'effectue en mode conversationnel avec l'ordinateur frontal du DSMR et utilise le réseau Transpac. Il est accessible à partir d'un micro-ordinateur, d'un terminal ou d'un minitel. Le minitel est un équipement constitué d'un poste téléphonique couplé à une imprimante et à un écran auquel est associé un clavier similaire à celui d'une machine à écrire.

Ce système est parfaitement adapté à la transmission de faibles volumes d'information et est à ce titre utilisé par les petites installations. Celles-ci effectuent le plus souvent peu de déclarations dans lesquelles, par voie de conséquence, sont constatées régulièrement un nombre d'erreurs élevé.

Ce mode de transmission fait l'objet d'une présentation Poster dans le cadre du présent congrès.

Avant d'examiner les divers types de traitements effectués sur les données reçues, l'architecture du système national de centralisation informatisée de la comptabilité des matières nucléaires est décrite ci-après.

4. L'architecture informatique

La configuration matérielle utilisée dans le cadre de la centralisation des matières nucléaires est articulée autour de 2 mini ordinateurs Intertechnique Multiprocesseurs des Gammes IN 8000 et IN 4000 dont l'un est exclusivement dédié aux transmissions, et dont l'autre contient l'intégralité de la base de données du système.

4.1. L'équipement frontal

Il consiste en un mini ordinateur IN 8700 comprenant :

- une unité de calcul de 3 méga octets,
- une mémoire de masse de 300 méga octets,
- un lecteur de mini bandes,
- 5 terminaux de visualisation,
- une imprimante série 400 CPS,
- une imprimante laser canon pour les applications de bureautique,
- une imprimante AGFA P400 permettant d'assurer l'impression de 24 pages par minute,
- un interface de transmission X25 pour la connexion au réseau transpac et le dialogue avec l'ordinateur de base,
- un interface de transmission BSC pour les liaisons avec les sites distants.

4.2. L'équipement de base

Il consiste en un mini ordinateur IN 4400 comprenant :

- une unité de calcul de 3 méga octets,
- une mémoire de masse de 300 méga octets,
- un lecteur de mini bandes,
- 15 terminaux de visualisation,
- une imprimante série 400 CPS,
- une seconde imprimante rapide laser AGFA P400,
- 1 imprimante laser canon pour les applications de bureautique.

Pour en terminer avec cet aspect de l'architecture informatique, il convient d'indiquer qu'il existe une concentration importante sur le site du Centre d'Etudes de Cadarache, des inspecteurs chargés d'assurer le contrôle national. Le DSMR a donc été conduit à mettre en place une architecture analogue dans ses locaux situés sur ce Centre afin de permettre à ces inspecteurs d'assurer leurs fonctions dans les meilleurs conditions possibles. Cette architecture comporte un ordinateur IN 5054, lui-même alimenté par le système de Fontenay-Aux-Roses à travers un équipement frontal constitué par un micro ordinateur Bull.

Outre les opérations nécessaires à l'acquisition des données c'est sur les équipements de Fontenay-Aux-Roses qu'est effectuée l'intégralité des traitements nécessités tant par le contrôle national que par les contrôles internationaux.

5. Le traitement des données

Dans ce chapitre les traitements des données effectués au titre du seul contrôle national sont distingués de ceux réalisés pour les besoins des contrôles internationaux.

5.1. Traitement des données au titre du contrôle national français

5.1.1. Acquisition des données

Le premier traitement effectué au titre du contrôle national sur les données reçues consiste en leur contrôle, puis en leur intégration dans la base de données. Il est fonction du mode d'acheminement utilisé pour leur transmission.

5.1.1.1. Acquisition des données sous forme de documents papier transmis par voie postale

L'acquisition de telles données est opérée par saisie manuelle au travers d'un logiciel permettant le contrôle des données saisies, tant dans l'absolu que par comparaison avec les déclarations saisies antérieurement et déjà intégrées dans la base. Toute anomalie donne lieu, grâce à la tenue à jour en permanence d'un fichier des comptables et des signataires recensés dans les installations, à l'information immédiate par téléphone de l'opérateur à l'origine de la déclaration et à la demande d'actions correctives.

5.1.1.2. Acquisition des données transmises par voie télématique en modes BSC 2780 ou Minitel

De telles données sont reçues sur l'ordinateur frontal et font l'objet, deux fois par jour, d'un transfert sur l'ordinateur contenant la base de données. Préalablement à l'intégration de ces données dans la base, celles-ci sont contrôlées par un logiciel qui conduit soit à leur acceptation, soit à leur rejet. Les données rejetées font l'objet d'un compte rendu précisant les raisons de leur rejet. En fin de traitement, ce compte rendu est retransféré sur l'ordinateur frontal et mis à disposition des correspondants. Lors de la transmission suivante, l'opérateur de l'unité émettrice viendra puiser dans un espace qui lui a été préalablement réservé sur l'ordinateur frontal le compte rendu de la transmission antérieure. Ainsi, compte tenu d'un traitement bi-quotidien des données, un opérateur est informé de la validité des informations, qu'il a transmises au cours d'une demi journée J, dans la demi journée J + 1.

On notera par ailleurs que dans le cadre de transfert de matières nucléaires entre 2 correspondants A et B, le système de compte rendu permet aussi à B de recevoir, grâce au DSMR et par voie télématique, la déclaration de livraison émise par A.

En dernier lieu, en fin de mois les documents papier, que les utilisateurs de transmission par voie télématique doivent envoyer également, sont corrélés aux déclarations précédemment transmises par voie télématique, afin de s'assurer de la présence de la preuve de la déclaration au sens juridique du terme, et d'en authentifier l'émetteur par sa signature.

5.1.2. Traitements

Dans le cadre national, le traitement des données reçues a notamment pour but de produire, soit périodiquement, soit à la demande, des listages informatiques destinés à divers utilisateurs.

Mensuellement au 3ème jour ouvrable du mois m + 1 sont ainsi établis et adressés au gestionnaire de chaque sous-ensemble technique, un état récapitulatif des variations de stock prises en compte par le système national au cours du mois "m" et un état des stocks inchangé au cours du mois considéré. Sont ainsi adressés annuellement aux exploitants des installations environ 80 000 pages de ces listages. Les gestionnaires des matières nucléaires des sous-ensembles techniques sont tenus de vérifier ces documents par rapport aux données prises en compte localement et d'informer sans délai le DSMR des divergences constatées.

Est par ailleurs édité avec cette même périodicité mensuelle un état récapitulatif des mouvements de matières nucléaires qui auraient dû donner lieu à une autorisation de transport délivrée par le Ministère chargé de l'industrie. Cet état permet d'effectuer, encore manuellement pour quelques mois mais informatiquement d'ici fin 1991, les corrélations entre les données comptables et l'autorisation de transport.

Enfin et bien que n'entrant pas au sens strict dans le cadre du contrôle national, on notera que sont en outre édités mensuellement :

- des états récapitulatifs des variations de stock et des stocks de matières nucléaires des réacteurs à neutrons rapides gérés en cycle.

- des états récapitulatifs des variations de stock et des stocks de matières nucléaires dont le CEA est propriétaire.

Trimestriellement, sont adressés aux autorités chargées du contrôle national les états des stocks de l'ensemble des installations françaises.

A ces tâches périodiques viennent s'ajouter tous les traitements nécessités pour répondre aux fréquentes demandes particulières :

- des exploitants,
- des autorités,
- des inspecteurs du contrôle national auxquels sont fournis pour chacune de leurs missions, des bilans des matières nucléaires de l'installation contrôlée, ainsi qu'un état récapitulatif des stocks, permettant d'effectuer le rapprochement avec les données comptabilisées localement.

5.2. Traitement des données au titre des contrôles internationaux

Déclaration à Euratom :

Comme indiqué précédemment et compte tenu des disparités des systèmes national et international, notamment au niveau de la fréquence, de la codification, et du support des déclarations, le DSMR procède mensuellement, pour le compte d'une grande partie des exploitants français, (à partir d'un logiciel relativement complexe, compte tenu des disparités), au transcodage en "langage Euratom" des déclarations de variation de stock comptabilisées au titre du contrôle national.

Ce transcodage porte sur les notions suivantes :

- la référence du sous-ensemble technique,
- l'élément,
- la teneur,
- l'irradiation,
- la forme physico-chimique,
- le type de variation de stock,
- l'engagement de contrôle.

Il est effectué pour l'ensemble des installations françaises soumises au contrôle Euratom, exception faite :

- des installations EDF qui effectuent elles-mêmes sous forme papier leur déclaration en utilisant le formulaire annexe n° II du règlement Euratom. Cette situation est provisoire, EDF mettant en place des procédures de transmission télématique de ces rapports au DSMR,
- de l'établissement COGEMA La Hague et de la société EURODIF qui constituent grâce à leur système informatique leurs propres rapports de variations de stock. Ces rapports sont toutefois adressés sous forme télématique au DSMR par le biais du protocole de communication BSC 2780 évoqué précédemment.

Les déclarations transcodées par le DSMR et celles reçues par ligne, sont ensuite enregistrées sur bandes magnétiques, puis adressées par courrier à la Direction du Contrôle de Sécurité d'Euratom, accompagnées d'un listage des données contenues.

Le cryptage de ces données, considérées comme sensibles par les autorités nationales, est à l'étude en vue de leur transmission télématique à Luxembourg.

Déclarations à l'AIEA :

Conformément à l'accord Euratom AIEA (INFCIRC 290), ces déclarations sont établies par la Direction du Contrôle de Sécurité d'Euratom, à partir des rapports comptables reçus des installations françaises, suivant la procédure précédemment décrite.

6. Protection des données et confidentialité

Dans le domaine de la protection des données et de la confidentialité, l'ensemble du système est basé sur les principes suivants :

- équipements installés dans des zones et locaux à protection renforcée,
- séparation physique des fonctions de chacun des ordinateurs,
- accès aux ordinateurs à partir d'identifiants et de mots de passe propres à chaque opérateur,
- accès personnalisés pour chaque opérateur : accès à certains comptes et dans chacun de ces comptes à certaines fonctions (lecture, écriture, mis à jour),

- transmissions télématiques réservées aux informations non classifiées pour les besoins de la défense,

- enregistrement des tentatives de connexion sur un journal des communications exploité quotidiennement.

En ce qui concerne la sécurité de fonctionnement, on notera :

- la réalisation de sauvegardes quotidiennes,

- l'alimentation électrique sur réseau secouru,

- la protection des machines par un onduleur et module de distribution électrique.

7. Les développements futurs

Afin de parfaire l'évolution du système de centralisation de la comptabilité des matières nucléaires aux fins de servir au mieux leur sécurité, diverses évolutions sont prévues ; certaines en cours de test actuellement entreront en service opérationnel d'ici fin 1991, d'autres seront réalisées au cours des années à venir.

7.1. Les développements dont la mise en service est prévue d'ici fin 1991

Ils concernent la généralisation d'une procédure d'accusé de réception et la mise en place de corrélations informatisées entre les déclarations de variation de stock et les opérations de transport et/ou d'exportation.

7.1.1. Généralisation de la procédure d'accusé de réception

Depuis son origine, le système actuel fonctionne en mode débit/crédit pour ce qui concerne les expéditions de matières nucléaires d'un ensemble technique à un autre. En effet, il n'avait pas été envisagé alors de gérer des masses de matières nucléaires en cours de transit sur le territoire national.

Ce crédit automatique, sur le compte du destinataire, des quantités de matières annoncées par l'expéditeur, n'interdisait cependant pas à ce premier de déclarer les éventuelles divergences constatées entre les quantités et qualités des matières annoncées et celles reconnues. Cette déclaration restait cependant une déclaration unilatérale et le chaînage avec la déclaration de livraison d'origine était délicat, notamment lorsque les écarts étaient constatés plusieurs mois, voire plusieurs années après l'arrivée des matières chez le destinataire. Pour ces diverses raisons, une procédure d'accusé de réception va être généralisée d'ici fin 1991. Elle repose sur les principes énoncés ci-après :

Le destinataire doit prendre toutes les dispositions pour connaître de façon précise en quantité et en qualité toutes les entrées de matières nucléaires dans son établissement ou son installation.

Parmi ces dispositions doivent figurer des procédures relatives à la reconnaissance des matières entrant dans l'installation. Compte tenu des moyens techniques et des moyens de mesure dont dispose l'unité destinataire, la qualité et la précision de cette reconnaissance ne peut qu'être progressive dans le temps. Les procédures précitées doivent en conséquence mentionner les différentes étapes de ces contrôles dont le 1er doit intervenir dans les 24 heures qui suivent l'arrivée des matières dans l'installation.

C'est sur la base de ces contrôles préliminaires que le destinataire fera connaître son accord sur les données comptables relatives à l'expédition ou fera état des divergences constatées, en émettant un BDOMN de réception de matières nucléaires.

Celui-ci permettra :

- en comptabilité centralisée de chaîner les déclarations respectives du destinataire et de l'expéditeur,
- d'annuler et de remplacer, par l'accusé de réception, en comptabilité centralisée le crédit temporaire des comptes du destinataire par les données fournies par l'expéditeur,
- de ne créditer le destinataire des quantités et qualités de matières reconnues qu'à la date réelle d'arrivée de celles-ci dans son installation,
- pour l'organisme de contrôle d'être informé sans délai des éventuelles divergences entre expéditeur et destinataire.

Il convient cependant dès à présent de noter que cette procédure n'engagera en aucune façon la responsabilité de l'unité émettrice de l'accusé de réception lors de la mise en évidence ultérieure d'un litige portant sur des éléments d'information dont la vérification n'aurait pas été prévue aux contrôles précités.

Au plan des déclarations faites à Euratom, cet accusé de réception conduira :

- soit à une seule déclaration de type RN, RD ou RF lorsque la réception (accusé de réception) sera déclarée au cours du même mois que la livraison,
- soit à une première déclaration de type RN, RD ou RF générée par le crédit automatique temporaire du destinataire, puis pour la période au cours de laquelle sera déclaré l'accusé de réception à une annulation "D" de la déclaration de crédit temporaire et à une

déclaration d'ajout "A" pour l'accusé de réception.

7.1.2. Mise en place de corrélations informatisées entre les déclarations de variation de stock et les opérations de transport et/ou d'exportation

Au plan de la réglementation nationale, lorsqu'une livraison de matières nucléaires fait l'objet d'un transport, il appartient au transporteur ou à l'expéditeur qui l'a mandaté d'adresser à un service particulier de l'IPSN/DSMR 15 jours avant l'exécution du transport, certains documents en fonction du type de transport effectué et des qualités et quantités de matières mises en jeu. Il s'agit, soit d'un préavis, soit d'une demande d'autorisation spéciale. Après examen, ce service accorde au transporteur et/ou au destinataire, selon les cas, une autorisation d'exécution ou une autorisation spéciale.

Jusqu'alors il était procédé mensuellement et manuellement au rapprochement des informations fournies sur ces préavis ou demandes d'autorisations spéciales avec celles figurant sur les déclarations de variation de stock.

D'ici fin 1991 les références des autorisations d'exécution ou des autorisations spéciales seront communiquées aux opérateurs des installations concernées, charge à ceux-ci de faire figurer ces références sur les déclarations de variation de stock adressées à la comptabilité centralisée.

Le lien entre les transports et les variations de stock sera alors établi avec une grande fiabilité autorisant ainsi des traitements et comparaisons informatisées des données contenues.

7.1.3. Mise en place de corrélations informatisées entre les déclarations de variation de stock et les opérations d'exportation

Certains transferts de matières nucléaires vers l'étranger doivent donner lieu à une notification préalable d'exportation dans le cadre de l'article 24 du règlement d'Euratom ou dans celui d'un accord avec l'AIEA. (INFCIRC 290, INFCIRC 207 add 1).

Cette notification établie sur la base des données fournies par le Service d'application des contrôles internationaux de l'IPSN/DSMR donne lieu à l'ouverture d'un dossier spécifique. La référence de ce dossier sera, par un processus identique au précédent, transmis au gestionnaire de l'unité expéditrice ; il lui appartiendra de transcrire cette référence sur sa déclaration de stock permettant ainsi d'établir un lien étroit entre ces deux domaines et d'effectuer des traitements et comparaisons

7.2. Les développements prévus pour les années à venir

Devraient être entrepris en 1992

* au titre du contrôle national :

- la généralisation du cryptage des transmissions télématiques utilisées avec nos correspondants,
- une nouvelle analyse de sécurité du système informatique, notamment vis-à-vis de la protection contre les intrusions et les virus,
- l'examen des possibilités de retransmission par ligne télématique des documents édités mensuellement et adressés aux opérateurs,
- l'analyse, tant au plan technique que juridique, des systèmes de signature électronique et de télé-authentification visant à se substituer à la signature manuscrite portée sur les déclarations comptables, l'objectif visé est en effet à terme la suppression quasi totale de la circulation des documents comptables sous forme "Papier".

* dans le cadre des contrôles internationaux :

- la mise en service d'une liaison télématique cryptée avec la Direction du contrôle de Sécurité d'Euratom afin de permettre la transmission des rapports de variation de stock de l'ensemble des installations françaises, par ligne, et remplacer les bandes magnétiques utilisées aujourd'hui.

THE COMPUTERIZED NUCLEAR ACCOUNTANCY SYSTEM FOR THE NEW SIEMENS MOX-FUEL FABRICATION PLANT

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Abstract

According to the provisions of the Atomic Law of the Federal Republic of Germany a new production facility designed as largely atomated plant is under construction. For this new facility also the Computerized Nuclear Material Accountancy System had to be newly developed and integrated into a production information system as part of the overall CIM-Concept.

The Nuclear Material Accountancy System has been developed as a "on-line"-system on a Siemens MX 500-75 computer with the UNIX-operating system SINIX and the database INGRES. The chosen relational database system permits a high flexibility in the utilization of the accounting data and in the generation of records and reports. In the first implementation step the system will be operated as a off-line system with a manual data input via PC's at the working stations.

Introduction

Under the provisions of the Atomic Law of the Federal Republik of Germany the new production facility for the Mox-fuel fabrication had to be designed as highly automated plant. Part of the concept to reach the required standard of remote controlled production is the CIM Concept (Computer Integrated Manufacturing) which integrates the data processing systems such as

- production planning (PPS)
- laboratory management (LVS)
- quality assurance (CAQ)
- nuclear material control and accountancy (KMÜ)

into a production information system. Intelligent workstation systems are the interface between the process control and the master control level. These systems are connected with the master control level data procesing system through the SINEC H2B - and/or SINEC-H1-bus. The data processing system architecture is shown in Fig. 1.

Basically, the data processing-supported nuclear material control and accountancy system has the following tasks:

- collecting the data of any movement of nuclear material during the production with the possibility to call at any time the current data of the nuclear material distribution within the production plant by booking them "on site" as real inventory. In addition also the book inventory of the plant and of the material balance areas according to the Particular Safeguards Provisions (PSP) can be called at any time.

- detection of "hidden inventory" and "hold up" by comparison of real and book values in the accounting positions and by analysis of the accounting data collected.

- preventive control for meeting material related restrictions due to license requirements, criticallity control and radiation protection.

- generation of accounting reports according to national and international commitments and regulations as well as the generation of internal documentation and statistics.

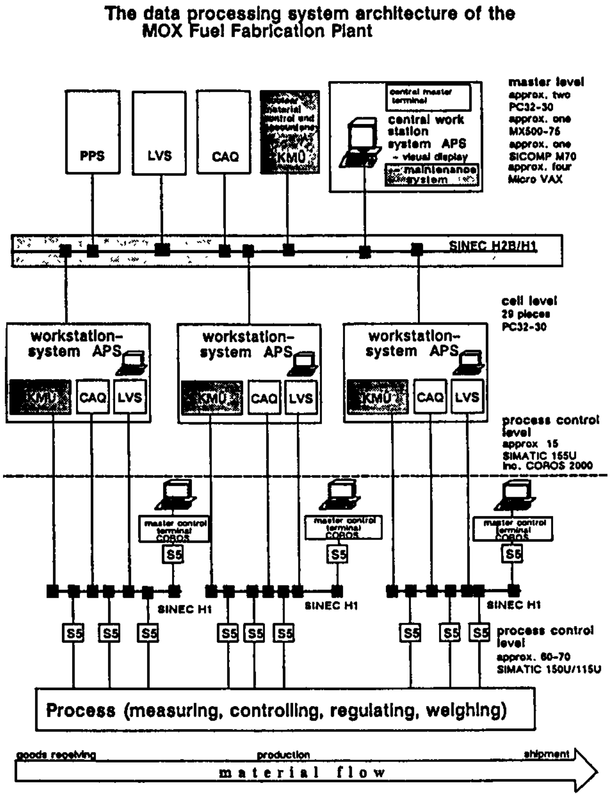

Fig. 1

Scope of functions

The nuclear material control and accountancy system is mainly based on measures and measurements in any case necessary for operation purposes with regard to quality control and quality assurance, supervision of the production and the flow control of material in order to assure the criticallity safety. For the purpose of the material control and accountancy as early as possible, identifiable, countable and traceable items are created in a discontinuous, batchwise production mode. This builts together with the division of the plant into small subunits for accounting purposes the basis of the accountancy system.

The production batches differ from each other by their different production - and quality levels and are divided into individual containers, which can be retraced. The data structures of the material and the location are connected with each other through the transfer function. The individual booking items of the system receive all data of the nuclear material with respect to quantity, batch, production - and quality status or their changes through the protocols about the nuclear material movement from one local balance unit to another.

The total and type of the data compiled on the basis of this principle permit continuous balancing and/or balance checking, with chronologically and also locally delineable resolving power.

The "chronological resolving power" is due to the determination of quality criteria by means of chemical analysis, a time-consuming measuring method which destroys material, and in which furthermore the analysis result is always available only after a certain time delay. This means that calculated or estimated values must be used for balancing or for compilation of an overview of the current distribution of nuclear material until analysis results are available, unless old analysis figures can be extrapolated.

The local resolving power with regard to distribution of nuclear material in the plant is, essentially, a function of the instrumentation, and of the division of the plant and flow into localizable and identifiable sub-units. Equipment and plant hold-up (hidden inventory) remains inaccessible for measurements under running process conditions. However, estimation is possible on the basis of the data obtainable from the accounting system.

Besides, the recording of the data transfer permits the represantation of a so-called "batch history" for each generated production batch. The batch-history shows the beginning, processing and losses etc. of the batches. Thus, it is possible to retrace the material history from the finished product to raw material and backwards.

The history of each container can be retraced and called through its container identification in such way as applied for the batch.

The system also allows to establish for any point in time the nuclear material inventories and their changes for each local area on the basis of the respective preceding inventory. These features of the accountancy system and the datatransfer to the inspectorates as described later in this paper also built an important basis of the safeguards concept for the new MOX-fuel fabrication plant./1//2/

According to the EURATOM-regulation, the book- and real inventories are managed separately from each other. The book inventories are composed of the sum of all inputs and outputs of the plant (or material balance-areas) whereas the real inventories are composed of the sum of booking item's inventories acquired through the data transfer function. The difference between the book - and the real inventories (Material Unaccounted For) results from positive or negative differences found out in the individual accountancy units by keeping the real inventories.

Softwarestructure

The accountancy system (KMÜ) was realized on a SIEMENS MX-500 computer by the use of SINIX as operating system and INGRES as relational database. As tools for programming the system were used

- ABF/OSL (4GL)
- Embedded SQL in C
- Vifred
- Report Writer

The use of a relational database management system allows at any time changes of relation of data in the tables. This special feature in using a relational database system results in a high flexibility in the report system as well as in the evaluation of those data.

Databases of the data processing system for the nuclear material accountancy which are identical, however, independent of each other cover

- the accountancy system of book and real inventories
- a production preparation - and planning sector
- an "inventory" sector.

Within the production preparation sector, it is possible for example, to keep any kind of data e.g. expected receipts or fuel element composition which will be later used in the real system as accountancy data. The purpose of the inventory sector is to freeze the status of inventory taking for later generation of the PIL after completion of the PIV so that after tag check and sample taking the production can go on without interruptions. The access to the individual sectors or the work in these sectors is possible with the same mask system.

The nuclear material accountancy system ist operated through a hierarchical menu tree. The operation is basically objectoriented, that means, the operator chooses at first the data he wants to process (for example a batch) then the function he intends to carry out using these data (for example, filing of new analysis data).

In processing any function, the operator is supported by the system which indicates him the existing data for his data selection. After the operator can select the required data and go into the requested processing function branch.

The menu tree always begins with a central log-in mask in which any operator must identify himself with his identity number and password against the system.

From the central mask, any application specific mask can be reached through data selection masks. This application-specific mask on its turn can have submasks. The nuclear material accountancy system offers the following functions:

- Administration

With the help of the application "Administration" a selected database can be locked and secured. In addition a copy of the LII can be generated.

- Parameterization

Covering the production plant configuration, which means, the definition of system parameters used by any nuclear material production control.

- Master data

This function sector defines the local structures of the plant such as accountancy units, rooms, material balance areas etc. as well as their material related limitations.

- Containers

Containing all information about valid container "names", and their specific data as tara weighs etc.

- Batch

In this section all data about the production batches, analytical results and the history of the batches are kept.

- Transfers

The production uses this function in order to inform the nuclear material accountancy system about any material movements.

- Limit checks

Comparising check for exceeded limits based on the parameterized limits, within data transfer dialogues. An exceeded limit is protocolled and can be called through an information function at any time.

- Real inventory and correction

This function provides above all the Division for Nuclear Material Control And Accountancy with information and is used for making data corrections necessary such as rounding, NT-corrections, determined differences in the real inventories of the accountancy units etc.

- Book inventory

Covering the balance reports such as inventory change - material balance report etc. printed in accordance with national and international regulations.

The protection against unauthorized access to the data inventory of the nuclear material accountancy system and the functions of the system is of great importance. Any user being authorized to have access to this supervision system is recorded in a table where one can see

- the user's name
- user's password (in cryptic letters)
- user class to which the user is allocated

In the user class, following authorizations are specified such as:

- Authorization for access to functions and masks
  autorizations for access to masks specify which masks, a user class is allowed to select. Authorizations for access to functions specify which funtions (function keys) a user class is allowed to initiate (operate) within a mask. In addition to each authorization for access to masks or functions of a user class, it is possible to file an autorization string which regulates the authorizations within the function, for example, within a master data mask. The contents is function-related and is evaluated only by the respective function itself.

- Authorization for access to the terminal
  authorizations for access to the terminal specify the masks and functions which are permissible on a specified terminal in the network.

- Authorization for access to a specified location
  authorizations for access to a specified location specifies for each user class such locations (production locations) where the functions are allowed to be applied.

Any trial for an unauthorized access to the system functions will be recorded in a separate table containing time, function and used identification, respectively pass-word.

Accountancy data transfer to an "In Field PC" of the inspectorates

For inspection and verification purposes the inspectorates will be supplied with a modified version of the system keeping only the safeguards relevant data as operators declaration running on a PC. Via a periodic data transfer by floppy disc or tape the inspectors will have available for their inspections acivities

- the actual material distribution in the facility (real inventory)
- the book inventories
- the running MUF-account
- transfer records
- records of any correction effecting book or real account.

In addition all data defining the system as parametrization, master data, container and batch data will be updated.

The modified version does not allow any change of data but contains the same record and report system. Due to the availability of the transfer records the inspectorates can select information needed to be compared with independently collected verification results.

The system includes an additional dBASE-interface for use of selected data in subsequent MS-DOS systems.

Hardware configuration

The nuclear material accountancy system has been designed as central database system where the material flow is acquired on decentralized workstation systems. In order to achieve a high safety degree against hardware components failure, the production is subdivided into cells to each one of them is allocated an intelligent workstation server. Here we started from the idea to maintain the material flow within each production cell, should the central system not be available. Each workstation server has the production data of its production cell. This means that all data are maintained twice within the whole network, on the one hand on the workstation systems and on the other hand, on the central computer.

The material flow is acquired through a screen mask on the individual workstation systems (PC 32.30). The workstation PC is connected with the nuclear material accountancy system computer through the SINEC H2B and SINEC-H1-bus. The operating system UNIX is used for these workstation systems. Some workstation systems are used as cell servers thus having their own database where their respective central database section is kept as redundant.

/1/ E. Haas, W. Hagenberg, A New Safeguards Concept at ALKEM MOX Fuel Fabrication Plant, Proc. 3rd Int. Conf. on Facility, Operation-Safeguards Interface, San Diego, 1987, p. 51

/2/ E. Haas, W. Hagenberg, M.J. Canty, Material Control and Accountancy at the Mixed Oxide Fuel Fabrication Plant Siemens Brennelementwerk Hanau, Proc. of the 30th Annual Meeting of the INMM, 1989, p. 745

AN INTEGRATED APPROACH TO PROCESS INFORMATION, NUCLEAR MATERIALS CONTROL AND ACCOUNTING IN BNFL's THORP FACILITY

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ABSTRACT

The paper descibes the integrated computer control system on British Nuclear Fuels' new Thermal Oxide Reprocessing Plant at Sellafield. It explains the hierarchical structure and the rôle of the major components.

The paper provides an outline description of the "Conventional" Nuclear Materials Control and Accountancy and the on-line Near Real Time Materials Accountancy Systems.

1 INTRODUCTION

British Nuclear Fuels' Thermal Oxide Reprocessing Plant (THORP), presently under construction at Sellafield in the UK, is the largest current process plant project in western Europe, with a design capacity of 1200 ton of Uranium per annum. The reprocessing plant is due for completion in 1992, and the fuel receipt and pond storage facilities are already operational.

The plant will operate as an integrated facility that includes Fuel Receipt and Storage, Head-End Plant, Chemical Separation, Purification, Finishing for both Uranium and Plutonium and waste handling processes prior to conditioning and subsequent storage of the waste in other facilities.

With the scale of THORP, marshalling together the inordinate quantity of process information and operational actions to a single location is a considerable task. The use of computer-based equipment has clear advantages and has been incorporated into an integrated control and information systems structure.

2 HIERARCHIAL SYSTEMS STRUCTURE

At the top of the hierarchical systems structure is the Chemical Plant Information Computer (CPIC). As well as CPIC, the hierarchical environment contains a large Distributed Control System (DCS), a Head-End Central Computer (HECC), an Automatic Sampling Computer (ASC), Receipt and Storage Information Computer and over a hundred programmable logic controllers. The major components reside on a local area network which also provides external connection to other facilities on the Sellafield site including:-

- Sellafield Laboratory Information Management System (SLIMS);
- Encapsulation Plant Information Computer, and
- Sellafield Nuclear Materials Accountancy and Safeguards Office (NMASO) Computer.

The network is based on the proprietary Ethernet data network and a simplified schematic of the network is shown in Figure 1. Communication is executed using the proprietary software packages SQLNET and DECNET.

3 SYSTEM MAJOR COMPONENTS

Chemical Plant Information Computer

The CPIC provides, as one of its rôles, the nuclear materials control and accountancy function for the THORP Head-End, Chemical Separation and Uranium and Plutonium Finishing. In addition to this rôle, the CPIC is required to provide the following facilities:-

- Production Planning;

- Data Archiving;
- Decision Support;
- Solvent Accounting;
- Tank Sentencing Data Analysis;
- Vessel Calibration Calculations, and
- Daily, Weekly and Monthly Report Generation.

Plant data that are required to perform the above tasks are received from the other systems on the network within a real-time environment an outline diagram of which is shown in Figure 2.

Distributed Control System

The bulk of the data coming to the CPIC will be from the DCS[1&2]. Plant parameters such as tank levels, temperatures, pressures, densities, weights, vessel status, etc. from each of the plant areas are stored by the DCS onto six associated historic recorders. Accordingly, both current and historic data records relating to process parameters are available to the CPIC, via the network, for subsequent analysis and interpretation.

Head-End Central Computer

Within the Head-End Mechanical Plant individual fuel element movements are tracked from building entry, at each stage of transfer, up to the dissolver batch by the HECC. When a rack arrives at the Head-End hand-over point, the fuel is validated against the current customer campaign before entry is allowed. The fuel is then moved through the feed-pond processes to the removal stations. The individual fuel elements are removed from the Multi-Element Bottle (MEB) or container and monitored. The fuel monitoring system is fed details about the fuel element from the HECC. The monitoring results are stored by the HECC againest the fuel element details. If the fuel element passes the set criteria for clearance to shear, the fuel element is allowed to transfer to the Shear Cave. When the fuel element is ready for shearing the HECC feeds the local control system with the associated cutting pattern for that fuel element. As the fuel element is sheared, the HECC records which dissolver and basket the fuel is dissolved in and allocates a unique batch number. Regular detailed reports are transmitted to the CPIC covering the fuel movements.

Automatic Sampling Computer

The Autosampling System[3] provides an automatic sampling facility for the chemical separation process and informs the CPIC each time a process control or accountancy sample is taken. The majority of the samples are scheduled but the system provides the facility to request additional samples when required; e.g. repeat samples for laboratory purposes.

Sellafield Laboratory Information Management System

Sometime later, dependent on the type and complexity of the analysis, the results of the analysis are sent by the SLIMS computer to the CPIC. This information is used in determining calibration factors, in determining the suitability of liquor for conditioning and tank sentencing and by the nuclear materials accounting function in deriving Uranium and Plutonium concentrations.

4 AN INTEGRATED APPROACH

Production Planning

The CPIC maintains the production plan, containing all fuel details required to run the overall production campaign and specific Head-End campaigns for onward transmission to the HECC. Collation of the production plan is initiated by the supervisor creating a list of MEBs/containers required for processing. The list is accessed by the NMASO computer which compiles futher information about the individual fuel elements from its records and transmits this back to the CPIC. Additional information is added by the supervisor as necessary, and only a supervisor can change the order or contents of the plan, although the plan is available for viewing by operators at all CPIC terminals. The CPIC transmits to Receipt and Storage the identification numbers of the required MEBs/containers for the planned Head-End campaign. The Receipt and Storage computer will use the information to identify the storage rack and hence the location of the required MEBS/containers in the storage ponds. This enables the work in Receipt and Storage to be planned to match the requirements of Head-End.

5 NUCLEAR MATERIAL CONTROL AND ACCOUNTANCY

The performance of the THORP materials control and accountancy system relies on the automatic recording and analysis of accountancy data taken by the CPIC and its subordinate computer systems from the plant instrumentation throughout THORP. Specific measurement requirements have been specified for identified vessels and at product filling stations solely for accountancy purposes. Within CPIC, two types of nuclear materials accounting will be provided:-

- "Conventional" Special Nuclear Materials Accountancy for Plutonium and Uranium, and
- Near Real Time Materials Accountancy (NRTMA) for Plutonium.

"Conventional" Materials Accountancy

For nuclear materials control and accountancy purposes the plant has been divided into a number of physically discrete accountancy areas which will form the basis of the Material Balance Areas (MBA's) for safeguards purposes. A schematic diagram of the arrangements is shown in Figure 3 and depicts the five main areas within the THORP complex.

Area 1/2 - Feed-Pond which includes the pond area, from Rack Transfer Machine handover to the Fuel Elevator. Head-End which includes the Fuel Shear, Dissolvers, Basket Handling and Hulls Monitoring equipment and Feed Clarification. The Head-End Accountancy Tanks (23 cubic metres) equipped with weight and level measuring systems are also located in this area.

Area 3 - The main Chemical Separation Plant, including HEP/SEP Buffer Tanks, Uranium and Plutonium Extraction and Purification Plant, Uranium Finishing and UO_3 drum weighing, Plutonium Nitrate Accountancy Harp Tanks and Liquid Waste Treatment systems.

Area 4/5 - Plutonium Finishing including Plutonium Nitrate Buffer and Feed Tanks, Plutonium Finishing Line and product weighing and preparation of Product Can for storage. Additionally, there is a Plutonium-Contaminated Material(PCM) handling area and storage capacity for Oxalate Mother Liquors (OML) from the finishing process. Plutonium Can buffer storage area including systems for can inspection prior to storage or export.

Area 6 - Main Plutonium storage area.

Area 7 - Main Uranium storage area.

In each case, transfers of materials between such areas, or from these areas to adjacent facilities, will be automatically identified by the subordinate plant control systems and signalled to the CPIC. Within the Chemical Plant materials tranfers from vessels across material boundaries are controlled and require authorization by operations personnel through the DCS. These transactions together with known steady state conditions trigger "Accountancy Transfer Reports" from the DCS to the CPIC. The reports are unique for each combination of issuing and receiving vessels. During the plutonium finishing line processes of can filling, packaging and storage, product accountancy data are collated, and validated. Data validation consists of checking can weights are within set limits, checking cans are in their expected locations and can numbers are within the range specified for the type of can. This data collection and validation is performed by the local control systems which regularly transmit the information up to CPIC through the DCS. Similar accountancy facilties are provided in the Uranium drum handling and storage areas with drum details being transmitted by the local control systems up to CPIC through the DCS. Such data, together with information from other THORP computers systems, are collated and stored in a relational database on CPIC. This will ensure that a single, integrated database of accountancy information is held on CPIC to meet the various statutory, commercial, materials accountancy and safeguards requirements under which THORP will operate.

The conventional accounting facility produces the physical inventory at the end of a production campaign and maintains complete transaction lists for all the materials balance areas throughout THORP. This information is transferred to NMASO to assist in the compilation of its sitewide accounts and reports.

An important consideration of the design objective is to ensure that the materials balance for individual areas can be rapidly updated to facilitate high standards of materials control. To that end, BNFL's NRTMA system will play a significant rôle.

Near Real Time Material Accountancy

BNFL has been engaged in the development of NRTMA since the early 1980s and has had a functional prototype system available for two years. That system is currently being restructured and enhanced for use in the Chemical Separation and Plutonium Finishing Areas of THORP. It will reside within the CPIC enviroment as an on-line facility. Accordingly, data collected and processed for "Conventional Accountancy" purposes will be readily available to the NRTMA program modules, together with addional information required by NRTMA, for further analysis. In all, the following data sources will be available to, and analysed by, the materials accountancy tasks on CPIC:-

- Plant instrumentation readings in engineering units (temperature, density, level and weight);
- Vessel status information from which accountancy transaction data "Events" can be determined;
- Vessel calibration data;
- Measurement uncertainty data for use in error propagation modules;
- Process models to derive the plutonium inventory in pulsed columns, evaporators and inter-vessel pipework;
- Data from the Plutonium Inventory Monitoring System (PIMS) installed in the Plutonium Finishing Line, and

Analytical data from plant samples.

Relevant data from each plant area will be processed by the NRTMA facility through a common series of program modules (although certain modules require optimization for specific reprocessing campaigns). The overall structure of the NRTMA facility is shown in Figure 4, and explained in more detail in other papers[4-6] being presented at this Symposium. Overall, however, the design philosophy of the NRTMA system developed by BNFL is based on the following four objectives:-

- To automate and integrate the capture and analysis of process data, thereby minimising operator involvement;
- To present the results of analysis in a clear, uncluttered manner to facilitate the use of NRTMA as a materials control tool;
- To ensure the facility is robust, both statistically and in respect of plant operational conditions, and

- To indicate, on the basis of the available information, the quality of the NRTMA balance derived by the system (in the event of spurious or missing data values).

The Near Real Time Materials Accounting Facility duplicates that of the "Conventional" nuclear accounting, but only in the plutonium materials balance areas. The significant difference between the facilities is one of timing, with the NRTMA facility being capable of determing the inventory in "near real time" while the plant is operational.

6 OPERATOR INTERFACE

The operator interface is via dual session, colour visual display units. These are located throughout the building but in particular within the management centre, the display enviroment is a menu driven hierarchy. The outline diagram of which is shown in Figure 5.

A menu display is a list of textual titles of options. Selection is by positioning of a cursor against the option, or entry of an associated screen number. The hierarchy allows restrictions to be placed on which personnel are authorised to access the menu and provides menus aimed at specific process cycles or facilities. Movement down the hierarchy provides either a more specific menu, or the final detailed "form". Information is accessed via the menu hierarchy and presented to the user in "form" type displays. These displays detail the database information in a simplified way. A "form" is as screen layout with a "fill-in-the-blanks" or "view-only" format of data, allowing information to be entered or queried. Access is password protected, with three levels of access:- Operator; Supervisor and Maintenance. One branch of the menu hierarchy covers accountancy whilst another branch is provided for the Euratom Safeguards Inspectorate to access agreed information.

7 QUALITY ASSURANCE

BNFL's engineering procedures require the process and mechanical offices to supply source documentation. The source documents were used to develop the THORP Division nuclear materials accountancy and safeguards strategies, thereby allowing the definition of the Works Accountancy Areas, the process information and the instrumentation required to determine the required accountancy transactions discussed earlier. This source information was then developed into the system's equipment definitions and specifications.

In order to produce computer software that operates as intended it is essential that the requirements are specified in a comprehensive and unambiguous manner. Accordingly, to ensure a quality product, quality assurance has to be applied throughout the life cycle of a system development on THORP. BS5750(Part 1) has been mandatory on all the control and information systems, and, via BNFL's engineering procedures, has required the System and Equipment Engineering Department to produce detailed User Requirements Specifications for each system.

The functional requirements for CPIC were defined in english text but were supported by a system context diagram, data flow diagrams and data table structure. In addition it was necessary to define and document the interface requirements to the other computer systems on the network.

The system supplier (in the case of the CPIC it was BNFL's own Corporate Management Services) used this document as a reference document for the production of a System Definition Document, which detailed how the requirements were to be met, including the logical data model, functional breakdown, screen layout, forms, menu structure, detailed table structure and sizing. Following approval, the system supplier then entered the detailed design stage, producing the physical data modeland the module design, leading to detailed software design specifications for each of the modules identified. The output was a system context diagram, entity-relationship diagrams, data flow diagrams, a data dictionary and module specifications.

The implementation phase then followed where software programmers used the detailed program specification to code and annotate the application software. A case tool embodying a data flow and data dictionary design methodology was used throughout the software design and implementation phases. The test requirements consisted of a comprehensive test plan which identified unit testing, integrated testing and final customer acceptance testing.

The CPIC software, in common with that for each of the information computers on the network is built around the same relational database software package utilising the associated fourth generation language and utilities. The use of the same well proven database and communications software packages throughout the integrated system design facilitates coding, testing, and commissioning, which, together with common hardware from the Digital range of VAX machines and terminals, provides an integrated system with high integrity and lower maintenance.

8 CONCLUSION

Overall, the adoption of a fully integrated control system on THORP provides BNFL with a powerful information system. It has been fully utilised to provide a reliable nuclear materials control and accountancy function.

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Figure 1

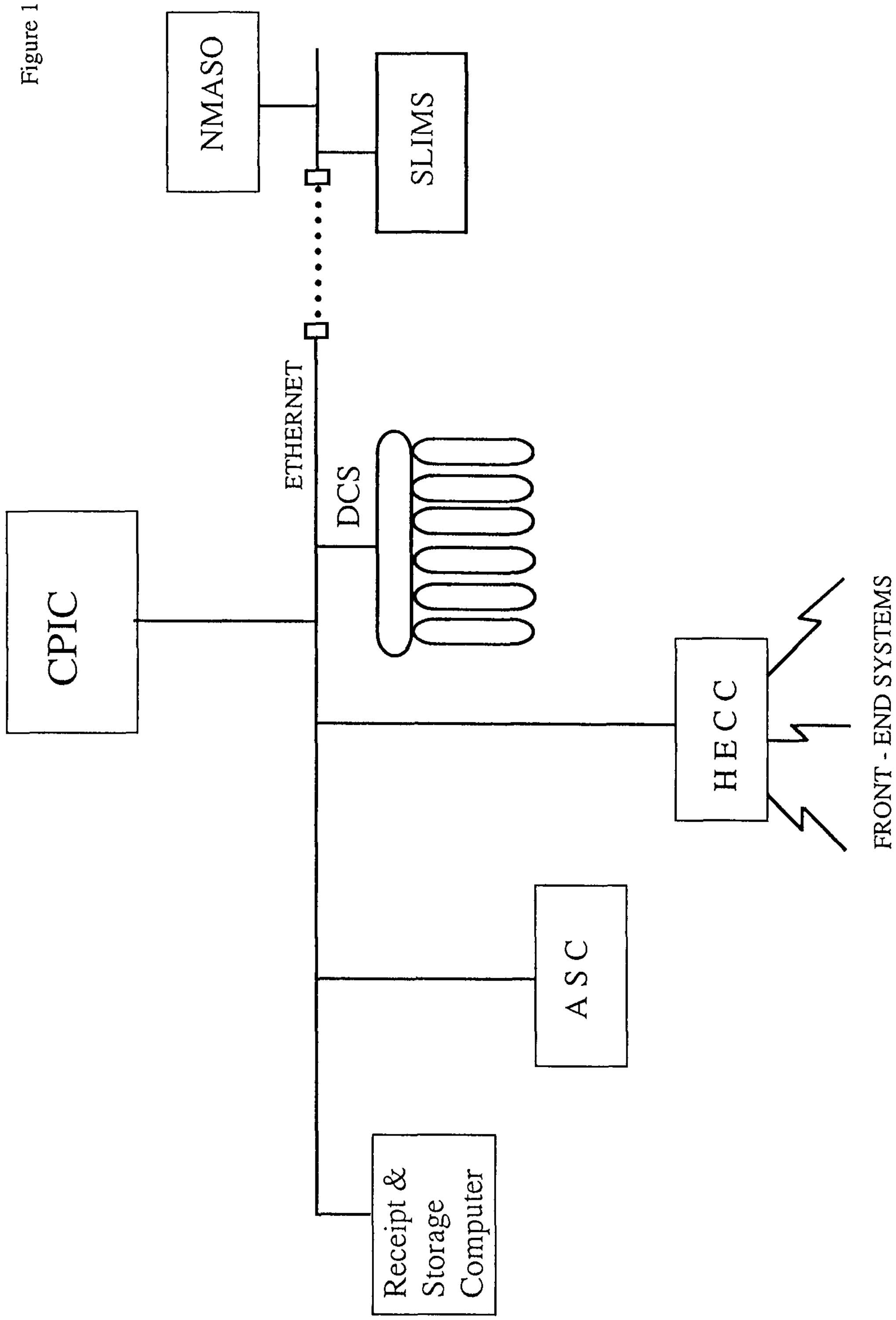

NMASO
SLIMS
ETHERNET
CPIC
DCS
HECC
FRONT - END SYSTEMS
A S C
Receipt & Storage Computer

Figure 2

CPIC Enviroment

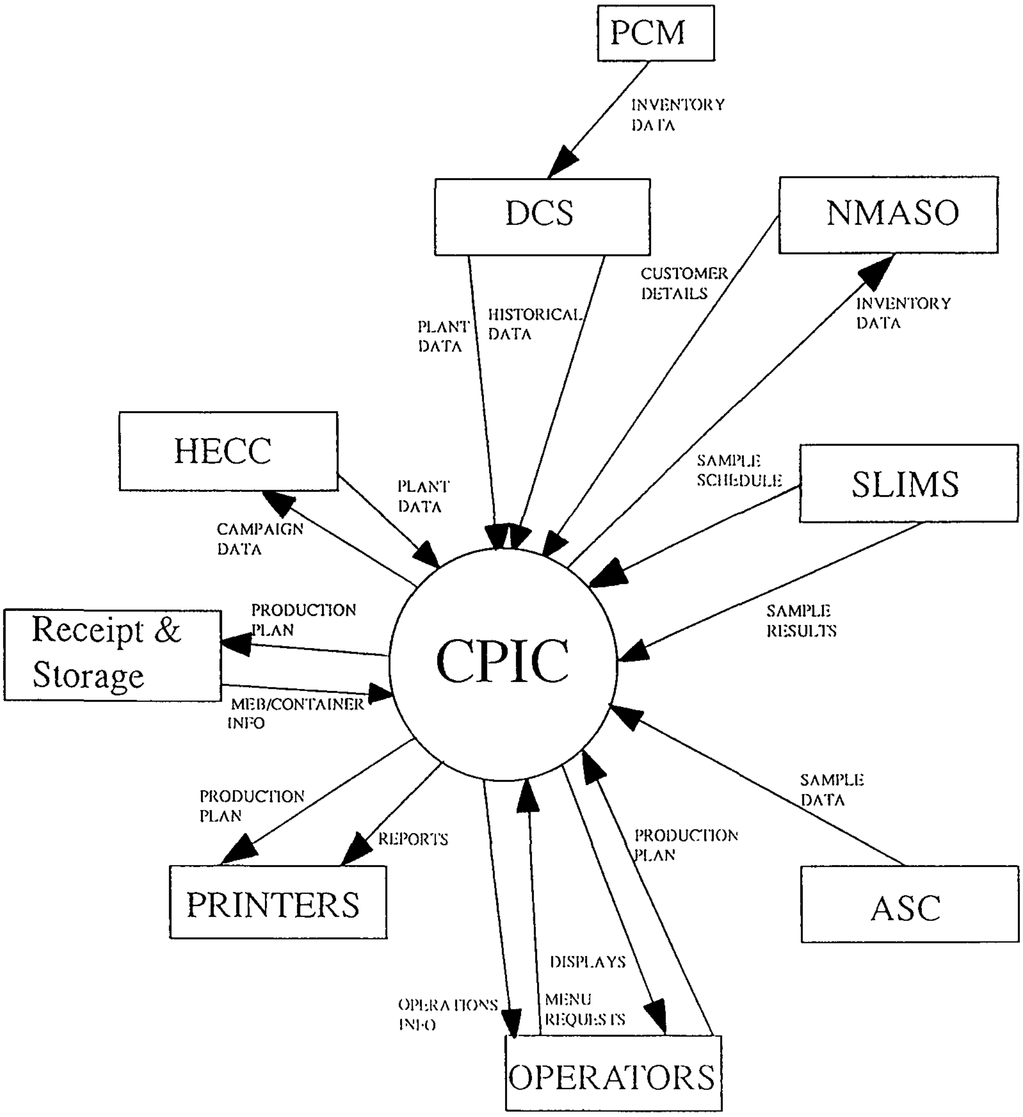


Figure 3

THORP Accountancy Structure

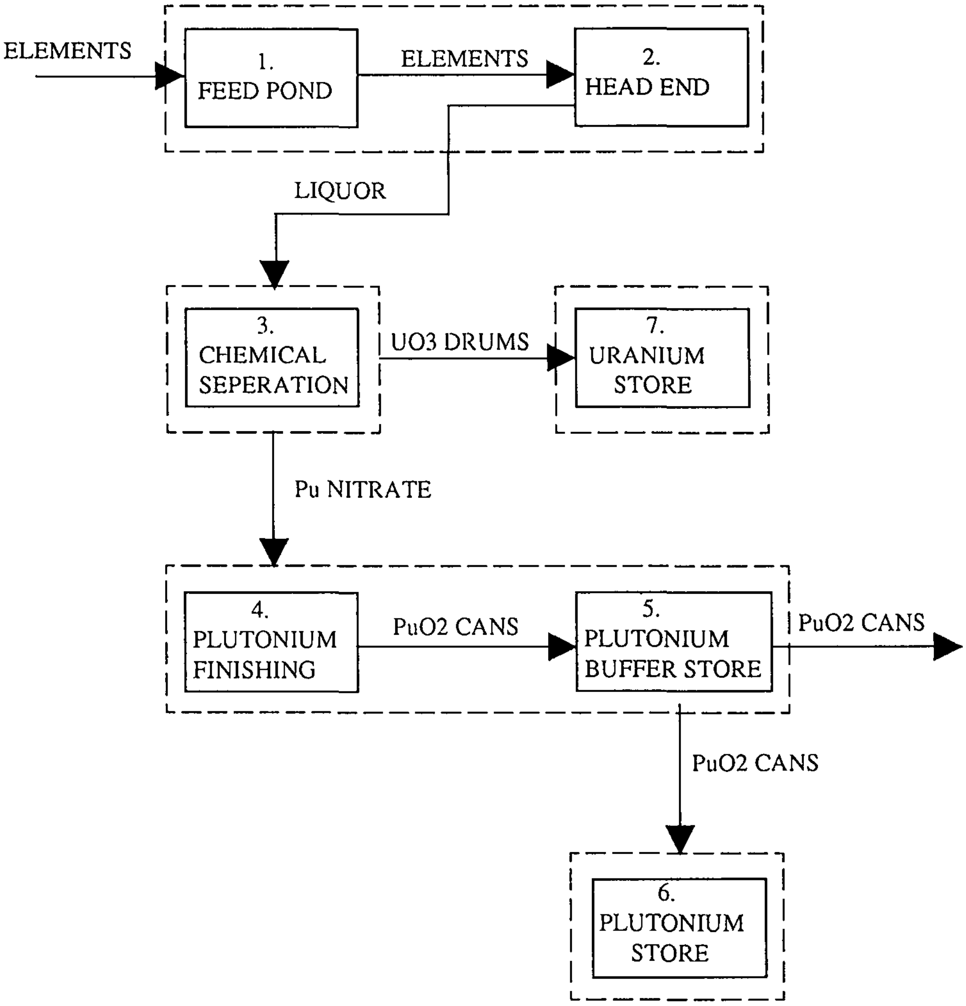


Figure 4

NRTMA STRUCTURE

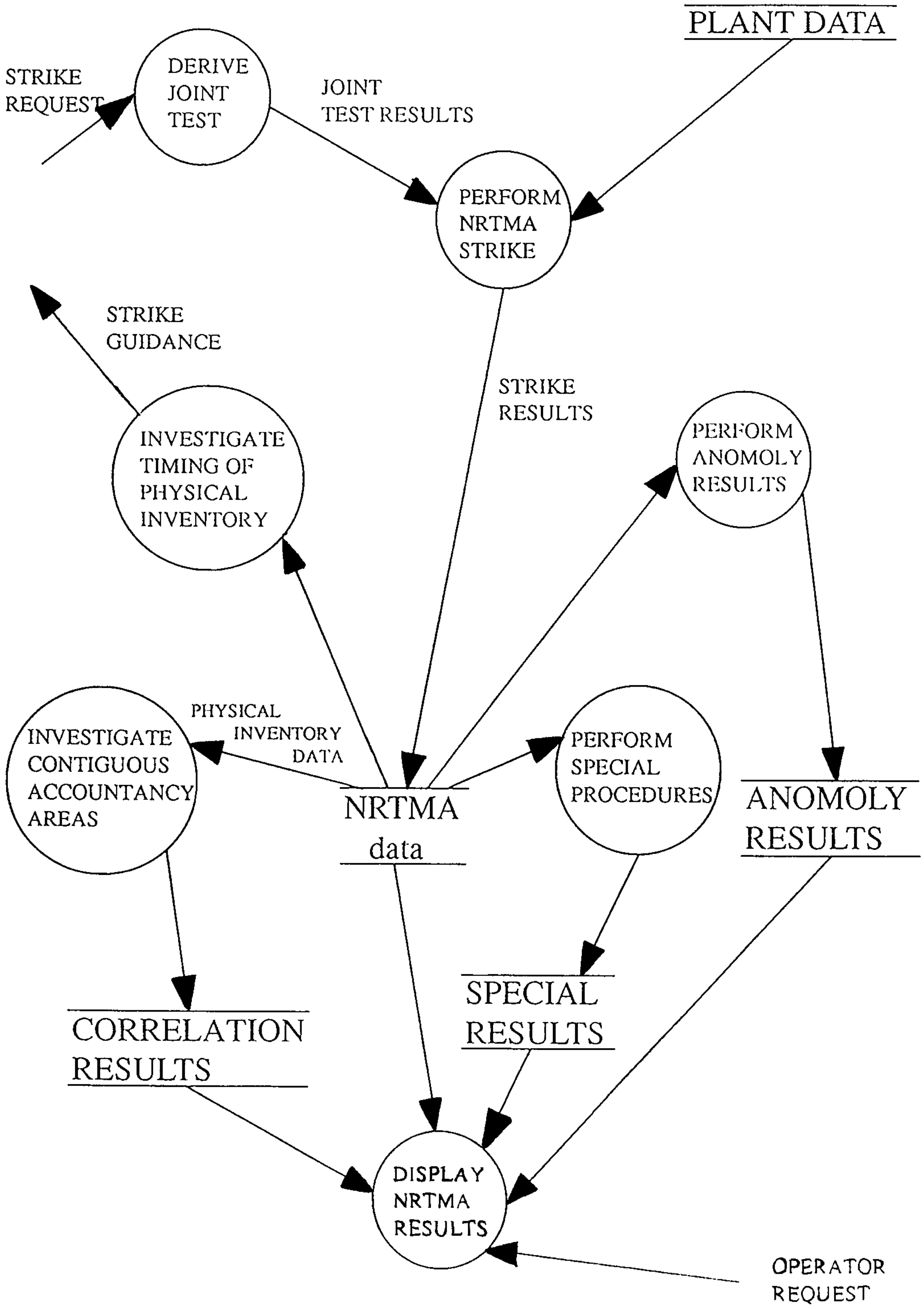
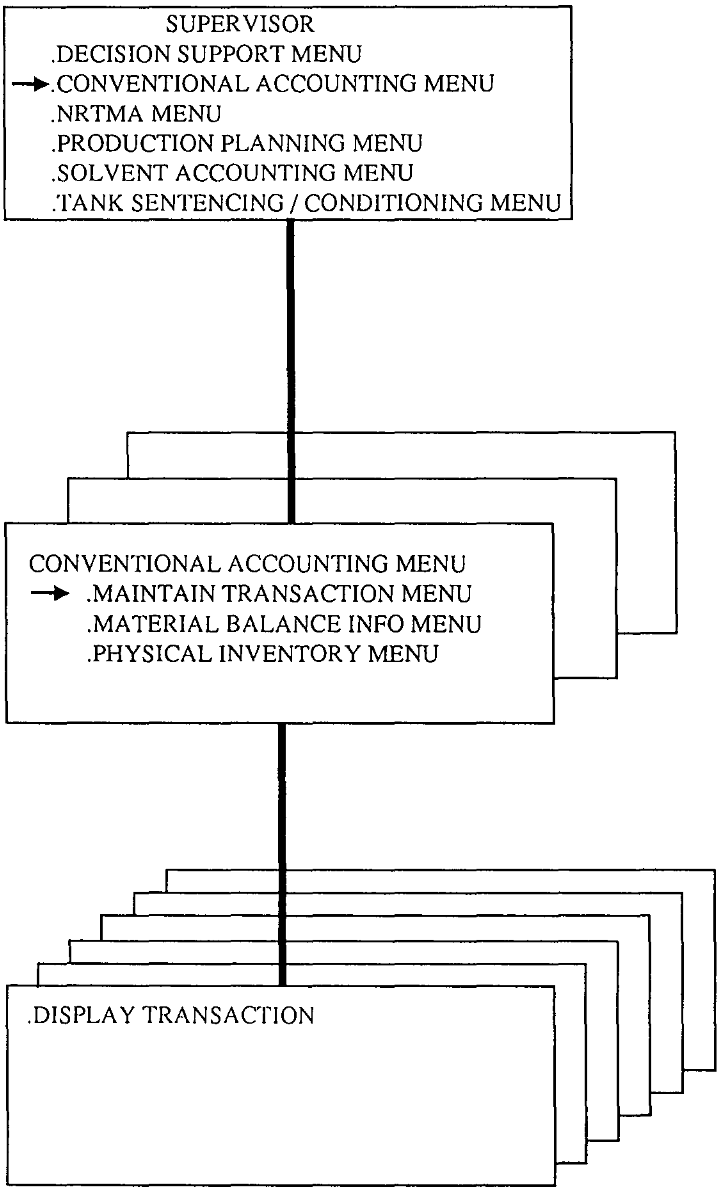


Figure 5

CPIC Menu Hierarchy



NEAR REAL TIME MATERIALS ACCOUNTANCY DEVELOPMENT PROGRAMME FOR THORP

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ABSTRACT

BNFL is currently designing and installing a fully automated system of data capture, storage, and processing for its Thermal Oxide Reprocessing Plant (THORP) at Sellafield. A prototype Near Real Time Materials Accountancy (NRTMA) system has been used to demonstrate the advantages of this method of materials control to the future plant operators and their feedback continues to be incorporated in the development of user interfaces. NRTMA has been included in the User Requirements Specification for Chemical Plant Information Computer, the top-tier computer which is being provided to archive, retrieve and analyse plant data.

The paper describes a development programme of performance and quality related improvements to the prototype NRTMA system. Furthermore, advanced diagnostic systems are described which will help the operator in the resolution of anomalies.

1 INTRODUCTION

Since the early 1980s, BNFL has been committed to the development of Near Real Time Materials Accountancy (NRTMA). NRTMA holds the greatest potential for improving materials control and accountancy in large commercial plants handling high strategic value materials and, therefore, THORP has always been seen as the main target for the benefits of this development.

At the last ESARDA symposium, the components of an NRTMA system for a commercial plant were described[1]. Since that time a major achievement in the development of a system for operational use has been the building of a prototype NRTMA system[2].

2 THE CHEMICAL PLANT INFORMATION COMPUTER

BNFL is currently designing and installing a fully automated system of data capture, storage, and processing for THORP. The components of this integrated computer system are described in another paper[3]. At the top of the hierarchy is the Chemical Plant Information Computer (CPIC) which is provided with functions for materials accountancy and control.

3 DEVELOPMENT OF NRTMA

Two years ago, a prototype computerized NRTMA system was developed in BNFL's Safeguards Section, using simulated data corresponding to steady plant operation, in order to establish the fundamental factors affecting the system's performance. Using the prototype NRTMA system, the Safeguards Section has been able to demonstrate how the advantages in statistical analysis of materials accountancy data can be made available to plant operators and non-specialist staff by means of a comprehensively engineered software package.

The prototype system has been fully validated and, under ideal flowsheet conditions, would meet the NRTMA requirements for THORP. However, from a materials control point of view, most plants are unlikely to operate under ideal conditions for the following reasons:-

- the in-process inventory will fluctuate due to normal plant behaviour, and as batches of material are received and despatched;
- the disposition of the inventory, and its measurement uncertainty, will vary with time, and
- the sizes, and the intervals between successive receipts and despatches of material will vary.

Accordingly, BNFL has adopted a strategy which has the following aims:-

- to minimize the amount of manual effort required by operators to set up test parameters, and
- to carry out initial interpretation of results.

In order to meet these ends, a number of performance/quality related improvements to the prototype NRTMA system have been identified which will increase the effectiveness of the THORP materials accountancy and control system. These improvements are discussed in detail later. Furthermore, advanced diagnostic systems will be provided to help the operator in the resolution of anomalies.

In order to support the developments described above, it is necessary to employ a model which can be used to simulate data more representative of real plant operating conditions. The basis of such a model existed in the form of the THORP stochastic models already being used to support plant design. However, some enlargement of the model was needed so that the necessary simulated data could be made available for further development and testing of the NRTMA system.

The NRTMA Development Programme for THORP comprises:-

- Migration of the Prototype System to VAX Hardware;
- Joint Test Generation;
- Timing of Physical Inventories;
- Error Propagation;
- Anomaly Resolution, and
- Enlargement of the Stochastic Model.

4 MIGRATION

The first element of the NRTMA Development Programme was the migration of the prototype NRTMA system from the IBM PC, used for the research hitherto, to VAX hardware, as specified for the CPIC. In addition, the structure of the NRTMA system was reviewed to bring it in line with the CPIC design, and similar Quality Assurance procedures adopted.

5 JOINT TEST GENERATION

From necessity, the performance of the prototype NRTMA system was based on flowsheet parameters, and assumptions made in regard of instrument performance, balance frequency and false alarm probability. When THORP is operational, some of these assumptions may prove inappropriate and parameters will vary between campaigns. This may have a significant effect on the performance of the NRTMA system, which can only be overcome by customizing and recalculating the test parameters for specific campaigns and Works Accountancy Areas (WAAs).

The Joint Test Generation function is being developed to allow the test parameters to be customized for specific campaigns and WAAs. This development will also allow the operator to ask "what if" type questions to predict the effect of changes in the operating conditions or measurement system.

The required false alarm probability must be specified before the Joint Test can be generated. Suppose that one Joint Test is generated for a campaign false alarm probability of 5%. It would then be possible to generate an alternative Joint Test for a campaign false alarm probability of, say, 10%. This second Joint Test, on account of its higher false alarm probability, will have a higher response. An alternative way of looking at this is that the second Joint Test will respond earlier to a protracted event (albeit with a higher risk of false alarm). This approach introduces the concept of an "amber" and "red" alarm. On the "amber" alarm, the operator is alerted that something may be wrong, and he has the opportunity to make preliminary investigations. The "red" alarm alerts him, with more certainty, that something is wrong, and action should be taken.

As part of this approach using two alarm levels, each Joint Test would have its corresponding set of Page's test statistics and the operator could be provided with additional screens and print-outs from the statistical analysis module. However, it would be clearer for the operator if this additional information could be presented without the need for additional statistics and screens. The Joint Test generation procedure is being extended in such a way that the two Joint Tests will use the same Page's test statistics but two different *H* values (alarm levels) corresponding to the two campaign false alarm probabilities. In this way, the additional alarm levels can be clearly displayed without the need for additional displays.

6 TIMING OF PHYSICAL INVENTORIES

The sole purpose of doing frequent in-process inventory determinations is to achieve good results and to demonstrate control over nuclear materials in a timely manner. A major factor which determines the response of the Joint Test is the uncertainty associated with closing the materials balance. The materials balance error is made up of contributions from the opening and closing inventories, and from the transactions taking place between the two in-process inventories.

The quality of the NRTMA system will depend on the timing of the in-process inventories, and will be influenced by factors such as:-

- the uncertainty in measuring the inventory;

- the uncertainty in measuring the cumulative throughput since the previous in-process inventory;
- the time since the previous inventory, and
- the time available to take the remaining inventories in the campaign (to be consistent with the number of balances stipulated at the beginning of the campaign).

The effect of these variables is not always simple or obvious. A reduction in the NRTMA system performance could occur if sub-optimal timings for in-process inventories were to be chosen. To avoid this happening, a function is necessary which will analyse the prevailing plant conditions and recent plant inventory and give readily usable advice to the operator to allow him to make the final choice for the timing of each in-process inventory.

There are a variety of ways of selecting times for the taking of physical inventories. One way would be to divide the time between the beginning and end of the campaign into the required number of balance periods and then calculate the appropriate times for striking the materials balances. Another way would be to strike the materials balance at a certain time every so many days. The basis of the Timing of Physical Inventories function is that the expected performance of the NRTMA system can be calculated at a greater frequency that that which is necessary to achieve the requisite number of balances in the campaign, and the decision of when to actually calculate the materials balance and apply the Joint Test made after analysis of this expected performance data. It is important to note that the materials balance would be calculated, possibly retrospectively, when the plant operating state was judged to be most favourable for the response of the NRTMA system. Furthermore, the NRTMA system does not demand that balances be taken at regular intervals of time.

Data will be available from the stochastic models and will be used to develop rules for evaluating the expected performance of the Joint Test for different timings of the in-process inventory. An important achievement for this function will be to show how the optimum times for calculating the materials balances can be proposed by the NRTMA system. Furthermore, it will be demonstrated that the response of the NRTMA system, using the timing of physical inventories module, is greater than if materials balances were taken at regular intervals, or when the plant was in a "standard state".

7 ERROR PROPAGATION

The prototype NRTMA system applies elementary measurement error analysis procedures. This function is being developed so that correlations in the data can be identified, calculated and then taken into account to obtain more realistic expressions for overall measurement uncertainties. False alarm probabilities, test procedure responses and anomaly resolution procedures are strongly influenced by measurement uncertainties and the incorporation of sufficiently rigorous error propagation procedures is a fundamental requirement. The error propagation function will require no input from the operator.

The only correlations taken into account in the prototype system are the negative correlations between successive balances. These correlations occur because of the common inventory determination. In total, there are simply one less than the number of balance periods of these correlations, and they are stored as a vector. When more extended correlations are considered, this method of storing the correlations is not suitable. A matrix is necessary. The first stage in providing for extended correlations has been the setting up of a matrix for storing extended correlations, and developing supporting software routines. This modification has already been incorporated.

The study of error propagation has been started by considering movements of highly active liquor from the Head-End to the HEP/SEP Buffers, and of plutonium nitrate concentrate within the harp tank suite. It is in such areas, where batches of liquor will be mixed and the concentrations calculated from previously recorded data for the components, that the error propagation process will induce correlations large enough to be important for the correct calculation of inventory uncertainty. The sources of significant correlations in the data can only be identified by examining campaigns of data in populated data base tables, and developing procedures for extracting those elements required for the calculation of the correlations. The THORP Stochastic Model will be used to generate data to allow error propagation procedures to be developed.

8 ANOMALY RESOLUTION

The anomaly resolution function in the prototype[4] system considers, one at a time, the following plausible models:-

- an abrupt loss/gain at any period;

- a protracted loss/gain beginning and ending at any periods, and
- an inventory error at any inventory determination.

New procedures are being developed to consider models with two or more components. The benefit for the operator is that, compared with the prototype system, it will be possible to present more information about the pattern of an anomaly. No additional input will be required by the operator to get this additional information.

Whereas the anomaly resolution function in the prototype system has only been able to consider plausible models one at a time, the extended function will enable conditions likely to occur on operational plant to be considered. For example, a bias on throughput determination may have been occurring for a number of balance periods when an error is made in determining the in-process inventory. In such circumstances, the anomaly resolution procedure must be able to recognise contributions to the overall anomaly from a number of sources if it is to generate the most useful diagnostics. The theoretical basis for fitting composite anomaly resolution models has already been developed. Consideration is now being given to how the additional information should be presented to the operator.

The anomaly resolution function in the prototype system was developed using data from a simulator which assumed steady flowsheet conditions. Each balance periods was taken as the same length, and regular throughput was assumed each balance period. The anomaly resolution procedure will be modified to accomodate plant conditions where the timing of physical inventories will be chosen to optimize the response of the NRTMA system, and throughput transactions will not necessarily occur at regular intervals.

The operator will be permitted to propose his own "models" for inclusion in the evaluation and ranking. The operator may receive information, outside of the NRTMA system, about the quality of data fed to the NRTMA system. For example, he might be informed, after a calibration check, that an instrument had been performing so as to introduce a bias or an additional measurement error, or he might be alerted to a mistake in an analytical result. The operator will wish to evaluate such information by examining its influence on the results, which may include an alarm, generated by the NRTMA system. Such an interactive diagnostic tool may prove itself particularly valuable during the commissioning phase, and the early stages of operation, when the base of experience is at its lowest.

9 STOCHASTIC MODEL

The THORP Stochastic Model is being improved in order to generate information about the quantities of plutonium associated with transfers and inventory items. Furthermore, the ability to introduce sources of measurement error is required so that the improvements to the prototype NRTMA system can be tested using data which closely resemble those possible from operating plant.

10 CONCLUDING REMARKS

In terms of construction and engineering, THORP is a major process plant project. In terms of statistical data analysis, the THORP NRTMA system is also a major project.

The culmination of development work since the early 1980s was the production of a prototype NRTMA system for THORP. This prototype is now being extended to produce an effective materials accountancy and control system for operational use.

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NEAR REAL TIME MATERIALS ACCOUNTANCY
RESOLUTION OF ANOMALIES

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ABSTRACT

When a single stream of materials accountancy data is tested, there may come a time when an anomaly is signalled. This means that, allowing for the false alarm probability which has been specified when setting up the testing procedure, the data stream appears to be inconsistent with there being no loss or gain of material. Test procedures are insufficient on their own; an anomaly resolution procedure is essential.

The purpose of the anomaly resolution procedure is to provide evidence of the size and duration of the irregularity which caused the alarm.

1 INTRODUCTION

An earlier paper[1] described how a Near Real Time Materials Accountancy (NRTMA) system could be broken down into a number of modules, each with a clearly defined function. Some modules are versatile and, therefore, capable of being installed as part of any NRTMA system. This paper concentrates on one such module - for anomaly resolution.

When a single stream of materials accountancy data is processed and the Joint Test applied to the resulting series of *SITMUF* values, there may come a time when one of the test statistics exceeds the alarm level.

For materials control purposes, it is just as important to detect an apparent gain of material as it is to detect an apparent loss. Therefore, two Joint Tests are employed to give a two-sided testing procedure and, altogether, four test statistics are used[3]. This means that, allowing for the false alarm probability which has been specified when setting up the testing procedure, an alarm from the testing procedure is a signal that the data stream appears to be inconsistent with there being no loss <u>or</u> gain of material. The testing procedure does <u>not</u> identify the cause of the alarm. Therefore, it is important to have an anomaly resolution procedure since test procedures are insufficient on their own.

A recent paper[2] in the ESARDA Bulletin showed the sound basis for an anomaly resolution procedure but pointed out that more development was required to produce a comprehensive procedure for operational purposes. Two areas for further work were identified. Firstly, those models which best fit the *observed* data should be ranked. Secondly, standard errors associated with loss, gain or error estimates need to be calculated. This paper addresses these two topics.

2 RESOLVING ANOMALIES BY MATHEMATICAL MODELLING

The basis of resolving anomalies by mathematical modelling has been introduced in an earlier paper[2]. The approach is made possible because of the properties of the *SITMUF* statistic. Firstly, sequences of *expected SITMUF* values can be calculated for various plausible loss, gain or error models. Secondly, the *SITMUF* series is made up of independent items with the same variance so that *observed* and *expected* sequences can be compared by the method of *least squares*.

3 EXPECTED *SITMUF* PATTERNS

For any plant which is suffering no loss or gain of material, the *expected* value of the Inventory Difference, *CUMUF* and *SITMUF* statistics, each period, is zero. Once a loss or gain occurs, the expected values of these statistics are no longer zero. When a loss occurs, the expected value of the Inventory Difference and *CUMUF* will be negative. Generally, for a loss in period i, the *expected SITMUF* value, W_j, in period j, is

$$W_j = 0 \qquad j < i$$

$$W_j < 0 \qquad j \geq i$$

SITMUF patterns depend on various plant operating parameters, and on materials measurement uncertainties, and will therefore be plant dependent. Before *expected SITMUF* patterns can be derived for specific loss, gain or error models, a plant model is necessary.

4 THE PLANT MODEL

Previous work[2-7] has chosen a campaign length of 240 days, divided into 40 balance periods of 6 days. The standard deviation of the throughput measurement error per balance period, T, set at 1 kg gives a standard deviation of the campaign throughput measurement error of 6.325 kg. This, and the standard deviation of the inventory measurement error, *I*, of 2 kg, is consistent with predictions for the THORP materials accountancy and control system[8]. This model will be used throughout the paper. Table I shows, for the specified plant model, the *expected SITMUF* values resulting from a gain of 1 kilogram in any one of the first 5 periods.

Table I: Expected *SITMUF* Values for a Gain of 1 Kilogram

Gains Period	Expected *SITMUF* Value in Period				
	1	2	3	4	5
1	0.44721	0.33218	0.22084	0.13937	0.08610
2	▨	0.41523	0.27605	0.17422	0.10762
3	▨	▨	0.40028	0.25261	0.15605
4	▨	▨	▨	0.39416	0.24349
5	▨	▨	▨	▨	0.39181

5 COMBINATION OF VECTORS

The values (0.44721, 0.33218, 0.22084, 0.13937, 0.08610), in Table I, constitute the *SITMUF* vector for a 1 kilogram gain in period 1. Subsequent rows of values constitute the *SITMUF* vectors for the same size gain in later periods. *SITMUF* vectors can be scaled, and added to one another. Because of these properties it is possible to calculate the *expected SITMUF* vector for any loss, gain or error model from a knowledge of the individual *expected SITMUF* vectors for a gain of 1 kilogram in any particular period. The number of basic scenarios for which the *SITMUF* vectors need to be known is therefore limited to the same number as there are periods in the campaign.

Table II shows how the *SITMUF* vector for a uniform protracted gain over periods 2 to 4 can be derived from the individual *SITMUF* vectors for a gain in each of the periods 2, 3 and 4.

Table II: Derivation of Expected *SITMUF* Vector for a Uniform Protracted Gain over Periods 2 to 4

Gain Period(s)	Gain (kg)	Expected *SITMUF* Value in Period				
		1	2	3	4	5
2	1		0.41523	0.27605	0.17422	0.10762
3	1			0.40028	0.25261	0.15605
4	1				0.39416	0.24349
2 to 4	3		0.41523	0.67633	0.82099	0.50716
2 to 4	1		0.13841	0.22544	0.27366	0.16905

Table III shows how the *SITMUF* vector for an inventory error at the end of period 3 can be derived from the *SITMUF* vectors for a gain in period 3 and a loss in period 4.

Table III: Derivation of Expected *SITMUF* Vector for Inventory Error at the End of Period 3

Gain Period(s)	Gain (kg)	Expected *SITMUF* Value in Period				
		1	2	3	4	5
3	1			0.40028	0.25261	0.15605
4	-1				-0.39416	-0.24349
1 kg Inventory Error				0.40028	-0.14155	-0.08744

6 ESTIMATION OF THE BEST FIT FOR A GIVEN MODEL

Suppose the Joint Test gives a signal in period *p*. It has already been shown[2] that, for anomaly resolution, it is important to consider *SITMUF* values up to the period after the alarm from the Joint Test, period *j*, where $j=p+1$.

Let the *observed SITMUF* sequence be Y_1, Y_2, .., Y_j. Let the *expected SITMUF* sequence corresponding to unit loss, gain or error, for a given model, be W_1, W_2, .., W_j. Let the unknown loss, gain or error be *L* kilograms. Now the *sum of squares* is

$$S_j = \sum_{i=1}^{j} (Y_i - LW_i)^2$$

What is wanted is the best estimate of *L*. In other words, find the value of *L* for which $\frac{dS_j}{dL} = 0$.

$$\frac{dS_j}{dL} = -2\sum_{i=1}^{j} Y_i W_i + 2L\sum_{i=1}^{j} W_i^2$$

When $\frac{dS_j}{dL} = 0$,

$$\hat{L} = \frac{\sum_{i=1}^{j} Y_i W_i}{\sum_{i=1}^{j} W_i^2}$$

Check the 2nd derivative

$$\frac{d^2 S_j}{dL^2} = 2\sum_{i=1}^{j} W_i^2$$

which is always positive, so the value obtained for *L* corresponds to a minimum for S_j.

Likelihoods and Standard Errors

If *x* is an observation from a normal distribution with mean μ and variance σ^2, then

$$f(x) = \frac{1}{\sqrt{2\pi}} e^{-\frac{1}{2}\left(\frac{x-\mu}{\sigma}\right)^2}$$

The probability that *SITMUF* ($\sigma^2 = 1$) takes a value in the interval x to $x + dx$ is given by

$$k\, e^{-\frac{1}{2}(x-\mu)^2} dx$$

In the case of *n observed SITMUF* values, Y_1, Y_2, .., Y_n, and *expected SITMUF* values, $G_1 W_1$, $G_1 W_2$, .., $G_1 W_n$ corresponding to a gain of G_1 kilograms for a given model (1) W_1, W_2,.., W_n, the joint probability of the observations, regarded as a function of the unknown gain, G_1, is called the Likelihood Function of the sample, and is written

$$L(Y|G_1) = f(Y_1|G_1)\, f(Y_2|G_1) \ldots f(Y_n|G_1)$$

$$= \prod_{i=1}^{n} k\, e^{-\frac{1}{2}\left(Y_i - G_1 W_i\right)^2}$$

$$= k^n e^{-\frac{1}{2}\sum_{i=1}^{n}\left(Y_i - G_1 W_i\right)^2}$$

For a second model (2), and a corresponding gain of G_2 kilograms

$$L(Y|G_2) = k^n e^{-\frac{1}{2}\sum_{i=1}^{n}\left(Y_i - G_2 W_i\right)^2}$$

Therefore, the relative likelihood of model (1) compared with model (2) is

$$\frac{k^n e^{-\frac{1}{2}\sum_{i=1}^{n}\left(Y_i - G_1 W_i\right)^2}}{k^n e^{-\frac{1}{2}\sum_{i=1}^{n}\left(Y_i - G_2 W_i\right)^2}} = e\left(\sum_{i=1}^{n}\left(Y_i - G_2 W_i\right)^2 - \sum_{i=1}^{n}\left(Y_i - G_1 W_i\right)^2\right)$$

The variance of $\hat{G}$, where $\hat{G}$ is the maximum likelihood estimate, is

$$\frac{1}{E\left\{-\frac{\partial^2 l}{\partial G^2}\right\}}$$

where $\hat{G}$ is the value of G which maximizes l, and l is log(Likelihood)

$$l = n \log k - \frac{1}{2}\sum_{i=1}^{n}\left(Y_i - G W_i\right)^2$$

$$\frac{\partial l}{\partial G} = \sum_{i=1}^{n} Y_i W_i - G \sum_{i=1}^{n} W_i^2$$

$$\frac{\partial^2 l}{\partial G^2} = -\sum_{i=1}^{n} W_i^2$$

and the standard error of $\hat{G}$ is therefore

$$\frac{1}{\left(\sum_{i=1}^{n} W_i^2\right)^{\frac{1}{2}}}$$

Example

For the plant model described above, and a campaign false alarm probability of 5%, it has already been shown' that a suitable Joint Test for detecting gains has the following parameters:-

H1 = 0, K1 = 3.4758, H2 = 7.8, K2 = 0.24389

The corresponding Joint Test for detecting losses has negative values for the test parameters:-

H1 = 0, K1 = -3.4758, H2 = -7.8, K2 = -0.24389

The Joint Test procedure is made up of two components, each of which is a Page's Test. For these components the two test statistics S1 and S2 are defined by :-

$$S1_0 = 0$$

$$S2_0 = 0$$

$$S1_i = \max(0, S1_{i-1} + Y_i - K1) \qquad i > 0$$

$$S2_i = \max(0, S2_{i-1} + Y_i - K2) \qquad i > 0$$

where $Y_1, Y_2, \ldots, Y_i$ is the series of *SITMUF* values generated from $ID_1, \ldots, ID_i$, the series of Inventory Difference values. The Joint Test is applied in such a way than an alarm is given if $S1_i > H1$ otherwise no alarm is signalled unless $S2_i \geq H2$.

Consider a sequence of observed *SITMUF* values. Table IV shows such a sequence.We wish to determine which loss, gain or error scenarios best fit the observed data.

Table IV: Observed *SITMUF* Values

Period	Observed *SITMUF* Value
1	0.419
2	-0.093
3	0.358
4	-4.735
5	-2.012

The Joint Test alarms in period 4, the SITMUF sequence is analysed for the first 5 periods, since as previously explained it is important to consider *SITMUF* values up to the period after the alarm.

All plausible models are considered and the expected *SITMUF* vector is calculated for each model. An abrupt model is where the loss or gain begins and ends in the same period, a protracted model is where the loss or gain extends over more than one period, and an inventory error model is where an error has occured at an inventory determination. For each model, the *observed* and *expected* sequences are compared by the *least squares* method and the best estimate of the anomaly and the *least squares* value are calculated. The models are then ranked in order of their *least squares* values and the likelihood of each model calculated relative to the most likely one.

Table V shows the results of the analysis performed on the above *SITMUF* sequence. The table shows that the most likely explanation of the alarm is that there has been an abrupt loss of 10.7kg in period 4. The data for this example were simulated using an abrupt loss of 10kg in period 4.

7 CONCLUDING REMARKS

In a recent paper[2], it was shown how a series of observed *SITMUF* values could be compared with the expected *SITMUF* values for a chosen loss, gain or error model, and the best fit of that model found using standard least squares techniques. This paper describes the underlying mathematical principles of a computer application which is used to evaluate an exhaustive range of models for abrupt loss (or gain), protracted loss (or gain) and inventory errors.

For the operator the product of the Anomaly Resolution module is directly useable information about the plant events most likely to have given rise to the alarm from the Joint Test procedure.

Table V: First Ten Loss/Error Scenarios for the Observed SITMUF Values

Model	Periods		Anomaly Estimate	Standard Error	Least Squares	Relative Likelihood
	First	Last				
1	4	4	-10727	2266	4.362	1.000
2	3	4	- 9280	2285	10.280	0.003
3	4	5	-12366	3127	11.136	0.001
4	3	5	- 9419	2540	13.025	<0.001
5	4	5	- 9352	2533	13.150	<0.001
6	2	4	- 7241	2088	14.756	<0.001
7	2	5	- 8061	2357	15.078	<0.001
8	3	3	- 5394	1918	18.871	<0.001
9	1	5	- 5681	2057	19.152	<0.001
10	1	4	- 4786	1803	19.735	<0.001

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FUZZY METHODS FOR SYSTEM PERFORMANCE

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Abstract

We propose a new method, based on the notion of fuzzy sets, to evaluate system performance. This method is applicable to an arbitrary system composed of the components that describe the system hierarchically. As an illustration, we introduce a new figure of merit, consistent with the recommendations of the Advisory Group Meeting on Containment and Surveillance. The fuzzy attributes, such as no-diversion, assurance, and reliability replace the subjective characterization parameters of individual components. Our methodology should be advantageous both in safeguards and in containment and surveillance problems.

1. Introduction

At the 6th Advisory Group Meeting on Containment and Surveillance held in Vienna in 1988, a framework was proposed to enable the characterization—with respect to assurance—of the performance of containment and surveillance (C/S) devices. Although one cannot quantify the performance of C/S in terms comparable to those of materials accountancy, the overall performance of a C/S system in terms of a factor of merit has been attempted /1/. This trend to express system performance in a formalized way is not limited to C/S. We mention but a few examples. Risk assessment to characterize the potential for liability costs associated with a facility for disposal of low-level radioactive waste was studied by Karam et al. /2/. The radiological sabotage consequence index was introduced by Altman and Hockert /3/. More recently, this type of qualitative analysis has been applied to risk of disclosure /4/ and vulnerability assessment to determine risks resulting from possible adversary actions /5/.

The purpose of this paper is to characterize the system performance, and also the system risk, by using an approach based on the notion of fuzzy sets and linguistic variables. The idea is to represent a fact as an evaluation and to represent a production rule as a transformation of evaluations. A fuzzy set can be regarded as function mapping the universe of discourse to a continuum of possible choices; such a function can be used to describe imprecise terms. Fuzzy sets, seen as states of fuzzy systems, thus become an algebra of approximate reasoning. The elements of this algebra are linguistic variables whose values are words or sentences in a natural or artificial language. For example, the values of the linguistic variable *age* may be young, middle-aged, old.

We summarize in Sec. 2 the classic approach to system performance, quantified in terms of a figure of merit. In Sec. 3, we provide the necessary definitions of the fuzzy sets theory and touch briefly on the applications of fuzzy sets to risk analysis. Section 4 is devoted to a fuzzy figure of merit. This will be based on linguistic rather than numeric measure.

2. Performance Characterization of C/S Systems

As indicated in Ref. 1, the overall characterization of the C/S system can be expressed in terms of a figure of merit that accounts for the system performance with respect to three major categories. These categories encompass the requirements of (1) analysis of diversion routes and their effectiveness; (2) system reliability, derived from mean time before failure (MTBF), false alarm (FA) rate, and detection probability (DP); and (3) system assurance (tamper resistance).

The proposed figure of merit M is defined as

$$M = ND \times R \times A \ , \tag{1}$$

where ND refers to diversion possibilities, R denotes the reliability, and A denotes assurance.

The diversion parameter ND and system assurance A are derived subjectively; they characterize diversion through subjectively assigned numbers (Tables I and II, respectively).

TABLE I. Characterization of No-Diversion

Diversion Analysis	Grading
No analysis or diversion route identified	0.0
Diversion route identified— low risk to operator	0.2
Diversion route identified— high risk to operator	0.5
No diversion route identified	0.8
No diversion route identified and effectiveness demonstrated	1.0

System vulnerability is treated in the same way (see Table III) but could be quantified if MTBF, detection probability, and false alarm rate have been determined from engineering tests on operation data.

In the following, we attempt to replace the numeric grading in Tables I-III by a linguistic grading reflecting our state of ignorance about numeric data.

Table II. Characterization of System Assurance

Assurance Analysis	Grading
Tamper route demonstrated no risk to divertor	0.0
Tamper route demonstrated—some risk to divertor	0.2
Tamper route believed feasible—but could not be demonstrated (w/o black hatting by a group of specialists)	0.5
Tamper route demonstrated—high risk to divertor	0.7
Tamper rout believed feasible—but could not be demonstrated with black hatting OR No tamper route conceivable—but w/o attempted black hatting	0.8
No tamper route conceivable by a group of specialists	1.0

TABLE III. Characterization of System Reliability

Reliability Analysis	Grading
MTBF or false alarm or detection probability—not acceptable	0.0
MTBF + false alarm + detection probability—lowest acceptable level low risk to operator	0.3
Medium MTBF + FA + DP	0.7
High MTBF + FA + DP	1.0

3. Imprecise Estimates of Performance

Fuzzy set theory, originally developed by Zadeh, is today a subject of review articles /6,7/ and monographs /8,9/. Fuzzy set theory was developed to generalize classical set theory in such a manner as to allow the possibility of partial membership in a set. In everyday life, one can find many examples of sets for which membership is not well defined. Some examples are the set of all tall men, the set of very large trees, or the set of all protective mechanisms that provide security against a certain threat.

In this section, we illustrate the notion of a membership function and define the operations of addition and multiplication of fuzzy sets. This will be followed by a description of the application of these concepts to risk analysis.

Fuzzy Sets

Intuitively, a fuzzy set is a class that admits the possibility of partial membership in it. Let $X = \{x\}$ denote a reference set (universe of discourse). Then a fuzzy set A in X is a set of ordered pairs

$$A = \{x, \chi_A(x)\} \quad , \; x \in X \; , \tag{2}$$

where $\chi_A(x)$ is termed the grade of membership of x in A. We assume for simplicity that $\chi_A(x)$ is a number in the interval [0,1], with grades 1 and 0 representing, respectively, full membership and full nonmembership in a fuzzy set. As an example, consider a linguistic variable *confidence factor* with values low, medium, and high, regarded as fuzzy sets. With the universe of discourse specified, arbitrarily, as the set of integers 1, ..., 9, the fuzzy sets low, medium, and high are represented graphically in Fig. 1.

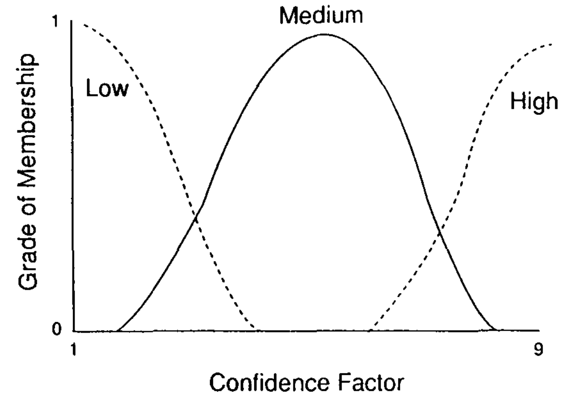

Fig. 1. Membership function for primary terms: low, medium, high.

A finite fuzzy set A having n elements in X is expressed as

$$A = \sum_{j=1}^{n} \chi_A(x_j)/x_j \; . \tag{3}$$

With this notation, the basic algebraic operations are introduced through the so-called extension principle:

$$A + B = \sum_{i,j}^{n} \min\left[\chi_A(x_i), \chi_B(x_j)\right] / (x_i + x_j) \; , \tag{4}$$

$$A \times B = \sum_{i,j}^{n} \min\left[\chi_A(x_i), \chi_B(x_j)\right] / (x_i \times x_j) \; , \tag{5}$$

$$A / B = \sum_{i,j}^{n} \min\left[\chi_A(x_i), \chi_B(x_j)\right] / (x_i / x_j) \; . \tag{6}$$

The operation of division requires that A / B should be reduced to a set of integers. Using the terminology borrowed from object-oriented programming, we have overloaded the operators +, ×, and /, thus extending their domain of definition to fuzzy sets.

The entities on which the basic operators act constitute either the primaries or hedged primaries of a natural language of linguistic variables. The primaries are *low, medium, high,* as well as the fuzzy numbers between 1 and 9. An example of hedged primary is *fairly low* or *pretty high*. The complete syntax of the natural language we use follows closely the monograph of Schmucker /10/. This syntax includes more involved constructs, such as relational phrases and composite relations.

Fuzzy Risk Analysis

Our performance evaluation method can be applied immediately to risk assessment. This method, used earlier in the context of risk analysis for computer security /11/ applies to a system composed of units that can describe the system hierarchically. One merely climbs the tree of decomposition, computing the risk of each interior node from the risk values of its descendants, until one arrives at the root node.

At the lowest level, the terminal nodes have three attributes associated with them. They are probability of failure, severity of loss, and confidence factor. The fuzzy product of these attributes, defined as an extension of the product of real numbers, results in a component risk indicator of the node. This is analogous to the figure of merit of Ref. 1. The component risk indicators of the descendants of each parent node are then weighted by the weight factors to produce the parent's node risk indicator. The weight factor, therefore, adds or subtracts weight from the risk indicators as they are merged into higher level indicators. The nonterminal nodes, which rely on their descendants, do not require the three attributes of terminal nodes.

When our algorithm is implemented on a computer, we are in a position to simulate a variety of systems studying their weakest nodes that lead to the highest risk. This is reminiscent of the method of Gayral et al. /12/ to assist a protection system.

4. Fuzzy Performance Characterization

We now construct the model that characterizes the performance of a safeguards system, like we did for the risk assessment model. As an illustration, we consider a simplified model of a facility consisting of two divisions, D1 and D2. Divisions D1 and D2, in turn, are composed of three and two sections, respectively. We refer to sections of D1 as S1, S2 and S3, whereas the sections of D2 are denoted by T1 and T2; this is indicated in Fig. 2.

At the lowest level, the component performance indicator is given by Eq. (1) as a product of *ND, R,* and *A*, as shown in Table IV. To assure the flexibility of our description, we do not provide a translation of Tables I - III in terms of fuzzy sets. Instead, we quantify the attributes of the components in terms of complex phrases compatible with the natural language we use. The overall figure of merit, on the other hand, is quantized at 8 levels; its value is determined through the shortest distance analysis /10/.

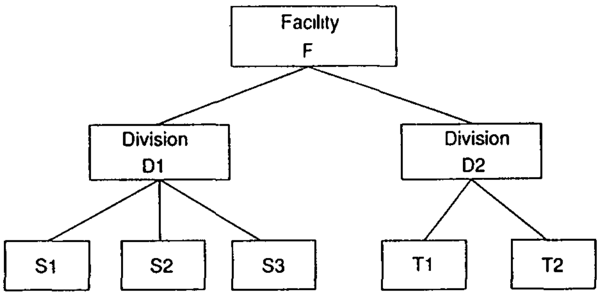

Fig. 2. Simplified scheme of a facility composed of divisions and sections.

TABLE IV. The Computed Section Performance Indicator

Section	Performance Indicator	No Diversion	Assurance	Reliability
S1	Fairly high	Very low	Pretty high	Medium to high

The fuzzy figure of merit of each parent node is obtained by combining the component risk indicator with the weighting factor specifying the proportion estimate of a child node. An example of this procedure is shown in Table V.

TABLE V. The Computed Division Performance Indicator

Division D1	Division Performance Indicator	Proportion	Section Performance Indicator
Section S1	Medium	About three	Fairly high
Section S2		Morl high	Medium to high
Section S3		High	High

Here, the shorthand "Morl" stands for "more or less." To arrive at the performance indicator for D1, we have computed a weighted average of the performance indicators M_j of the individual sections, according to the formula

$$M_{D1} = \frac{\sum_{j=1}^{3} W_j \times M_j}{\sum_{j=1}^{3} W_j} \qquad (7)$$

where the W_j's denote the weights describing the proportions associated with each section. In a similar manner, the facility performance indicator, which quantifies the overall figure of merit, is obtained as a weighted performance indicator of individual divisions. The described procedure can, obviously, be extended to more complicated subdivisions.

The key assumption is that only the terminal nodes of the hierarchy are described in terms of the diversion possibility, reliability, and assurance; the nodes at higher levels merely require the weighting factors.

5. An Example of Computation

The tree structure shown in Fig. 2 will now be supplemented by a specific description of the performance of individual nodes. Table VI details our example.

The computation yields the figure of merit *low* as a measure of performance for the facility. It is easy to see that, although weighted low, division D2 is a weak link in the overall performance. In fact, if the last row of Table VI is changed into ***high, high, one, medium***, the figure of merit changes into ***medium***. This example indicates how the worst performing link of a complex facility can be localized, thus leading to an improved performance.

TABLE VI. Performance of Individual Nodes of Facility F

Node	No Diversion	Assurance	Reliability	Weight
D1				Medium
D2				Low
S1	Very low	Pretty high	Medium to high	About three
S2	Low	Medium	Low	Morl high
S3	Low	Medium	Medium	High
T1	Low	Low	Low	High
T2	Low to medium	Medium	Fairly low	Low

6. Conclusions

We have proposed a new figure of merit, useful in C/S problems and in safeguards, based on linguistic variables. In many cases of practical interest, this may be a more adequate description of performance than the description employing numeric variables. Future research will focus on refining our model and applying it to realistic facilities.

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Optimal Data Verification Procedures

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Abstract

In order to verify the material balance data reported by plant operators, the safeguards authority performs independent measurements on a random sampling basis and compares the resulting data with the reported ones. It is shown that the so-called D-statistic, which originally has been justified with heuristic arguments, and since then has been used in practice for many years, is optimal if the total assumed falsification is small. Furthermore, numerical calculations indicate that for larger total falsification, where a more complicated test procedure would be optimal, the D-test is still useful from a practical point of view. For very large total falsification, the optimal test statistic is complicated; this, however, is not so important since here one is approaching the attribute sampling area.

Introduction

International nuclear material safeguards is by general agreement organized in such a way that the plant operators generate all the data necessary for the establishment of a material balance, that the inspectors verify the operators' data with the help of independent measurements and that - if there are no significant differences between the operators' data and the inspectors' findings - the material balance is established with the help of the operators' data.

In this paper the present state of data verification is discussed. In doing so, only *variables sampling* [1] procedures are considered which take into account measurement errors, and with the help of which the expected differences between the operators' reported data and inspectors' findings are quantitatively evaluated.

In the following we will have in mind primarily the verification of inventory data, first, because it is easier from a methodological point of view, and second, because it represents an especially important part of safeguards: whereas flow measurement data sometimes can be verified by comparing shipper and receiver data, there is nothing which can replace inventory data verification using independent measurements.

Data verification presents a *statistical problem* because of the random sampling procedure and, in case of variable sampling, because of the existence of statistical measurement errors. Furthermore, since at the end of the verification procedure a decision has to be taken whether or not the data of the operator are accepted, data verification in safeguards basically is a *test problem.* (An inspector may also be interested in estimating possible defects [2]; since, however, their use is not clear in international safeguards, estimation is not discussed here.) Finally, contrary to conventional statistical problems like quality control, there is a conflict situation between an operator who may falsify data - otherwise there would be no reason for verifying his data - and the safeguards authority which has to detect an eventual falsification. This means that data verification represents a *game theoretical problem.*

Any game-theoretical model of variable sampling data verification problems requires the definition of payoffs to both the inspector and the operator for detected and undetected illegal behavior as well as for confirmed and not confirmed legal behavior (false alarms). It should be mentioned that at least in case of false alarms the interests of both players are *not* opposite to each other. Therefore, non-zero-sum models have to be used.

It can be shown (see, e.g., [3]) that models of this kind can be decomposed into two sub-models such that the first sub-model deals only with the decision of the operator whether or not to act illegally, and the inspector's choice of the false alarm probability. The second sub-model deals with the operator's choice of an illegal strategy and the inspector's choice of a decision procedure for a given false alarm probability. This second sub-model, however, is a zero-sum game with the probability of detecting an illegal action as payoff to the inspector. This means that, for a given value of the false alarm probability, payoff parameters need not be known, which is very helpful for practical applications. In the following, we consider only the second "statistical" game-theoretical model.

Analytical Results

Let us assume that N data X_i, $i = 1, \ldots, N$, have been reported by the operator, and that $n (\leq N)$ data are verified by the inspector with the help of independent observations Y_i, $i = 1, \ldots, n$, on a random sampling basis. Since the inspector is not interested in the true values of the random variables X_i or Y_i, but only in the deviations between corresponding reported and independently generated data, he will construct his test procedure with the help of the differences of these corresponding data. We formulate this test problem as follows:

Definition 1. The differences Z_i, $i = 1, \ldots, n$, between the operator's reported data X_i and the independent findings Y_i of the inspector are assumed to be independently and identically normally distributed random variables with variances

$$\mathrm{var}(Z_i) = \mathrm{var}(X_i) + \mathrm{var}(Y_i) =: \sigma^2, \qquad i = 1, \ldots, n,$$

and with expected values

$$E(Z_i) = \begin{cases} 0 & \text{under } H_0 \\ \mu_i > 0, \quad i = 1, \ldots, n, & \text{under } H_1, \end{cases}$$

where H_0 is the null hypothesis (no data falsification) and H_1 the alternative hypothesis (data falsification). □

As a result of the game-theoretical analysis sketched before, we are looking for that test procedure which maximizes the probability of detection $1-\beta$, given a fixed value of the false alarm probability. In doing so, we assume that the operator – if at all – will falsify all data by the total amount μ in such a way that the probability of detection is minimized.

First, for maximum sample size $n = N$ an optimal test procedure is given:

Theorem 1. [4] Let the sets of pure strategies Δ_α and Γ_μ of the inspector and of the operator be given by the set Δ_α of all test procedures for the test problem given by Definition 1 with $n = N$ and given false alarm probability α, and

$$\Gamma_\mu := \{ (\mu_1, \ldots, \mu_N) : \textstyle\sum_{i=1}^N \mu_i = \mu > 0, \\ 0 \le \mu_i, \quad i = 1, \ldots, N \},$$

and let the payoff to the inspector be the detection probability $1-\beta$, i.e., the probability of rejecting H_0 if in fact H_1 is true. Consider the two-person zero-sum game $(\Delta_\alpha, \Gamma_\mu, 1-\beta)$, where the values of μ, σ and α are known to both players.

Then the saddlepoint strategy of the operator is given by $(\mu/N, \ldots, \mu/N)$, that of the inspector is a test given by the critical region

$$\{ (Z_1, \ldots, Z_N) : \sum_{i=1}^{N} Z_i > \sigma \cdot \sqrt{N} \cdot U(1-\alpha) \},$$

and the value of the game, i.e., the guaranteed optimal probability of detection $1-\beta^*$ is

$$1 - \beta^* = \Phi\big(\mu/\sigma \cdot \sqrt{N} - U(1-\alpha)\big),$$

where $\Phi(\,.\,)$ is the normal distribution and $U(\,.\,)$ its inverse. □

According to this Theorem, the test statistic is $D := \sum_i Z_i$. This is the well known D-test statistic which was proposed first in 1970 [5] for use in nuclear material safeguards; at that time, it was justified by heuristic arguments.

Second, let us assume that only *one datum* drawn at random out of N reported data (minimum sample size) is verified by the inspector. The following solution to this problem is more complicated than the preceeding one:

Theorem 2. [6] Consider the two-person zero-sum game $(\Delta_\alpha, \Gamma_\mu, 1-\beta)$ as in Theorem 1, however with $n = 1$. Let $\mu = \mu^*(N)$ be the unique solution of $f(\mu) = 0$, where the function $f(\mu)$ is defined by

$$f(\mu) = \Phi\big(U(1-\alpha) - \frac{1}{N} \cdot \frac{\mu}{\sigma}\big) - \\ \big(\Phi(U(1-\alpha) - \frac{\mu}{\sigma}) + (N-1) \cdot (1-\alpha)\big)/N.$$

Then for $\mu < \mu^*(N)$ the saddlepoint strategy of the operator is $(\mu/N, \ldots, \mu/N)$, whereas for $\mu \ge \mu^*(N)$ it is $(\mu, 0, \ldots, 0)$ or $\ldots$ or $(0, \ldots, 0, \mu)$.

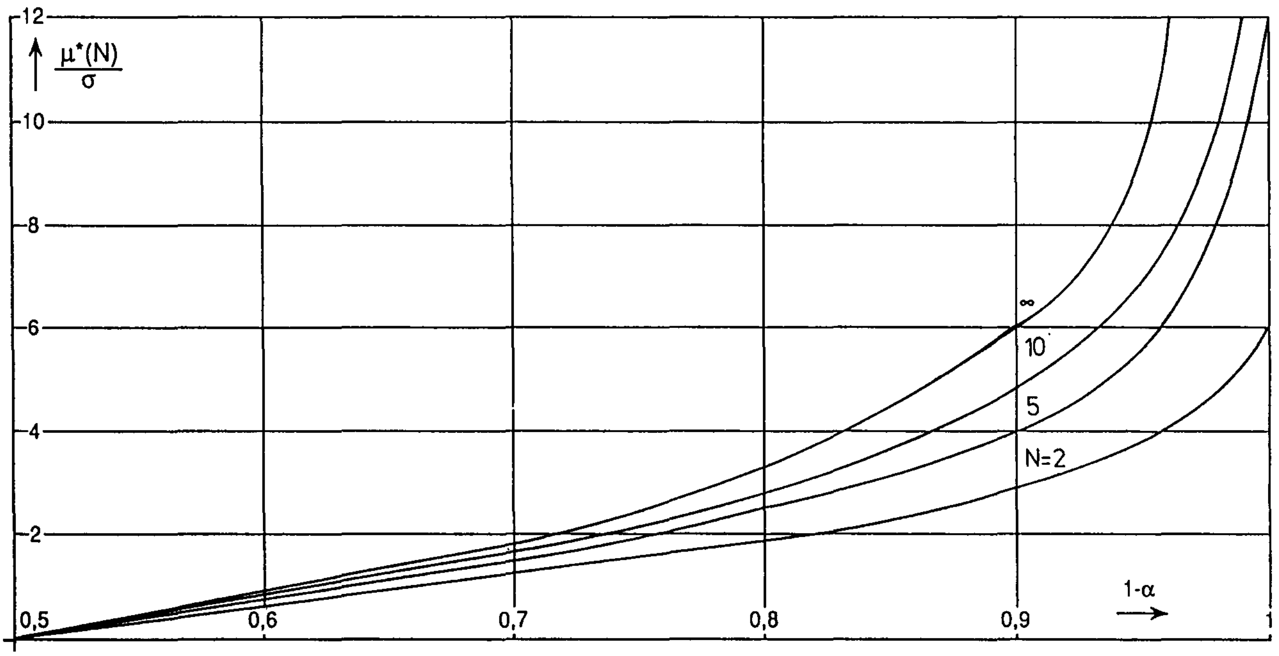

Figure 1: Critical falsification μ*(N) for various values of N and limiting value μ*, as given by Theorem 2, as function of no-false-alarm probability 1-α.

The saddlepoint strategy of the inspector is the test given by the critical region $\{ Z : Z > \sigma \cdot U(1-\alpha) \}$; it is independent of the strategy of the operator.

The value of the game, i.e., the guaranteed optimal probability of detection $1-\beta^*$, is

$$\begin{cases} \Phi(\frac{1}{N} \cdot \frac{\mu}{\sigma} - U(1-\alpha)) & \text{for } \mu \leq \mu^*(N) \\ \Phi(\frac{\mu}{\sigma} - U(1-\alpha))/N + (1-\frac{1}{N}) \cdot \alpha & \text{for } \mu \geq \mu^*(N). \end{cases}$$

The sequence $\{\mu^*(N)\}$ for $N = 1, 2, \ldots$ of critical falsifications is strictly increasing; it starts with $\mu^*(2) = 2 \cdot \sigma \cdot U(1-\alpha)$ and converges to a limit value μ^* which is implicitly determined by the equation

$$\phi(U(1-\alpha)) \cdot \frac{\mu^*}{\sigma} + \Phi(U(1-\alpha) - \frac{\mu^*}{\sigma}) - 1 + \alpha = 0,$$

where $\phi(\,.\,)$ is the normal distribution density. ☐

The critical value $\mu^*(N)$ as well as the limiting value μ^* are graphically represented in Figure 1.

This result recurs in some form or other throughout the problem area. It has an intuitive interpretation: If the total falsification is small, then from a falsification point of view it is best to distribute it on all N data in order to conceal the falsification by the measurement uncertainty. If, on the other hand, the total falsification is large, it can no longer be concealed in this way, so the number of falsified data has to be as small as possible to minimize the probability that the falsified datum is verified and thus discovered.

Third, we consider the general case $1 < n < N$, again without restriction to single falsifications. So far, no general solution to this problem is known. One only knows that for small total falsification μ again the D-statistic is optimal:

Theorem 3. [4] If the total falsification μ satisfies the condition

$$\mu \leq \frac{\sqrt{n}}{\binom{N-1}{n-1}} \cdot \mu^*(\binom{N}{n}), \quad 1 \leq n < N,$$

where $\mu^*(\,.\,)$ is given by Theorem 2, then the D-test as well as the equal distribution of the falsification are saddlepoint strategies of the game $(\Delta_\alpha, \Gamma_\mu, 1-\beta)$. ☐

There are further analytical results for the general problem considered here, e.g., for the case where there exists an upper limit μ_r for the falsification of one single stratum [7]. There is, however, no general theorem which describes the saddlepoint strategy of both the inspector and the operator for *all* values μ of the total falsification.

Numerical Calculations

It has been shown numerically that for arbitrary values of N and $1 < n < N$ and for large total falsification μ, the D-statistic is no longer the best test statistic for the inspector. Nevertheless, for the moment we continue to consider the test based on the D-statistic and ask which falsification strategy minimizes its detection probability for given value of μ. Theorem 3 tells us that for small values of μ the equal distribution $(\mu/N, \ldots, \mu/N)$ of the falsification is optimal, i.e., saddlepoint strategy for the inspector.

So far, it cannot be proven analytically, but there is strong numerical support [8], that in addition to the equal distribution $(\mu/N, \ldots, \mu/N)$ only the similarly defined

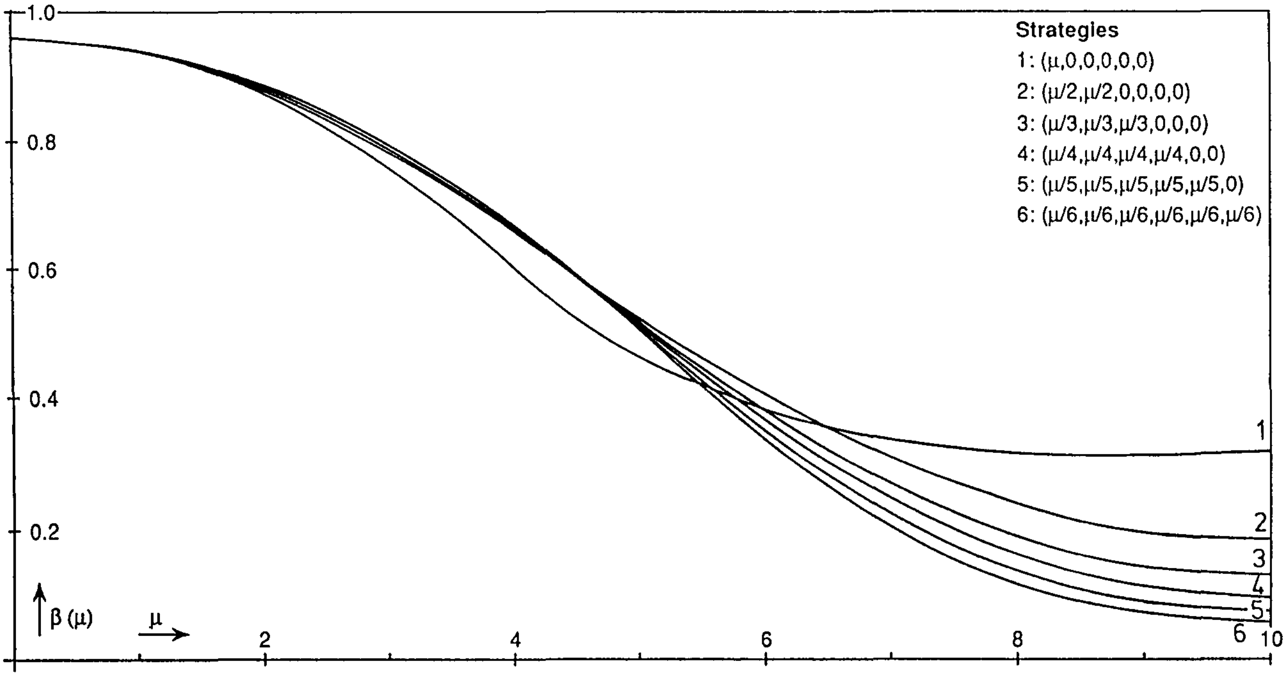

Figure 2 : Probability of non detection $\beta(\mu)$ for the D - test as function of total falsification μ for $(N,n) = (6,4)$ and various falsification strategies

strategies $(\mu/(N-1), \dots, \mu/(N-1), 0)$, or, analogously, $(\mu/(N-2), \dots, \mu/(N-2), 0, 0)$, ..., $(\mu, 0, \dots, 0)$, minimize the detection probability of the D-test so that the optimization procedure can be limited to these marginal strategies.

For these marginal strategies, with r falsified out of N data, the probability of detection is given by $1-\beta(\mu)$, where

$$\beta(\mu) = \sum_l \Phi(U(1-\alpha) - \frac{l}{\sqrt{n}\cdot r} \cdot \frac{\mu}{\sigma}) \cdot \frac{\binom{r}{l}\binom{N-r}{n-l}}{\binom{N}{n}}$$

with summation for $\max(0, n+r-N) \leq l \leq \min(n, r)$. Special cases are for $r = N$:

$$\beta(\mu) = \Phi(U(1-\alpha) - \frac{\sqrt{n}}{N} \cdot \frac{\mu}{\sigma})$$

and for $r = 1$:

$$\beta(\mu) = \Phi(U(1-\alpha) - \frac{1}{\sqrt{n}} \cdot \frac{\mu}{\sigma}) \cdot \frac{n}{N} + (1-\alpha) \cdot (1 - \frac{n}{N}).$$

For $n = 1$, these expressions are obviously the same as those given by Theorem 2; for $r = n = N$, we get the expression given by Theorem 1.

An overview on all possible optimal falsification strategies for $1 \leq N \leq 18$ and $n \leq N$ is given in Table 1, which shows that there are only four different cases:

n \ N	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18
1	a	a	a	a	a	a	a	a	a	a	a	a	a	a	a	a	a
2	d	b	a	a	a	a	a	a	a	a	a	a	a	a	a	a	a
3		d	b	c	a	a	a	a	a	a	a	a	a	a	a	a	a
4			d	b	b	c	a	a	a	a	a	a	a	a	a	a	a
5				d	b	b	b	c	a	a	a	a	a	a	a	a	a
6					d	b	b	b	c	c	a	a	a	a	a	a	a
7						d	b	b	b	b	c	c	a	a	a	a	a
8							d	b	b	b	b	b	c	c	a	a	a
9								d	b	b	b	b	b	b	c	c	a
10									d	b	b	b	b	b	b	b	c
11										d	b	b	b	b	b	b	b
12											d	b	b	b	b	b	b
13												d	b	b	b	b	b
14													d	b	b	b	b
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Table 1: Optimal falsification strategies for the D - test. Further explanation see text.

(a) The only optimal strategies are the equal distribution for small μ and the one point distribution for large μ.

(b) All marginal falsification strategies mentioned above are optimal within well defined ranges of μ, starting with the equal distribution for small μ and ending with the one point distribution for large μ.

(c) Except for $(\mu/3, \mu/3, \mu/3, 0, \dots, 0)$, all marginal falsification strategies are optimal.

(d) Only the equal distribution is optimal as shown by Theorem 1.

It should be kept in mind, however, that the optimal falsification strategies in cases (b) and (c) are *not* saddlepoint strategies: The best test against these strategies is *not* the D-test, but a much more complicated test given by the Neyman-Pearson Lemma (see, e.g., [9]).

Let us consider now explicitly an example of case (b), where all marginal falsification strategies are optimal within well defined ranges of μ. Figure 2 gives the probability of non-detection as a function of μ for $N = 6$, $n = 4$, $\sigma = 1$ and $\alpha = .05$ which illustrates two very interesting aspects: First, in that region of μ where the one-point falsification (1) is not optimal, all probabilities of non-detection are very close to that of the equal distribution for given value of μ, except for that of the one point distribution. Second, in the optimality region of the one point distribution, the probability of non-detection approaches very quickly its asymptotic value which is given by $(1-\alpha)\cdot(1-n/N)$ for arbitrary values of N and n.

These results are very important for practical applications. Quite generally, one may conclude that it is sufficient to consider only the equal and the one point distribution of the falsification. This has been proven to be correct for $n = 1$, and shown numerically for $n \leq N/2$. For all other cases, the probabilities of detection of all other marginal falsification strategies which are optimal for some ranges of the total falsification μ, are so close that the differences are practically irrelevant.

This means that one has just to look for that value $\mu^*(N;n)$ of μ, where the probabilities of detection for the two extreme marginal falsification strategies intersect: $\mu^*(N;n)$ therefore is given as the unique solution of $g(\mu) = 0$, where $g(\mu)$ is given by

$$g(\mu) = \Phi(U(1-\alpha) - \frac{\sqrt{n}}{N} \cdot \frac{\mu}{\sigma}) - \Phi(U(1-\alpha) - \frac{1}{\sqrt{n}} \cdot \frac{\mu}{\sigma}) \cdot \frac{n}{N} - (1-\alpha) \cdot (1 - \frac{n}{N});$$

for $N \to \infty$ the sequence $\{\mu^*(N;n)\}$ converges for fixed n to a limiting value $\mu^*(n)$ which is implicitly determined by the equation

$$\phi(U(1-\alpha)) \cdot \frac{\mu^*(n)}{\sqrt{n}\cdot\sigma} + \Phi(U(1-\alpha) - \frac{\mu^*(n)}{\sqrt{n}\cdot\sigma}) - 1 + \alpha = 0.$$

If we compare this with the limiting critical falsification given by Theorem 2, we observe that we just have to replace μ^* by $\mu^*(n)/\sqrt{n}$.

As a result for given values of N and n, we have to take for $\mu < \mu^*(n)$ the equal distribution, and for $\mu > \mu^*(n)$ the one-point distribution; for the latter case the probability of no detection is practically equal to $(1-\alpha)\cdot(1-n/N)$.

Concluding Remarks

Let us conclude our deliberations with some remarks on several strata. Stratified sampling means considering data in different classes or "strata". This is appropriate when there are different types of measured entities, like volume, weight, etc., or to achieve a greater uniformity if the measurements are of widely different magnitudes, for example. In case of several strata of data to be verified, there is a new element, namely how the inspector shall allocate his inspection resources (like manhours or money) to the different strata. Furthermore, how shall he do this if the verification of one datum in one class takes more effort than in a different class? There is no general solution to these problems in view of the difficult situation even for one stratum. Therefore, special cases and approximations are usually considered [3]. Both are oriented at the existing solutions for one stratum, especially at those for the case where there exists an upper limit μ_r for the falsification of one single stratum.

One important special case, also from the practical point of view, is that if the operator acts illegally, he falsifies all data of one stratum by the same particular amount which is typical for that stratum. In view of Theorem 2, one expects this assumption to be reasonable for small total falsification μ. In this case, the set of illegal strategies of the operator is the set of the single falsifications for all classes. The inspector's set of strategies is the set of possible decision procedures for given false alarm probability and the set of possible modes of allocating the inspection effort to the different strata.

Considering the strata sample sizes of the inspector as continuous variables, this problem can be solved in a satisfying way: The saddlepoint test statistic is the weighted sum of D-statistics for the single strata, and the optimal allocation of the inspection effort is determined by the well known Neyman-Tschuprow formula (see, e.g. [10]).

Another important special case is given by the assumption that the operator falsifies only parts of the data of the strata by strata typical amounts. This may be reasonable for larger total falsification. This case cannot be treated analytically with the same rigor as that described before. If one assumes, however, the D-statistic to be useful also for this case, then one arrives again at solutions which can be applied in practical situations. In particular, one obtains again the Neyman-Tschuprow sampling procedure. This is very satisfying since the inspector thus gets a sampling procedure that does not depend on the scheme by which the operator will falsify data, if at all.

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THE COMPUTER PROGRAM MEMO - A TOOL TO ESTABLISH DETAILED MEASUREMENT MODELS FOR NRTA TEST PROCEDURES

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Abstract

According to the Treaty of Non-Proliferation of Nuclear Weapons (NPT), the International Atomic Energy Agency (IAEA) executes frequent inspections in nuclear facilities. In this connection, a computer program called PROSA was developed at the Karlsruhe Nuclear Research Center. PROSA evaluates nuclear material balances using sequential test procedures based on the measurement model of the facility. In order to enable the determination of the measurement model, a computer program MEMO has been developed, in addition. In a first step, MEMO creates the measurement system of the facility which includes the facility design and the measurement uncertainties of each of the material-balance components. On the basis of real detailed measurement data, MEMO then allows to compute the detailed measurement model related to the balance periods considered.

1. Introduction

According to the Treaty of Non-Proliferation of Nuclear Weapons (NPT), the International Atomic Energy Agency (IAEA) executes frequent inspections in nuclear facilities. For inspection planning and evaluations, a set of technical criteria has been defined by the IAEA for internal use.

One important goal of international safeguards is to detect a possible loss of material timely - in particular as far as bulk handling facilities of industrial scale are considered. That means, about monthly inspections are required in order to make sure that no diversion of nuclear material has occurred. It is obvious that this cannot be performed in the 'classical' way of material balancing which means process stop, clean-out and verification of all nuclear materials. Advanced methods are required which enable the IAEA to apply acceptable safeguards measures whilst plant operation is continued. Investigations related to this subject have been performed in several countries. These activities are usually associated with the term Near-Real Time-Accountancy (NRTA).

The Karlsruhe Nuclear Research Center started research and development activities on NRTA many years ago. The basic idea was to establish short-time material-balances from those measurement data on process inventories which are primarily collected for process-control purposes. Then, sequential statistical tests are applied to the short-time material-balances in order to decide whether or not the observed differences between book and measured inventories can be assigned to measurement uncertainties.

In this context, a computer program PROSA /1/ (Program for Statistical Analysis of NRTA data) was developed as a tool to apply truncated sequential test procedures to a given sequence of material-balance results.

The evaluation by statistical tests is based on the measurement model of the facility. Sequential evaluation requires sequential measurement modelling which leads to a variance/covariance matrix of the material-balance sequence. This variance/covariance matrix is the condensed form of the sequential measurement model.

2. The Material-Balance Problem

Material accountancy is an important tool for international nuclear safeguards. The aim is to detect a possible loss of material timely and with high probability. The 'classical' method of material balance performing one or two balances per year is not able to meet the goal of timeliness in industrial plants with high throughput. Therefore, advanced methods have to be applied which evaluate the generated sequential material balances during plant operation. These methods are summarized under the term Near-Real Time-Accountancy (NRTA). NRTA means that the whole reference time is subdivided into several periods and the material balance is closed after each period. So one gets a sequential series of material-balance results MUF(1), MUF(2),...,MUF(N). The terms MUF and N denote Material Unaccounted For and number of periods, respectively. Each single MUF value can be determined according to the following equation:

$$MUF(i) = I(i-1) + T(i) - I(i).$$

I(i) is the measured ending inventory of period i, I(i-1) is the measured ending inventory of period i-1, and T(i) is the measured net transfer within period i. Due to measurement uncertainties, the MUF values in general do not amount to zero, even in the case of no loss of material. That means, at the end of each balance period i, a decision has to be taken whether or not the deviations of the sequence MUF(1),...,MUF(i) from the expectation values zero can be explained by measurement uncertainties. To take this decision in an objective manner, sequential statistical test procedures are required. The evaluation of the series of MUF values with such tests is based on the detailed measurement model of the facility considered.

3. Determination of a Detailed Measurement Model

The determination of the detailed measurement model for real sequential process data is an essential step in applying NRTA-test procedures. One can assume that in most cases all material-balance terms can be calculated by the mathematical product of two single determinations, for example, volume measurement and concentration measurement. For computing the measurement model, each process component is considered separately using its individual random and systematic measurement uncertainties. With the knowledge of the variances of each inventory and transfer measurement, and with flow-sheet information supplied by the operator, all variance and covariance calculations can be performed.

3.1 Determination of the variance of the inventory of one component

The amount of plutonium (Pu) of each single process component of the facility is - without limitation of general validity - the mathematical product of the volume measurement (VOL) and of the volumetric Pu-concentration measurement (CVP):

$$(1) \quad Pu = VOL * CVP.$$

Both measurements are assumed to be random variables with random and systematic components. The standard deviation of the two types of errors are assumed to be known from operators quality control programs. The mean values are assumed to be zero for both errors, so that each measurement is an unbiased estimate of the true value.

Using truncated Taylor series expansion in the mean values E(VOL) and E(CVP) one gets:

$$\begin{aligned}(2) \quad VOL * CVP &= E(VOL) * E(CVP)\\ &+ (CVP - E(CVP)) * E(VOL)\\ &+ (VOL - E(VOL)) * E(CVP)\end{aligned}$$

which results in the reasonable equation:

$$(3) \quad E(Pu) = E(VOL) * E(CVP).$$

With regard to the variances, the following expression holds:

$$\begin{aligned}(4) \quad var(Pu) &= (E(VOL))^2 * var(CVP)\\ &+ (E(CVP))^2 * var(VOL).\end{aligned}$$

On the right hand of Formula (4), the terms E(VOL) and E(CVP) appear which represent the true values of volume and concentration. These values are even unknown, however, there is the possibility of replacing them by the measurement values, because each measurement is an unbiased estimate of the true value. Doing this, detailed knowledge of all measurement data which means the volume measurement and the concentration measurement of each process component is necessary.

With this procedure, the variance of the amount of plutonium of each process component can be calculated. The variance of the inventory of each component of the facility is split into a random component and into a systematic component. Both components are of random origin. They differ only in the way of error propagation. In this case of a model reprocessing facility, systematic errors are assumed to be constant for the whole model campaign. Furthermore, in order to consider the influence of measurement errors in a reasonable way, each uncertainty component - random and systematic as well - is split into an additive part and into a multiplicative part. So one gets at least four different kinds of measurement uncertainties:

SAD (systematic, additive),
SMU (systematic, multiplicative),
RAD (random, additive), and
RMU (random, multiplicative).

3.2 Determination of the dispersion matrix

In order to determine the measurement model, each process component is considered separately using its individual random and systematic uncertainties of the volume and of the concentration measurement as well. Within one balance period, all measurements of inventories are in general independent to each other from the statistical point of view. However, with respect to the influence of systematic measurement uncertainties, there are covariances between inventory measurements from balance period to balance period. Such covariances are also exist between an input transfer and inventory of the current balance period, and between output transfers (product or waste) and inventories of the balance period next to the last one.

All these variances and covariances have to be calculated and considered by the computer program in order to determine the so-called dispersion matrix (variance/covariance matrix) of the MUF series /2/. This matrix is the condensed form of the sequential measurement model and provides all necessary information to perform statistical test procedures, for example, using the computer program PROSA. This matrix represents the special measurement model calculated from special series of measurement data by the use of the only measurement system of the facility which is valid in general.

4. The Computer Program MEMO

Up to now, those sequential measurement models have been calculated with a computer program running on a host installation. In order to make this procedure userfriendly - as done earlier with the program PROSA - , now a first attempt was made to implement also the program MEMO (Program to Establish a Measurement Model for NRTA Procedures) onto a portable Personal Computer system.

4.1 General Structure of MEMO

In a first step, MEMO has to create the **measurement system** of the facility which includes the **facility design** describing each of the material-balance components and the **measurement uncertainties** associated with each of the appropriate measurements.

This measurement system of the facility has to be created only once and remains unchanged as long as there is neither a change in the facility design, nor in the measurement uncertainties of the measurement methods applied for concentration determinations, nor in the measurement uncertainties of the measurement devices used for volume determinations.

Using these general features for the creation of the measurement system of the considered facility, in a second step MEMO is able to compute the special sequential measurement model related to a chosen number of material-balance periods. Thereby, two additional data sets are necessary, namely the **detailed measurement data of the campaign** and a **run-time file** as well.

The **campaign data** contain all measurements of volume determination and of concentration determination. They must be stricly correlated to the facility design. In the facility design, all balance components - inventory batches and transfer batches as well - are defined and the appropriate measurement methods are indicated. The batches are marked by a batch-code number and by a description name. The description name enables the computer program to detect the kind of batch treated and, therefore, via the design data to connect each batch with the appropriate measurement-uncertainty data.

The **run-time file** controls the starting point of balancing and the number of balance periods to be computed. Furthermore, setting a printout option, one can choose the volume of recorded output data which will be written on a result file. There is the possibility to record only the total balance results and the contributions to the dispersion matrix of all computed balance periods. In addition, these data can also be complemented either by recording the measurement system (design data and measurement-uncertainty data) or by recording of all the batch data which contribute effectively to the material balance and to the dispersion matrix as well.

However, the most important output data of MEMO are the two result files called **MUF** and **COVA**. These result files are generated in order to be used as input data for the performance of the statistical test procedures implemented in the program PROSA.

4.2 Description of the Model Facility Implemented in MEMO

In order to demonstrate the functioning of MEMO and its linkage to the already existing progam PROSA, a model facility was created. The design of this model facility is based on **ten inventory components** and **three transfer components**, resulting in a total of thirteen batches which have to be considered within each balance period. The ten inventory components are distributed among **six facility areas** with up to two components each.

In total, there are eighteen measurement points in order to compute the batch amounts of plutonium and their contributions to the dispersion matrix. According to the ten inventory batches, there are also **ten volume-measurement points**. Together with further **eight concentration-measurement points**, they represent the complete measurement-uncertainty data of the facility.

4.3 Current Limitations and Future Aspects of MEMO

For reasons of run time and of necessary core memory as well, this first version of MEMO is limited in the following way:

- the maximum number of facility areas is limited to six areas,
- the maximum number of groups within each area is also limited to six groups each,
- the maximum number of components within each group is also limited to six components each,
- and the number of balance periods which can be computed by MEMO is limited to a maximum value of 20 periods.

In the future, it is intended to extend these limitations to values of more than ten facility areas, of more than ten groups per area, of more than ten components per group, and of fifty balance periods which can be treated, as well.

Furthermore, a menu-guided user shell will be implemented in MEMO which shall enable the user to establish its individual measurement system (facility design and measurement uncertainties) related to a facility of its own.

Due to the fact that the measurement data of the considered campaign must strictly correlate with the facility-design data, the authors intend to give any support in order to transpose such measurement data according to the facility design created.

5. Concluding Remarks

With the PC-version of MEMO, an important step is solved for applying sequential statistical test procedures implemented in PROSA. The combined program packages of PROSA and MEMO might be very suitable for routine field use not only with respect to reprocessing facilities but probably also for applications in other fields.

6. References

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/2/ U. Bicking, W. Golly, R. Seifert, "Establishing of a Detailed Measurement Model for NRTA Procedures", 11th Annual ESARDA Symposium on Safeguards and Nuclear Material Management, Luxembourg, May 30 to June 2, 1989, p. 315 - 318.

Acknowledgement

The authors wish to thank Mr. G. Nägele who was buisy at KfK with these problems a few years ago. The first draft version of this computer program is mainly based on his reflections of that time.

NEAR REAL TIME MATERIALS ACCOUNTANCY SYSTEM FOR THORP

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ABSTRACT

This paper describes the prototype Near Real Time Materials Accountancy (NRTMA) system which forms the basis of the NRTMA system being developed for inclusion in the Thermal Oxide Reprocessing Plant (THORP) Chemical Plant Information Computer.

The NRTMA system incorporates the joint Page's test on *SITMUF* (the Joint Test) which has been developed in BNFL to meet the requirements for a robust testing procedure capable of both responding effectively to abrupt losses/gains, and also controlling protracted materials loss.

The NRTMA system is presented as a menu-driven application. Emphasis is placed on minimizing and simplifying operator input whilst maximizing the return to him of readily useable information.

1 INTRODUCTION

This paper describes the prototype Near Real Time Materials Accountancy (NRTMA) system which forms the basis of the NRTMA system being developed[1] for inclusion in the THORP Chemical Plant Information Computer.

2 THE PLANT MODEL

The prototype uses data from a simulator which models the characteristics BNFL expects of THORP[2]. Previous work[3-6] has chosen a campaign length of 240 days, divided into 40 balance periods of 6 days. The standard deviation of the throughput measurement error per balance period, T, set at 1 kg gives a standard deviation of the campaign throughput measurement error of 6.325 kg. This, and the standard deviation of the inventory measurement error, I, of 2 kg, is consistent with predictions for the THORP materials accountancy and control system. The above values for I and T are used as basic parameters in the simulator. The simulator allows abrupt and protracted losses/gains, and inventory errors, to be incorporated into the data stream.

3 THE PROTOTYPE SYSTEM

The prototype NRTMA system is the culmination of development work in BNFL since the early 1980s. The system is presented as a menu-driven application running on an IBM PC. The rest of this section of the paper is a description of the functions included in the system.

The Copyright Screen

The Copyright Screen, Figure 1, the first screen seen by the user, shows, amongst other information, the date and version of the application, and the date when the prototype NRTMA system was first created. The user presses <SPACE> to move on to the Joint Test Module.

The Joint Test Module

The NRTMA system incorporates the joint Page's test on *SITMUF* (the Joint Test) which has been developed in BNFL to meet the requirements for a robust testing procedure capable of both responding effectively to abrupt losses/gains, and also controlling protracted materials loss. In this prototype system, the Joint Tests are generated off-line and read in from a file. The Joint Test Module Screen, Figure 2, shows the parameters which will be used in the two Joint Tests. The user presses <SPACE> to move on to the Main Menu.

The Main Menu

The Main Menu, Figure 3, displays the current period and measurement system. The latter is discussed more fully later as part of the Utilities Module. An option is selected by pressing a single key.

Get Data for Next Period and Apply Joint Test

This module is used to read in the receipts, shipments and closing inventory for the next balance period. In addition, the data is processed in order to calculate the corresponding value for *SITMUF*, and the Joint Tests are applied.

Standard Errors of Throughput and Inventory Module

This module, Figure 4, displays, for each balance period, the standard errors associated with receipts, shipments, wastes, overall throughput, and inventory.

Transactions and Balances Module

This module, Figure 5, displays, for each balance period, the opening inventory, receipts, shipments, waste, closing inventory, inventory difference and *CUMUF*. Note, from the displayed data, how the opening inventory equals the closing inventory of the previous period, and that negative values for the inventory difference and *CUMUF* statistics correspond to an apparent loss of material. Graphical display of the Inventory Difference and CUMUF statistics, and of the inventory is also provided.

Sequential Analysis Statistics Module

This module, Figure 6, displays, for each balance period, the inventory difference and *SITMUF* statistics, together with the four statistics associated with the application of the two-sided Joint Test[7]. Graphical display of the test statistics and alarm levels is also provided.

Anomaly Resolution Module

This module, Figure 7, displays the results of the anomaly resolution function. The mathematical aspects of anomaly resolution are described in other papers[7 & 8]. The anomaly resolution procedure considers, one at a time, the following plausible models:-

- an abrupt loss/gain at any period;
- a protracted loss/gain beginning and ending at any periods, and
- an inventory error at any inventory determination.

For each model, the following information is provided:-

- the first and last periods which define the loss, gain or error model;
- the lower and upper 95% confidence limits of the size of the anomaly;
- the likelihood of the model relative to that of the most likely one, and
- a comment which is "Protracted Loss (or Gain)", "Abrupt Loss (or Gain)" or "Inventory Error".

Special Procedures Module

This module, Figure 8, is password protected and allows the experienced user to customize the way in which the anomaly resolution function is implemented.

Utilities Module

This module allows the user to switch between the primary (main) and secondary (back-up) measurement systems[2].

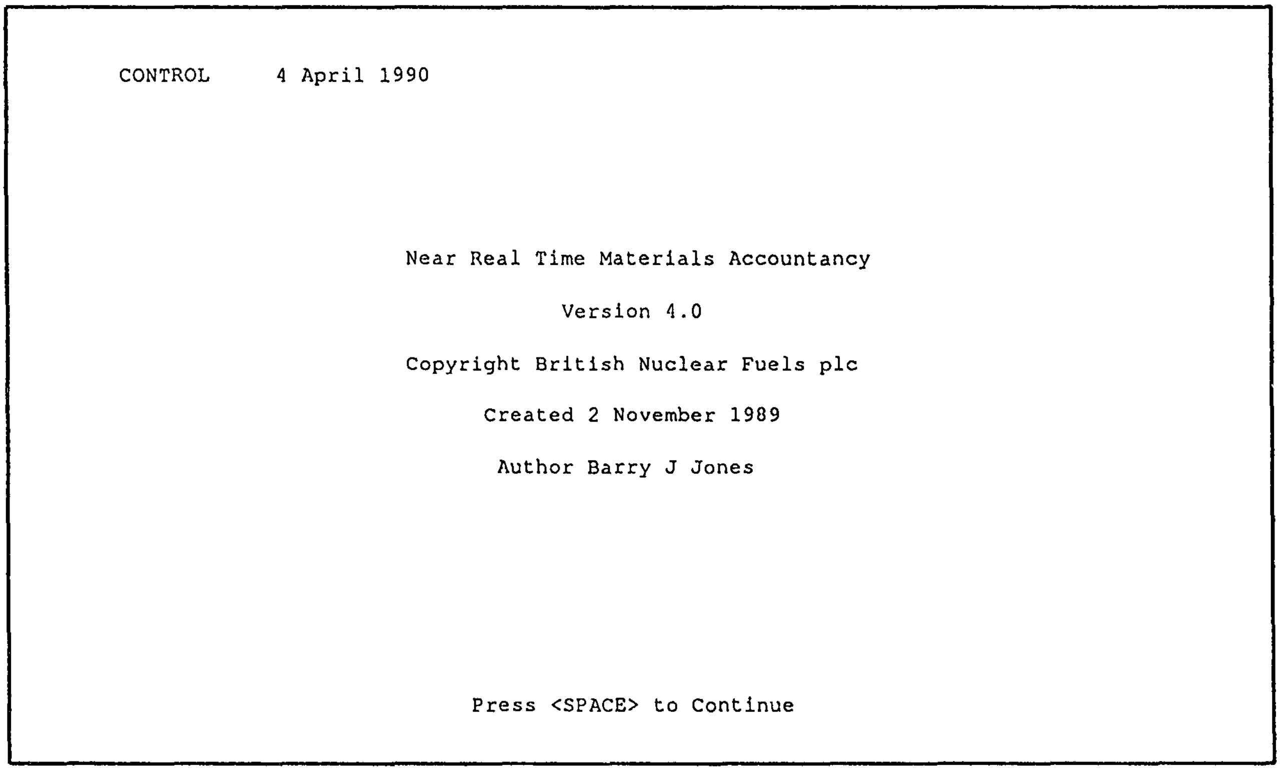
Quit Function

This option allows the user to leave the NRTMA application.

4 CONCLUDING REMARKS

Emphasis is placed on minimizing and simplifying operator input whilst maximizing the return to him of readily useable information.

Figure 1: Copyright Screen



```
CONTROL     4 April 1990



                    Near Real Time Materials Accountancy

                                Version 4.0

                    Copyright British Nuclear Fuels plc

                          Created 2 November 1989

                           Author Barry J Jones




                         Press <SPACE> to Continue
```

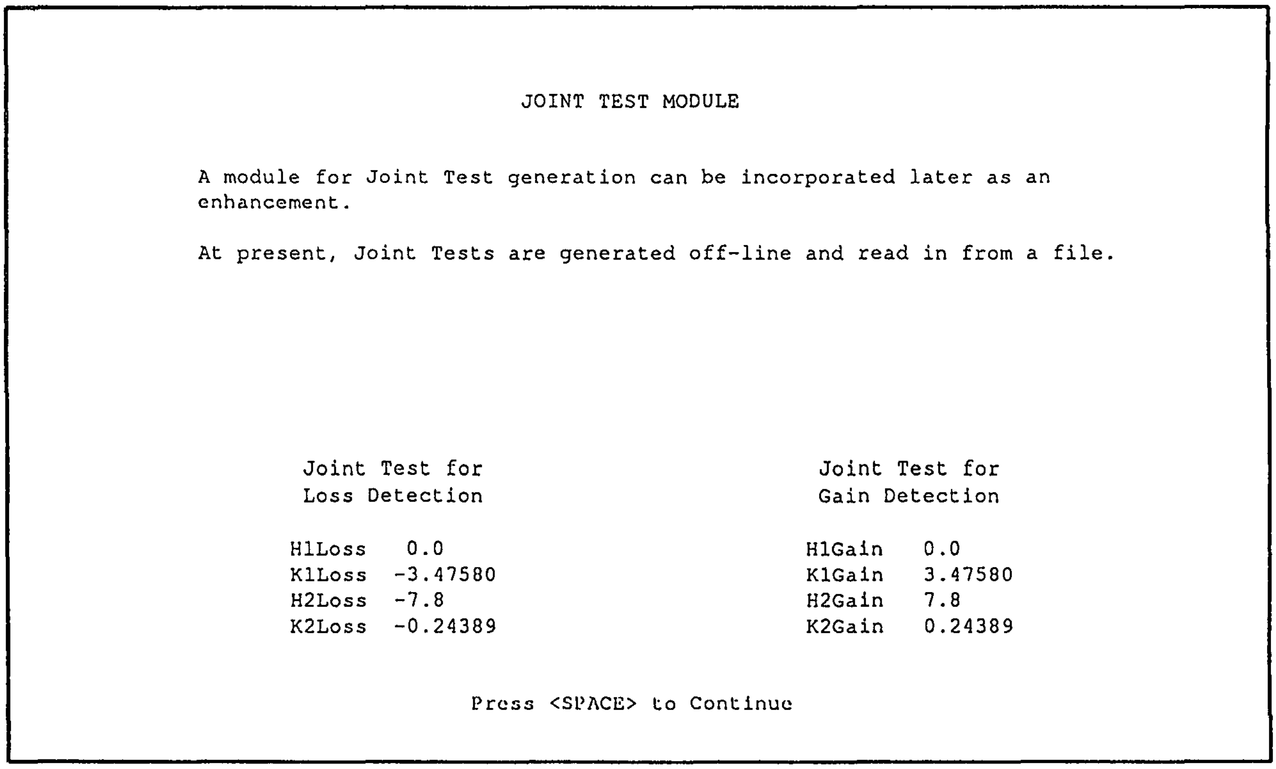


Figure 2: Joint Test Module

```
                        JOINT TEST MODULE

     A module for Joint Test generation can be incorporated later as an
     enhancement.

     At present, Joint Tests are generated off-line and read in from a file.




         Joint Test for                          Joint Test for
         Loss Detection                          Gain Detection

        H1Loss    0.0                           H1Gain    0.0
        K1Loss   -3.47580                       K1Gain    3.47580
        H2Loss   -7.8                           H2Gain    7.8
        K2Loss   -0.24389                       K2Gain    0.24389

                       Press <SPACE> to Continue
```

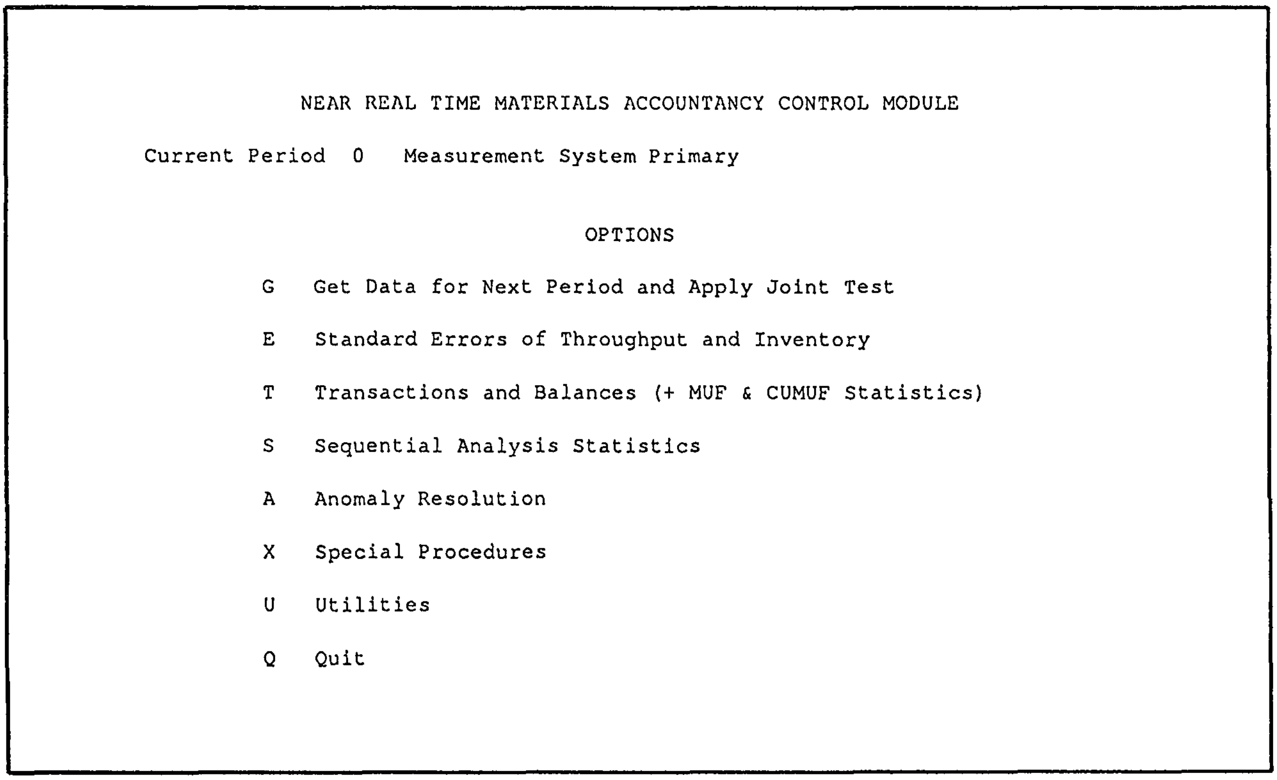


Figure 3: Main Menu

```
          NEAR REAL TIME MATERIALS ACCOUNTANCY CONTROL MODULE

   Current Period  0   Measurement System Primary


                              OPTIONS

        G   Get Data for Next Period and Apply Joint Test

        E   Standard Errors of Throughput and Inventory

        T   Transactions and Balances (+ MUF & CUMUF Statistics)

        S   Sequential Analysis Statistics

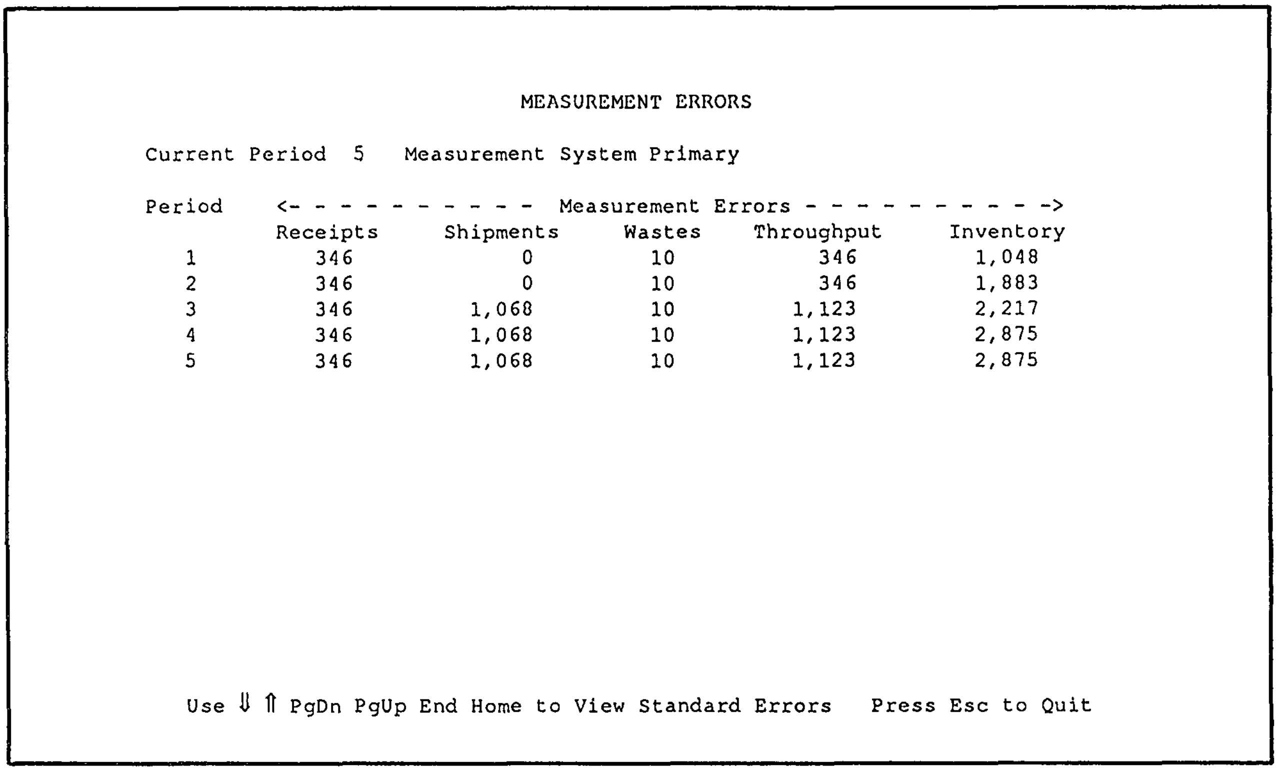
        A   Anomaly Resolution

        X   Special Procedures

        U   Utilities

        Q   Quit
```

Figure 4: Standard Errors of Throughput and Inventory



```
                         MEASUREMENT ERRORS

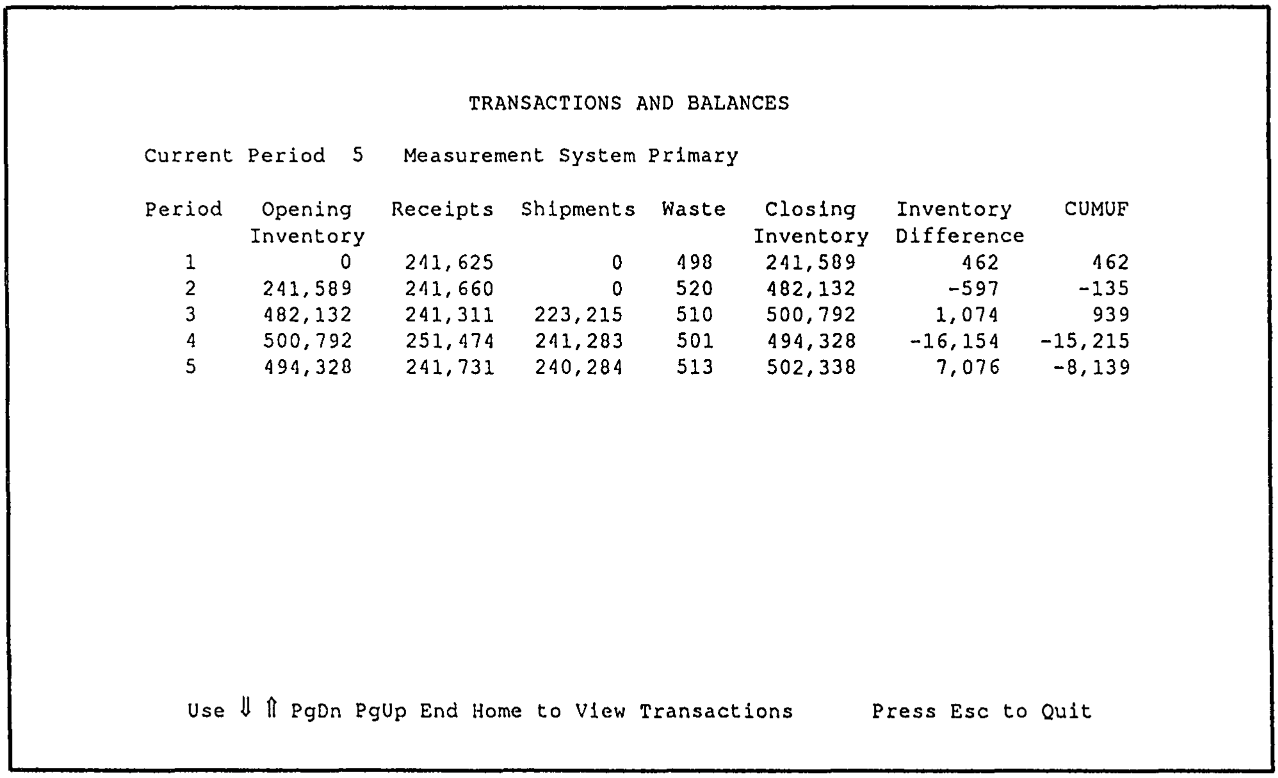
Current Period  5   Measurement System Primary

Period    <- - - - - - - - - - Measurement Errors - - - - - - - - - ->
          Receipts      Shipments      Wastes     Throughput      Inventory
  1          346             0           10           346           1,048
  2          346             0           10           346           1,883
  3          346         1,068           10         1,123           2,217
  4          346         1,068           10         1,123           2,875
  5          346         1,068           10         1,123           2,875




   Use ⇓ ⇑ PgDn PgUp End Home to View Standard Errors   Press Esc to Quit
```

Figure 5: Transactions and Balances (+ MUF & CUMUF Statistics)



```
                       TRANSACTIONS AND BALANCES

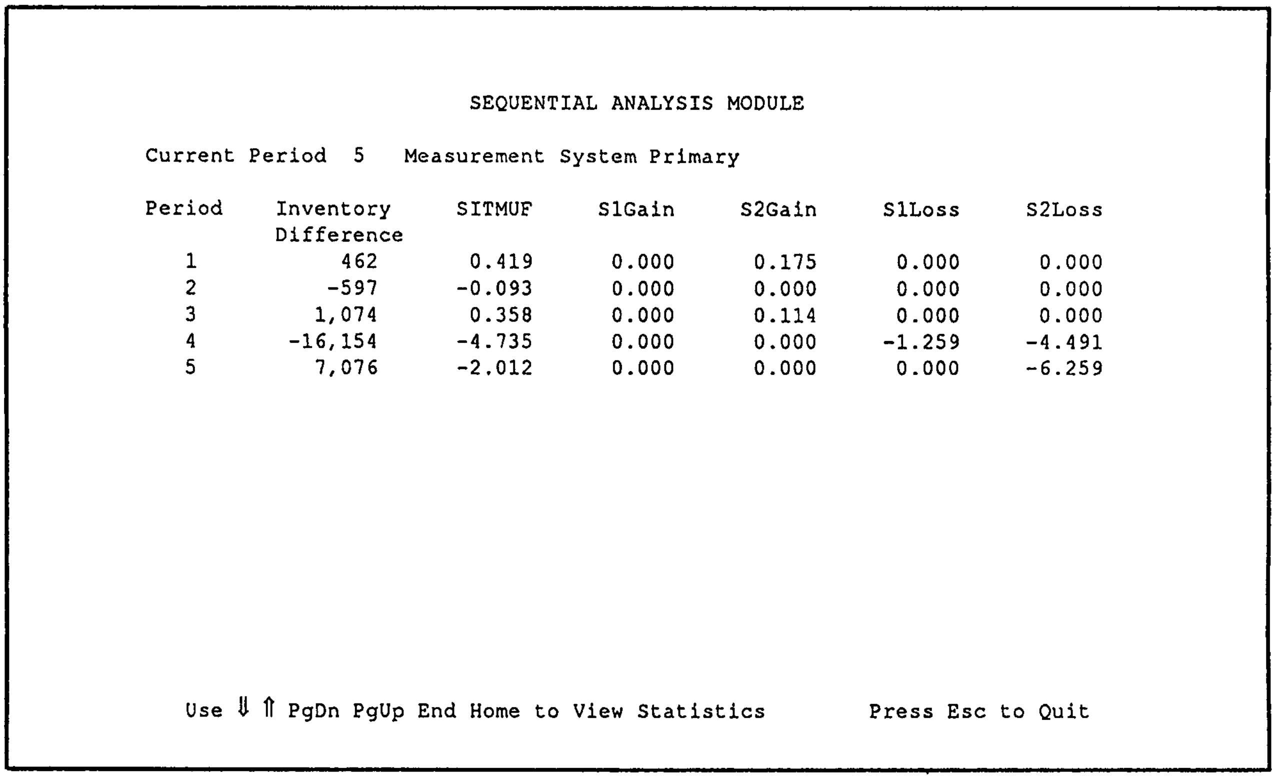
Current Period  5   Measurement System Primary

Period   Opening    Receipts  Shipments  Waste   Closing   Inventory    CUMUF
         Inventory                               Inventory Difference
  1            0    241,625         0     498    241,589        462       462
  2      241,589    241,660         0     520    482,132       -597      -135
  3      482,132    241,311   223,215     510    500,792      1,074       939
  4      500,792    251,474   241,283     501    494,328    -16,154   -15,215
  5      494,328    241,731   240,284     513    502,338      7,076    -8,139




   Use ⇓ ⇑ PgDn PgUp End Home to View Transactions      Press Esc to Quit
```

Figure 6: Sequential Analysis Statistics



```
                           SEQUENTIAL ANALYSIS MODULE

Current Period  5   Measurement System Primary

Period    Inventory     SITMUF     S1Gain     S2Gain     S1Loss     S2Loss
          Difference
   1           462       0.419      0.000      0.175      0.000      0.000
   2          -597      -0.093      0.000      0.000      0.000      0.000
   3         1,074       0.358      0.000      0.114      0.000      0.000
   4       -16,154      -4.735      0.000      0.000     -1.259     -4.491
   5         7,076      -2.012      0.000      0.000      0.000     -6.259

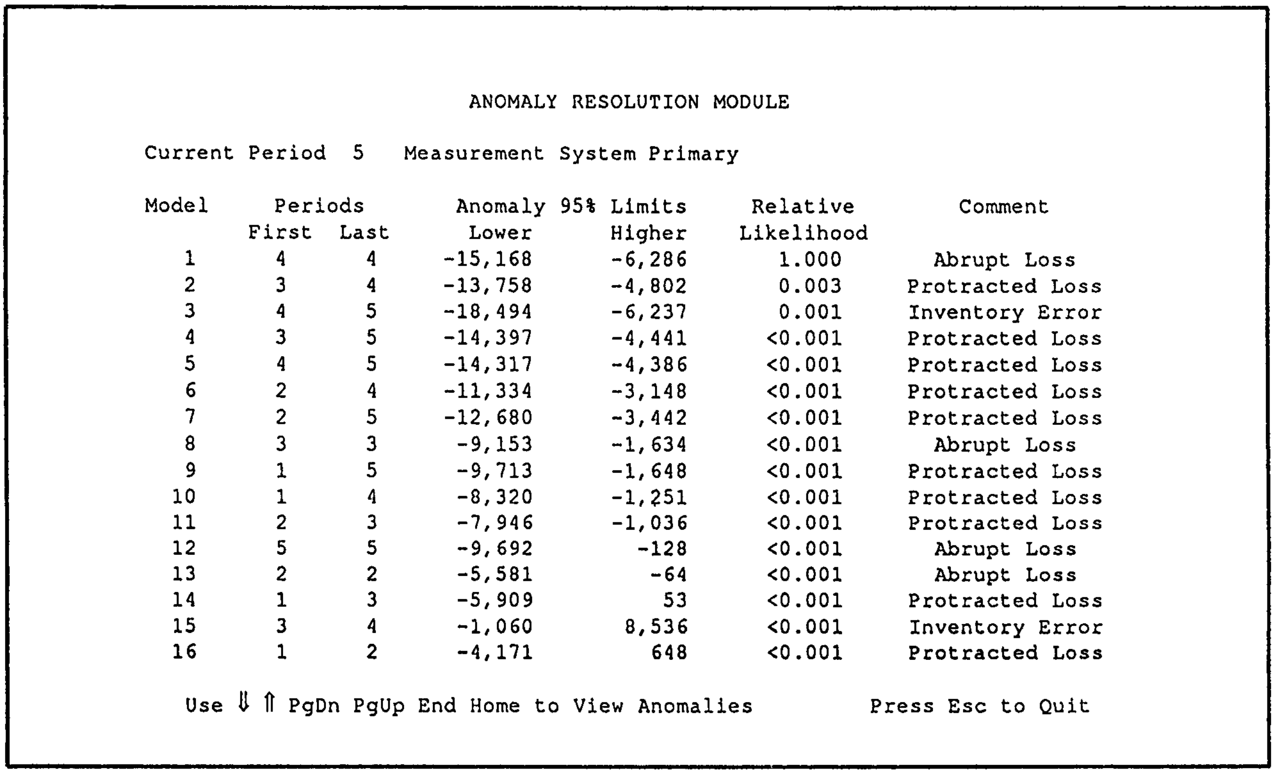






   Use ⇓ ⇑ PgDn PgUp End Home to View Statistics        Press Esc to Quit
```

Figure 7: Anomaly Resolution



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                           ANOMALY RESOLUTION MODULE

Current Period  5   Measurement System Primary

Model     Periods      Anomaly  95% Limits     Relative          Comment
        First  Last     Lower       Higher    Likelihood
   1      4     4     -15,168      -6,286       1.000        Abrupt Loss
   2      3     4     -13,758      -4,802       0.003      Protracted Loss
   3      4     5     -18,494      -6,237       0.001      Inventory Error
   4      3     5     -14,397      -4,441      <0.001      Protracted Loss
   5      4     5     -14,317      -4,386      <0.001      Protracted Loss
   6      2     4     -11,334      -3,148      <0.001      Protracted Loss
   7      2     5     -12,680      -3,442      <0.001      Protracted Loss
   8      3     3      -9,153      -1,634      <0.001        Abrupt Loss
   9      1     5      -9,713      -1,648      <0.001      Protracted Loss
  10      1     4      -8,320      -1,251      <0.001      Protracted Loss
  11      2     3      -7,946      -1,036      <0.001      Protracted Loss
  12      5     5      -9,692        -128      <0.001        Abrupt Loss
  13      2     2      -5,581         -64      <0.001        Abrupt Loss
  14      1     3      -5,909          53      <0.001      Protracted Loss
  15      3     4      -1,060       8,536      <0.001      Inventory Error
  16      1     2      -4,171         648      <0.001      Protracted Loss

   Use ⇓ ⇑ PgDn PgUp End Home to View Anomalies         Press Esc to Quit
```

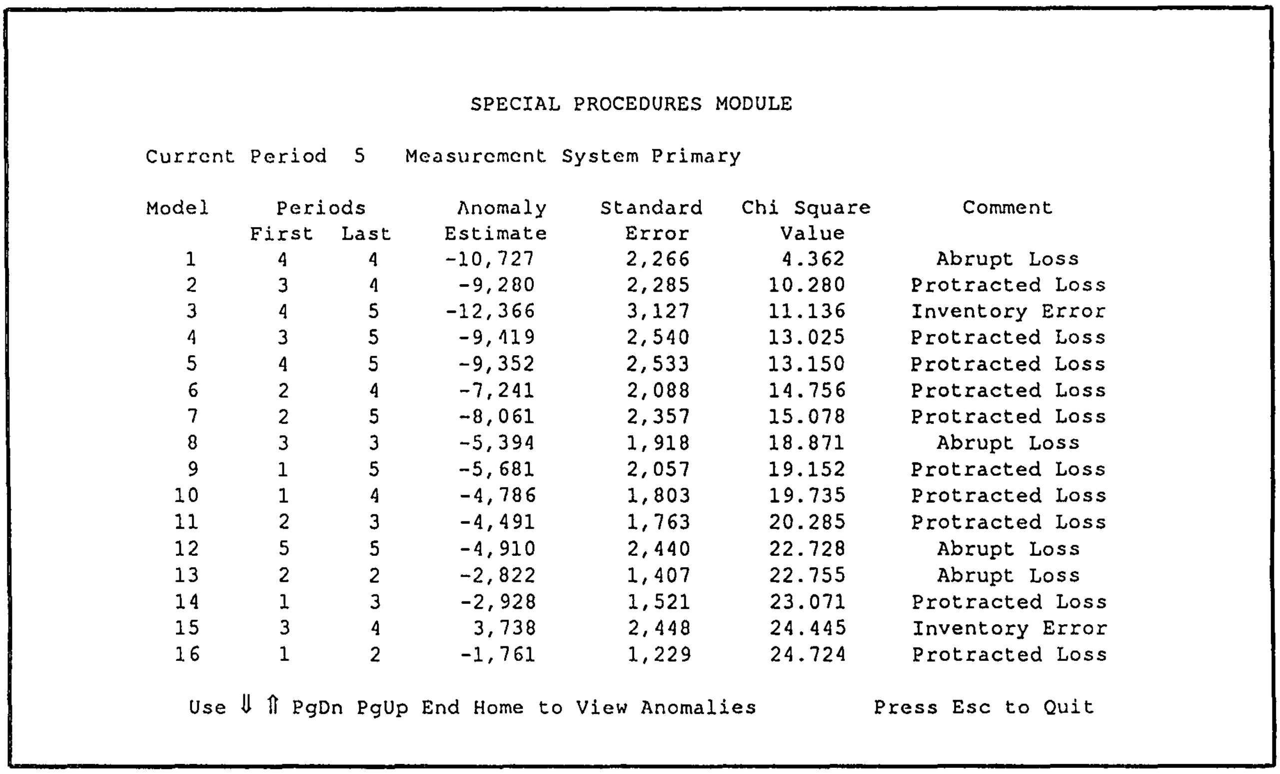

Figure 8: Special Procedure

```
                          SPECIAL PROCEDURES MODULE

  Current Period  5   Measurement System Primary

  Model     Periods       Anomaly    Standard  Chi Square        Comment
          First  Last     Estimate     Error     Value
    1       4     4       -10,727      2,266      4.362       Abrupt Loss
    2       3     4        -9,280      2,285     10.280     Protracted Loss
    3       4     5       -12,366      3,127     11.136     Inventory Error
    4       3     5        -9,419      2,540     13.025     Protracted Loss
    5       4     5        -9,352      2,533     13.150     Protracted Loss
    6       2     4        -7,241      2,088     14.756     Protracted Loss
    7       2     5        -8,061      2,357     15.078     Protracted Loss
    8       3     3        -5,394      1,918     18.871       Abrupt Loss
    9       1     5        -5,681      2,057     19.152     Protracted Loss
   10       1     4        -4,786      1,803     19.735     Protracted Loss
   11       2     3        -4,491      1,763     20.285     Protracted Loss
   12       5     5        -4,910      2,440     22.728       Abrupt Loss
   13       2     2        -2,822      1,407     22.755       Abrupt Loss
   14       1     3        -2,928      1,521     23.071     Protracted Loss
   15       3     4         3,738      2,448     24.445     Inventory Error
   16       1     2        -1,761      1,229     24.724     Protracted Loss

     Use ⇓ ⇑ PgDn PgUp End Home to View Anomalies         Press Esc to Quit
```

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7 JANETTE WARK, "Near Real Time Materials Accountancy: Resolution of Anomalies", *Proceedings of the Thirteenth Annual ESARDA Symposium on Safeguards and Nuclear Material Management*, -, (1991)

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MODEL-BASED DIAGNOSIS
IN NEAR-REAL TIME MATERIALS ACCOUNTANCY

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Abstract

NRTMA systems must be capable of diagnosing anomalies as well as detecting them because of problems associated with, for instance, obtaining *apriori* knowledge of systematic errors. A question therefore arises as to whether methods developed for fault diagnosis may be equally applicable in NRTMA. This possibility is examined and a model-based approach to diagnosis is described.

1. Introduction

It has been argued elsewhere /1-3/ that near-real-time materials accountancy (NRTMA) systems installed in reprocessing plants must diagnose as well as detect because of the likely occurrence of false alarms induced by systematic errors, transcription errors and so on. If a NRTMA system is to perform effectively in a reprocessing facility, it must therefore be able to build on experience gained in previous campaigns and investigate data irregularities.

The idea of an intelligent knowledge-based system (IKBS) providing valuable assistance in this situation has been mooted /1/ previously. A number of approaches are possible depending on factors like the type of knowledge available, methods of representation and manipulation. The simplest and most widely proposed method for representing knowledge is with if-then rules; for example

> if *the accountancy tank has just been emptied*
> then *the level in the plant feed tank > 1m.*

A large number of these rules can be strung together to relate observed symptoms to implied underlying causes and faults. Such systems tend to lack depth because although terms like *empty* and *full* can be quantified the relationships between the various plant components do not represent the mechanism by which these rules are obtained: the underlying physical relationships have been eliminated from the description. There is therefore a strong argument in favour of incorporating deep, or model-based, knowledge into any diagnostic system. This in turn requires mechanisms to reason about the models constructed.

Although little has been published about the model-based diagnosis of anomalies in NRTMA, considerable research/4,5,6/ has been carried out into model-based diagnosis of faults in general. The question therefore arises as to whether this research is equally applicable to NRTMA. There is no conceptual difference between faults and anomalies. Materials accountancy is to do with ensuring that the measured or estimated flow of material through a plant balances; a fault may be deemed to be anything that upsets this balance.

This paper first puts model-based diagnosis into context by briefly outlining an IKBS structure to meet NRTMA requirements It goes on to discuss the most relevant aspects of current research into fault diagnosis and proposes a diagnosis methodology that is suitable for the information content that is likely to be available from reprocessing plants. That is a methodology which will accommodate the quality of data and models thought to be available. A diagnostic procedure is outlined and its capabilities are assessed by examining its performance on a reprocessing plant simulation.

2. A Suitable Knowledge-Based System Structure for NRTMA

It is argued elsewhere /7/ that an appropriate knowledge-based structure could have the following knowledge sources:

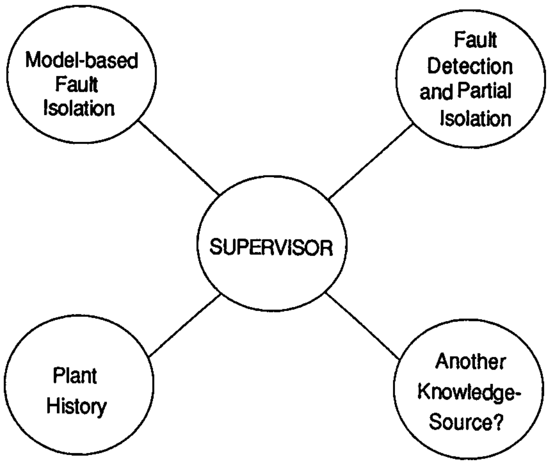

Figure 1 : Knowledge Sources

The system could be invoked everytime a set of plant measurements becomes available. This data would then be input to the fault detection and partial isolation knowledge-source which would be subsequently activated.

The primary role of this knowledge-source is to detect the occurrence of an anomaly and having done so, to perform a pattern matching exercise on, for instance MUF, CUMUF and CUSUM plots. It outputs two lists: a list of symptoms or discrepancies and a list of assertions which point to possible classes of anomalies that could account for the patterns observed. It would be unusual for the charts to identify an anomaly uniquely; their role is to focus attention. Two issues complicate the recognition process, that of noise and of multiple faults (anomalies). Both can largely be overcome by adding any fault, that is remotely likely, to the list. Care must then be taken to ensure that the most likely are considered first.

The Supervisor is driven by data flowing from the fault detection and partial isolation knowledge source. It has two roles, that of an evidence gatherer and hypothesis generator and that of an *adjudicator*. We use the term *gatherer* to refer to the former. The basic mechanism behind the Gatherer is as follows. On receipt of a list of possible fault scenarios, the Gatherer takes each fault scenario in turn and invokes one or more of three options: a model-based analysis, a reference to history or its own assessment. In most situations it will invoke all three. Each option returns either a statement of its deliberations in the form of hypothesis explanations or nothing at all. Finally the *adjudicator* is invoked to assess the hypotheses gathered. Little research has been carried out into the process of adjudication. The current state assumes that the operator will make his own assessment based on the hypotheses formed.

Model-based fault diagnosis is therefore viewed as contributing only one aspect to the overall strategy.

3. Introduction to Model-Based Fault Diagnosis

What do we mean by model-based fault diagnosis?

In its simplest form /5/ it can be considered as consisting of a parallel simulation predicting plant measurements, a comparison between these predictions and the actual measurements and a means of generating fault hypotheses on the basis of any discrepancies obtained.

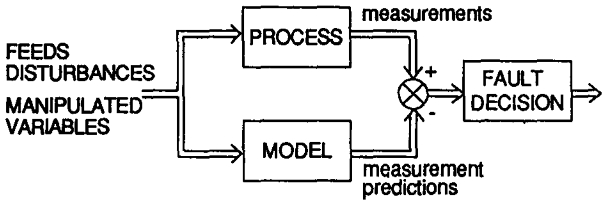

Figure 2 : Model-based Fault Diagnosis

A slightly more sophisticated arrangement /8/ is where the simulation outputs model parameters instead.

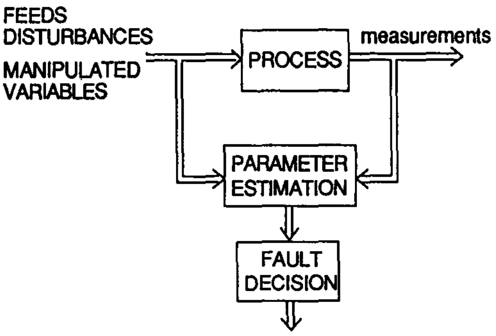

Figure 3 : Model -Parameter-based Fault Diagnosis

Processes need not be modelled quantitatively, various techniques have been proposed which reason with qualitative models instead /9,10/. However these are unlikely to be of interest to materials accountancy which is singularly numerical in nature. Virtually all research into quantitative techniques has focused on plants or systems that are *information rich*: either in the sense that there is a proliferation of sensors throughout the plant, the outputs of which are of a sufficiently high quality and are recorded at frequencies much higher than that of underlying process fluctuations or in the availability of models that are accurate in terms of structure and parameters or both. If this was the case in NRTMA, then only actual diversions would be alarmed because the detection system would be able to compensate for any other anomaly that might arise. Reprocessing plants tend to be *information poor*. Howell /11/ has proposed a common sense approach to fault diagnosis in information poor processes and it is this approach that is followed here.

It is not our concern here to digress into models, measurements and their inaccuracies. The reader is asked to accept that such inaccuracies can exist and can have various affects on the discrepancies observed. To give an idea of scale, the plant model discussed later requires 26 variables to describe the time variations in its principal inaccuracies assuming that the structure is correct. There is no guarantee that a particular pattern of discrepancies can be attributed to a particular fault because various permutations of model and measurement system inaccuracies may have a similar effect. The following formula is therefore proposed /11/ to represent the various possibilities.

The process, plant or system is divided into parts along physically meaningful boundaries and a model is constructed for each part. Variables describing the flow across boundaries are defined as path variables. Neither measurements nor model inaccuracies need be confined to these boundaries and a Principle of Re-Distribution /7/ is introduced to formalize this new representation. This principle provides a common approach to the categorization of the effects that individual model inaccuracies, measurement errors and faults have on the discrepancies. Any model inaccuracy, measurement error or fault can be identified with a single element contained in one of two mutually exclusive sets, Θ, the set of path errors, and U, the set of non-path errors. One difference between them is that each element of Θ is always present which is not the case with elements of U. For instance, an analogue measurement can only be obtained to the accuracy of the measuring device; some error will always exist. A set Θ^* is then defined as containing those elements of Θ that are significantly in error in the sense that, for instance, the error cannot be explained by normal inaccuaracies in the measurement system.

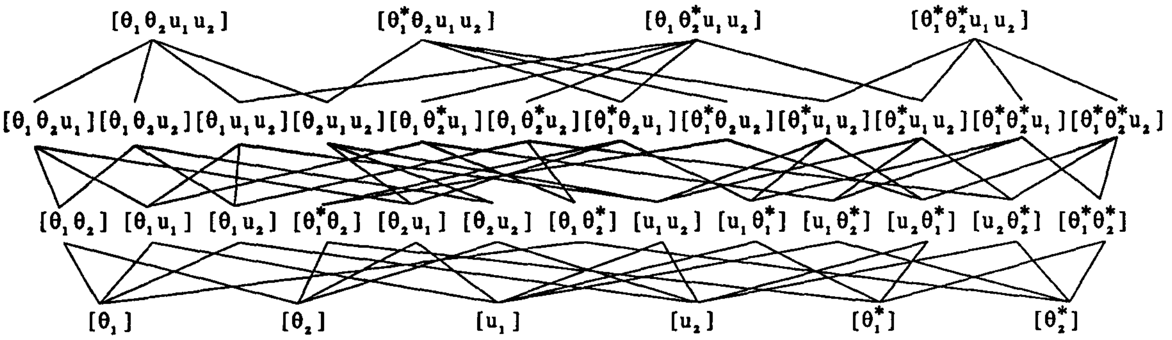

Figure 4 : A Typical 4-Element Candidate Space

A candidate is now defined as a particular hypothesis for how the plant differs from the model. That is, it is a set of elements of $\Theta \cup U$ whose values could, hypothetically, be manipulated to explain the discrepancies observed. Figure 4 shows the candidate space for a simple example containing just two path and two non-path errors. Each set of explanations, indicated by [.], is possible giving 35 candidates in all. More realistic situations would involve much larger candidate spaces. The space, shown in Figure 4, can be viewed as a lattice composed of 4 sub-lattices, each starting at a different 4-element node with only one node containing purely insignificant errors. If considerable uncertainty surrounds the diagnosis, then candidates at the top of the lattice are likely to be true; the greater the certainty, the more likely the candidates towards the bottom of the lattice become.

The number of explanations contained in each of the candidates towards the top of the lattice are likely to be far greater than the number of symptoms available to perform a diagnosis. In these circumstances it is difficult to discriminate between the various candidates unless some of the explanations can be eliminated on grounds of inter-dependence or through non-symptom related arguments.

Howell /11/ has proposed a way forward based on common sense reasoning and has developed techniques to evaluate the various sets of possibilities. Based on subjective probabilities and estimation theory, they are beyond the scope of this paper.

4. Model-Based Diagnosis in NRTMA

Figure 5 shows the knowledge-based system that has been developed to combine the ideas of the previous two Sections.

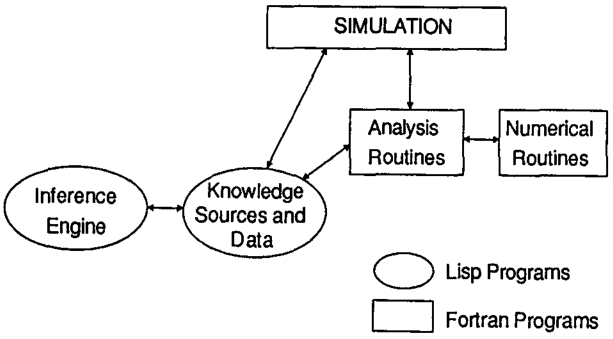

Figure 5 : Proposed Knowledge-based System

It consists of a hybrid lisp/Fortran environment with the lisp environment acting as host calling Fortran routines when necessary. A hybrid implementation is preferred because it combines the numerical affinity of Fortran with the list and symbolic processing powers of lisp. The Fortran environment is composed of simulation, analysis and numerical routines. The numerical routines are a library of routines to perform matrix algebra, to solve a set of linear simultaneous equations and to generate random numbers for test purposes. The lisp environment consists of knowledge-sources, an inference engine and various global data structures and methods.

5. An Example

This Section discusses the steps that have been taken to develop a model-based diagnosis system for a particular plant; in this case the hypothetical solvent-extraction and concentration plant shown in Figure 6. Ideally, there would be three stages to such a development:

1. form a skeletal knowledge-base and produce simulation and analysis routines;
2. test the functionality of the resulting system using data output from the simulation;
3. assess its performance on the real plant.

Unfortunately neither a plant nor resources were available to perform the third stage. A relatively superficial assessment of its performance was therefore made by testing it against a simulation designed to reflect some of the uncertainty surrounding the model.

5.1 Knowledge-sources, Simulations and Analysis

There is insufficient space here to discuss the knowledge-sources that must be constructed to interpret alarms generated from control charts, represent operator experience and so on. The discussion will be limited to the model-based part of the system.

With the absence of a real plant for comparison, it was assumed that the structure of the model incorporated into the fault diagnosis system was the simplest possible. That is, one with perfect mixing in all the buffer tanks and with the ratios of the various feeds in the solvent-extraction plant maintained constant. The only random errors were those assumed to exist in the individual measurements.

System performance was then examined by diagnosing faults generated from a simulation in which non-perfect mixing and variations in a number of feeds and parameters were, or were not, be imposed. To be concise, only two plant simulations are considered here,

Plant A:

i) random errors applied to individual measurements;

ii) fluctuations in each of the elements initially contained in the set of simulation variables, Θ:

Plant B: as above plus

iii) only the top quarter, of the total possible volume, of each buffer tank is mixed;

iv) 0% mixing in the buffer tanks prior to sampling;

v) 0.5% fluctuation in the nominal load of the solvent-extraction plants;

vi) 0.5% fluctuations in each of the feeds to the first cycle of the solvent-extraction plant:

where an n% fluctuation is modelled as a multiplicative gaussian random error with standard deviation, n/100. In all, 16 different random sources were included in Plant B in addition to those used to corrupt the individual measurements. It must be emphasised again that these plant simulations were performed for the purposes of experimentation and not to depict reality.

5.2 Performance of Model-Based Reasoning

System performance was assessed by examining its ability to detect and diagnose the following *faults*:

A. an erroneous measurement of volume (equivalent to 3.7 σ_{MUF}) of the plant feed tank (buffer tank A) on period 5;

C. a diversion from the concentrator (equivalent to 4.2 σ_{MUF}) on period 5;

D. solvent_extraction_A plant load incorrectly specified on period 5 as 60% instead of 100% load; this will cause the simulation to either over or under predict the inventory in the plant feed tank with an associated under or over prediction of the inventories of buffer tanks B and C. In addition, the estimated inventory of the relevant solvent extraction plant will be in error.

The simulation runs were restricted to 16 periods in all cases because of the not insignificant amount of data required like switching times. Ten experiments were performed, giving ten sets of data in all. Although an attempt was made to make the 10 sets different, a certain amount of similarity still existed because of difficulties in generating operational times which do not result in tanks being emptied.

Table 1 shows the results obtained when model-based reasoning was applied to data collected at the end of a single period, on the assumption that all path faults were insignificant. This is a limitation of the current implementation.

The following observations were made.

Fault A: the correct measurement error was identified in all cases;

Fault C: diagnosis was incorrect because it should have been performed over two periods although it did identify the correct plant component;

Fault D: one diagnosis, per plant, failed completely whilst the other 9 required at least 2 non-path explanations. A question therefore arose as to whether or not these 2 explanations could be replaced by a single, significant path explanation. That sub-lattice containing no insignificant path errors was therefore examined for those datasets containing Fault D and the following results were obtained:

		CANDIDATES						
		p = path np = non-path						
Plant	Fault	np	p+np	2np	2p+np	p+2np	3np	np identified
A	A	6	4					pft_vol
	C	7	3					prod_storA_vol
	D					8	1	pft_vol +buf_tankB_vol (+prod_storA_vol)
B	A		7		3			pft_vol
	C	1	7		2			prod_storA_vol
	D					8	1	pft_vol +buf_tankB_vol (+prod_storA_vol)

Table 1: Fault Studies using Insignificant Path Sub-lattice

Plant A - $7*p^* \& 3*2p^*$

Plant B - $5*2p^* \& 1*(p^*+np) \& 4*3p^*$

where a particular p^* in every case was that composite element that contained both solvent_extraction_plant_A_load and solvent__extraction__plant__B__load. Each sub-lattice containing a single significant path error which was identical to one of those identified was searched next. Only 2 significant path errors were successful in the case of Plant A, the 2 loads and the following candidates were identified,

Plant A - solvent_extraction_plant_A_load*
+ (7 & 3*p)

Plant B - solvent_extraction_plant_A_load*
+ (4*p & 2*np & 4*2p)

5.3 Performance Overall

The discussion will be limited to that pertaining to Plant A, Fault A because there is insufficient space to describe all the results here. Indeed this example is only given to elaborate on system performance and to demonstrate its potential. Considerably more development is required before the system can be viewed as being operationally viable.

At present, the consequences generated by the Control–chart and Supervisor knowledge-sources are held in

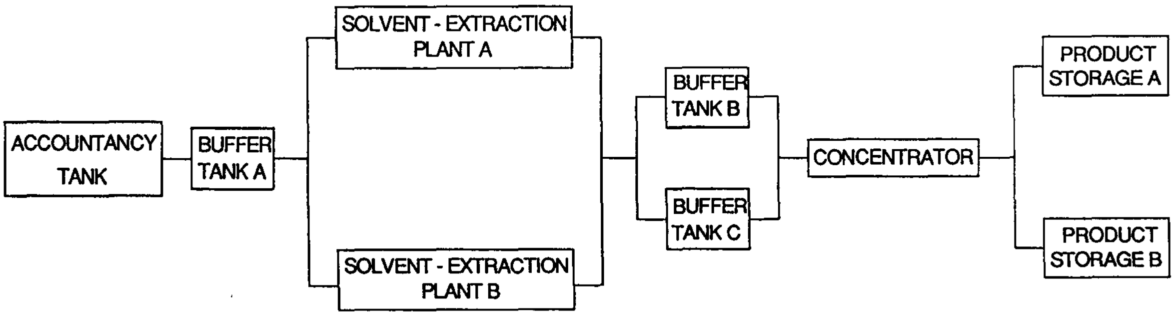

Figure 6 : Plant Layout

a common assertion base, *goals*. Model-based reasoning and history have their own assertion bases, *sim_asserts* and *corr_asserts*. The following assertions have been selected from the contents of the various assertion bases generated when the system was invoked at the end of the period subsequent to the anomaly having occurred.

goals - as produced by the Control Chart knowledge-source

```
(INV-LOSS+SIM-OUT YESTERDAY)
(INV-LOSS YESTERDAY)
(SINGLE-TRANSFER-ERROR YESTERDAY)
(SINGLE-INV-ERROR YESTERDAY)
(CUSUM-P YESTERDAY)
(MUFTEST-P YESTERDAY 316.498)
```

The Supervisor takes each assertion indicating an anomaly (ie the top 4) and assesses the possibility by generating LOOK-FOR assertions. These, in turn, are acted upon.

goals - produced by the Supervisor,

```
(LOOK-FOR-FAULT S-I-E YESTERDAY
     (YESTERDAY YESTERDAY))
(LOOK-FOR-FAULT S-T-E YESTERDAY
     (YESTERDAY TODAY))
(LOOK-FOR-FAULT I-L YESTERDAY
     (YESTERDAY TODAY))
```

sim_asserts - as produced by the analytical approach,

```
(S-I-E ((PFTVOL-MEAS -9.20707)) NIL 1)
(INTERPRET-MODEL S-I-E
          ((0 NIL (((1 -0.492931)
                    (991 -9.20707)))
                    (0.965009))))
```

Only one possible explanation was generated on the second row of the insignificant path error sub-lattice and the search was terminated. (The first row having identified no suitable candidates.) A reduction of -9.2*l* in the simulated volume in the plant feed tank would explain the measurements. That is, there was a measurement error of -9.2*l* as compared to an actual error of -10*l*. An insignificant error in the volume input to the plant is also needed to explain the discrepancies.

6. Conclusions

A knowledge-based system structure has been described which incorporates and subsequently analyses the outputs of statistical tests applied to MUF series with surface knowledge about previous experience, operator heuristics and plant peculiarities and with model-based reasoning.

It has been shown how anomalies can be diagnosed by taking a common sense approach to reasoning about differences between simulations based on quantitative models and plant data. A number of successful experiments have been performed to prove the efficacy of the approach. However considerable effort is still required to demonstrate its potential in general. In particular, experiments to date have been with simulated data in a university environment. A demonstration on a real plant is required before the approach would be acceptable to the NRTMA Community.

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TORNESS COMPUTERISED FUEL RECORDS SYSTEM

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Abstract

Early during the construction of Torness Power Station it was decided that a computerised record system would be developed to enable the Station to comply with current legislation relating to the control of fissile material. The resultant system was delivered in stages to match the commissioning of the various sections of the Fuel Route and has been used successfully now for five years. Information on fissile material is updated in 'real-time' and all statutory and mandatory reports required by Regulatory bodies are provided, thus eliminating potential sources of errors.

1. Introduction

Torness Power Station, located near Dunbar in East Lothian Scotland, is owned and operated by Scottish Nuclear Limited. Torness Power Station consists of two Advanced Gas Cooled (AGR) nuclear reactors with a total generating capacity of 1400 megawatts.

The Computerised Fuel Records system (Fuelroute) is a complex data processing system which tracks the movement and storage of all Fissile Material and associated Reactor components from receipt on the Station, to dispatch off the Station for reprocessing or disposal into the High Active Debris Vault in the case of non-reusable items. The system was designed to provide a database of current and historical transactions which could be interrogated to provide the wide variety of reports required by Scottish Nuclear personnel and outside regulatory bodies.

The software requirements of the Fuelroute Programme was specified by the Fuel Records Engineer at Torness Power Station and produced in accordance with an approved Project Plan by ScottishPower Plc. It was agreed at the onset that all Software would be delivered from ScottishPower plc. in batches to match the commissioning of the various sections of the physical Fuel Route.

2. History

At an early stage during the construction phase it was decided that, wherever possible, all information at Torness Power Station would be 'Computerised'; since there was no known computer system readily available it was necessary to develop a computerised Fuel Records system from scratch.

This computerised record system now forms part of the Torness Management Information System and is considered as the definitive Station based Fissile Material Record. The system is used for recording the description, location, movement and use of all Fissile Material and associated components at Torness Power Station.

The system has been used successfully now for five years and has proved to have three very important advantages over a paper based system:

1) the data base is always up to date, being updated in 'near' real time

2) data is subject to rigorous vetting, thus giving confidence in the information at all times

3) the database can be easily and speedily interrogated day or night to give either standard or miscellaneous reports. The database can also be interrogated remotely from the Company Headquarters in the unlikely event of a serious Nuclear Site Incident.

Since the software was delivered in phases, this allowed the users to become familiar with using it gradually and to gain confidence in the system. It also allowed the project team designing the software to take into account users' comments. Throughout the design and production phases close collaboration was maintained at all times between Station Staff and ScottishPower Information Systems Department.

3. Requirement

At the commencement of the design stage of the computerised record system it was stated by Station Management that the following requirements were paramount and had to be met in totality by the system:

- To be able to demonstrate that the Nuclear Site Licence conditions are being complied with in respect to the movement and storage of the Fuel Route items during the life of the station.
- To enable auditing of the above Nuclear Site Licence conditions by Regulatory Bodies.
- To computerise the record keeping of all operations within the physical Fuel Route.
- To maintain a database of all above transactions for the Station's Lifetime Records.

For the Physical Fuel Route at Torness Power Station to be able to function effectively the following information must be available at all times:

- Details of all fuel elements, associated reactor components, fuel flasks, skips and bottles delivered to the Station eg. Identification Numbers, Location and contents.
- Fuel and components specified for fuel assembles, ie. Stock Levels, Identification Numbers and Location.
- Fuel and components used in fuel assembles.
- Discharge instructions detailing the channel to be discharged and where the constituent parts of the fuel assembly are to be stored.
- Conformation of the dismantling process and compliance with the discharge instructions.
- Details of Irradiated and Un-irradiated fuel, Irradiated fuel flasks etc. dispatched from the Station.
- Records of all Fissile Material movement within the station.

4. Hardware

This programme runs on a clustered Digital VAX computer system, comprising two VAX 8550's and a VAX 6310. The programs and data occupy 30 Megabytes of disk storage and access by users is via terminals or personal computers attached to a local area network. Security on the VAX system is ensured by use of file protection, access control lists and further restrictions built into the executable image.

It is Station Policy that all Engineers have personal computers or have access to a terminal when required. In addition to personal terminals there are also a number of terminals strategically located around the plant and throughout the physical Fuel Route. These terminals are linked to the cluster via a Local Area Network which utilises glass fibre cable because of the distances involved. Having a number of terminals available enables all Station Staff, wherever they are located, to have access to the information as required.

The cluster is also linked via a Wide Area Network to other computers including the development machine.

5. Software

The system was produced according to a Quality Assurance Plan which followed the guidelines contained within BS:5750 and thus detailed responsibilities, deliverables, standards/methods/tools to be used to ensure quality, and reviews.

The software was designed using a structured methodology. Various computer languages were used where applicable (ie COBOL, BASIC, FORTRAN, and DATATRIEVE - DEC high level query language) and the screen user interface uses DEC's Terminal Data Management System (TDMS).

The comprehensive database is a series of Index Sequential files which were designed to minimise redundancy of data. Record Locking is utilised to ensure that no inconsistencies in the data arise from multi-user access.

The project team developed the system on their own computer which is 150 kilometres from the target computer at Torness Power Station. When preparing the Software Requirements Specification, and when unit and system testing etc, maximum use was made of the Wide Area Network (WAN) linking the two sites. This method worked very well indeed and allowed the project team to develop the computer system very much in partnership with the end users.

6. System

The multi-user system is menu based with extensive on-line help facilities. It is updated in 'near' real time by the Sections responsible for Fuel Records, Reactor Physics, Fuel Handling Operations and Fuel Handling Maintenance.

In addition to the 'Live' database a Training database is maintained for staff in these sections. This Training database is an exact copy of the live programme and as such can provide realistic training for all.

Security within the Station Vax Computer system is strictly controlled in the first instance by the Computer Section only issuing User Accounts to named individuals and each user having their own password to permit access to the system. There are further security controls built into the system, e.g. all users are placed into sections and access to various activities are limited to nominated sections and in some cases to named individuals.

Security within the Fuelroute System is further controlled with access only being granted to named users by the Fuel Records Engineer. Only approved users are allowed access to the software and each approved user has access only to the facilities required to carry out his involvement within the Fuel Route. The data is thoroughly vetted before the database is updated and an audit trail, detailing who performed a particular transaction and when, is maintained of all transactions. Many of the security features of the VMS operating system are also utilised to ensure the security of the data.

Data is protected from loss by the implementation of 'Volume Shadowing' which ensures that a back-up copy of the data is always available within the system. In addition to Volume Shadowing all data is archived on a weekly basis and is stored at two separate locations for security reason. This system of duplication of files ensures that, even in the worst possible scenario, the maximum loss of information is one week's data which could be recovered within a fairly short period of time.

7. Reports

It is one of the functions of this computer system to provide status reports on demand. These reports can either be produced in a paper format or magnetic tape for down loading directly onto other computer system as required.

Examples of computer produced reports are:

1) Euratom Reports eg. ICRs, MBRs and PILs - These above reports can be produced in the format of Magnetic Tape for direct transfer of data to Euratom Headquarters in Luxembourg

2) Details of the isotopic content of the High Active Debris Vault - Required for the government's Scottish Office.

3) Reactor Core contents.

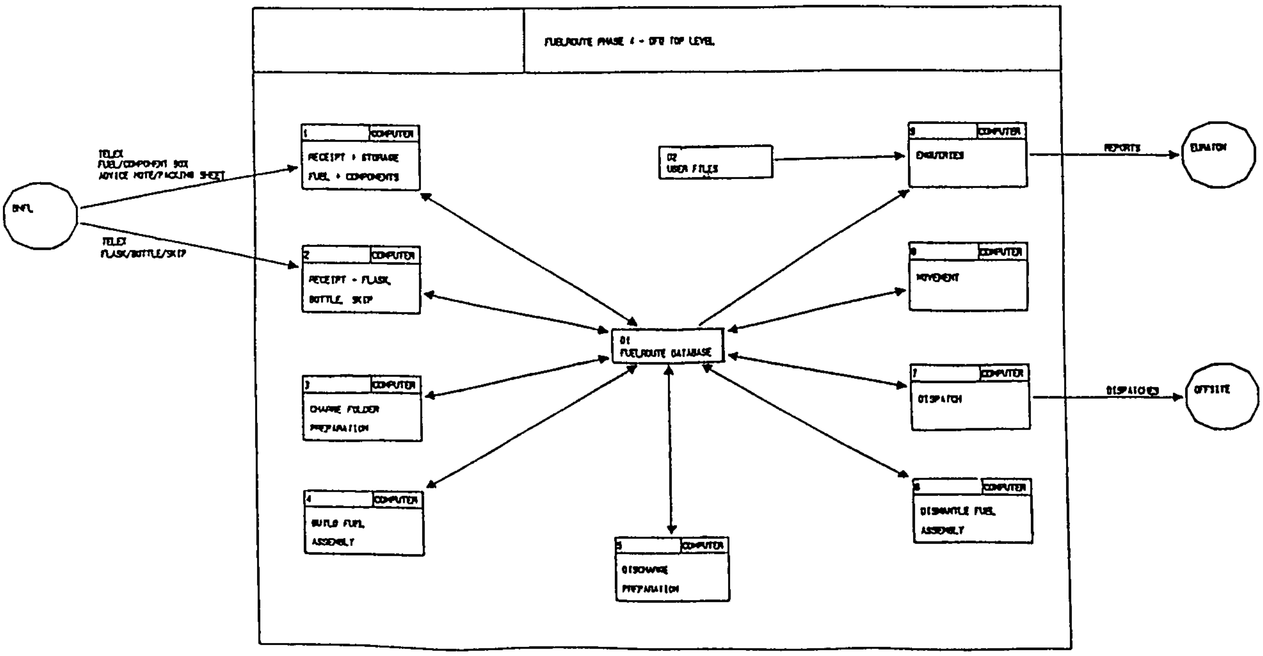
4) Station Stock levels of all Reactor Fuel and components

5) History of all fuel assemblies during their lifetime at Torness.

8. Top Level Functions

This section contains a top level description of each of the main functions that make up the computer system and the top level Data Flow Diagram (figure 1) shows them diagramatically.

Fig. 1 - Top Level Data Flow Diagram (DFD)



FUNCTION 1: FUEL & COMPONENT RECEIPT AND STORAGE

The Receipt and Storage function deals with the delivery to Torness of boxes of fuel elements and reactor components and their associated telex, advice note and packing sheet details, it also deals with their storage within the reactor building. The following items are delivered in boxes - Fuel Elements, Tiebars, Bottom Support Reflector Assemblies (BSRAs), Top Reflectors Assemblies (TRAs), Central Inertial Collectors (CICs), Finned Upper Stabilising Brushes (FUSBAs), 'E' Seals, Collets and Sleeves.

FUNCTION 2: FLASK, BOTTLE, SKIP RECEIPT AND STORAGE

This function also acts as a goods inwards, providing facilities to enter flasks, bottles and skips, coupled with their telex, advice note and packing sheet details into the system.

FUNCTION 3: CHARGE FOLDER PREPARATION

This function handles the inter-departmental preparation of a statutory charge folder which is required to be produced prior to the loading of a fuel assembly into a reactor channel. The charge folder contains the details of the fuel elements and components to be used in the building of a fuel assembly and the tests that they must undergo as defined by Reactor Physics Section.

FUNCTION 4: BUILD FUEL ASSEMBLY

This function handles the actual building of the fuel assembly as specified within the charge folder. At this stage individual fuel elements and components which have failed the specified tests may have to be replaced by compatible ones. This sub-system also provides a facility for quarantining (and un-quarantining) the failed fuel elements and components and will confirm that all alternatives used are of the same type as the original specification.

FUNCTION 5: DISCHARGE FOLDER PREPARATION

This function handles the multi-departmental preparation of the statutory discharge folder which is required to be produced prior to the removal of any fuel assembly from a reactor channel.

FUNCTION 6: DISMANTLE FUEL ASSEMBLY

This function handles the actual dismantling of the fuel assembly as specified in the discharge folder.

After a fuel assembly has completed its life in the reactor and using the appropriate discharge folder, the fuel assembly may be dismantled and its constituent fuel elements and components are placed, as specified on the discharge folder, into skips, bottles or into the High Active Debris Vault (HADV). The facility allows the user to overrule the discharge folder instructions for operational reasons. Other items may also be dispatched to the HADV. The isotopic contents of the fuel elements may be calculated when required.

FUNCTION 7: DISPATCH

This function completes the Fuelroute cycle by acting as goods outwards. It controls the dispatch of boxes, fuel elements, components, flasks, bottles and skips from the Station.

FUNCTION 8: MOVEMENT

This function allows the Fuelroute System Manager to move any Fuelroute entity throughout the station (eg. a fuel assembly may be moved from the Build Area to the Reactor via the Fuelling Machine; a fuel or component box may be moved from one store to another).

FUNCTION 9: ENQUIRIES

This function handles all reports and enquiries that are available via the menu. The Euratom reporting is included within this function. A draft ICR, MBR, or PIL report may be produced prior to the required date. This facility means that the report may be checked, the database modified if required, and then the actual report may be produced on the required date. The database may also be interrogated at any time using procedures written by the users in a high level query language. These are not incorporated into the menu system.

9. SUMMARY

The Computerised Fuel Records System as outlined above, has now been in use at Torness Power Station for five years and has been found to be invaluable in enabling the Station to comply with all Mandatory and Statutory requirements relating to the control of Fissile Material within the site.

It has been found that with the use of this computerised system the Fuel Records Team can operate more efficiently and with only one third the staff required to run a comparable non-computerised system.

It is our opinion that this system could, with various levels of modification, be used to perform the same controls at any other Nuclear Power Station.

PEPSICODE - A COMPUTER CODE FOR THE DYNAMIC SIMULATION OF SNM HOLD-UP IN MIXER-SETTLERS

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Abstract

Hold-up of Special Nuclear Material in processing facilities is an insidious problem from both safety and safeguards point of view.

Large inventory differences (IDs) are often proven to be caused by wrong estimates of hold-up, consequently, for a more efficient SNM accounting system, a new awareness about the problem of hold-up estimation has been created.

In order to simulate the dynamic behaviour of mixer-settlers in an extraction solvent process, ENEA, in cooperation with the University of Rome, developed a Purex Extraction Process SImulation Code (PEPSICODE).

PEPSICODE, due to a peculiar hydrodynamic modelling, describes time evolution of U, Pu and fission products concentrations both for transient and steady-state conditions.

Simulation of an extraction campaign of EUREX plant will be reported.

1. Introduction

Within the next years, three new commercial reprocessing plants will start the operation with a total capacity of about 2800 t HM/y. That will means, for each plant, a total solvent extraction system inventory of 10÷20 Kg Pu.

With enhanced material control and accounting practices and procedures, inventory differences and, consequently, the problem of quantifying residual process material (hold-up) have been enphasized.

Then, the importance of knowing uranium and plutonium concentrations vs. time in process equipment is become a more and more felt requirement in process control and Near Real Time accounting procedures for nuclear material.

In a PUREX extraction process, mixer-settlers contactors are characterized by relatively low throughputs and large hold-up volumes.

Measuring hold-up dynamic behaviour is an extreme difficult topic, it is more convenient an estimation by studying and developing mathematical models of the hydrodynamic characteristics of the contactors.

In the framework of the ENEA activities relevant to the Safeguards implementation in reprocessing plants, a general computer program simulating the process section and the measuring system of the EUREX pilot reprocessing plant (Fig.1 and Tab.1) has been developed to evaluate the performances of near real time accountancy procedures /1, 2, 3/. With the aim to implement a subroutine for the hold-up estimation a Purex Extraction Process SImulation Code (PEPSICODE) has been developed by the University of Rome "La Sapienza" and ENEA.

PEPSICODE provides time varying estimates of the SNM hold-up residing in a mixer-settler from the start-up until the steady state is achieved.

It can be very useful in studying also process shutdown and transient conditions including problems of sensitivity and stability.

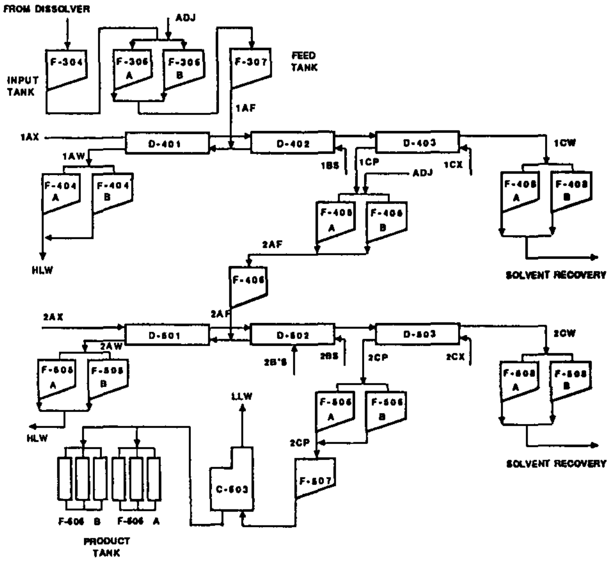

Fig.1 - Scheme of the simulated Eurex process section, /1/.

Comp.	V (l)	Description	Pu (g/l)
F-307	360	Feed 1°Cycle	2.5
F-404	180	Aqueous waste 1°C	$2\ 10^{-4}$
F-408	350	Organic waste 1°C	$1\ 10^{-5}$
F-405	35	Product adj. 1°C	0.41
F-406	35	Feed 2°C	0.41
F-505	100	Aqueous waste 2°C	$2\ 10^{-5}$
F-508	350	Organic output 2°C	-
F-506	40	Product 2°C	2.7
F-507	40	Feed Evaporator	2.7
C-603	14	Evaporator	100
F-606	125	Output	100
D-401	54.4	Codecontamination battery 1°Cycle (8 st.)	
D-402	68	Scrub battery 1°Cycle (10 st.)	
D-403	68	Strip battery 1°Cycle (10 st.)	
D-501	74.8	Codecontamination battery 2°Cycle (11 st.)	
D-502	74.8	Coscrub battery 2°Cycle (11 st.)	
D-503	108.8	Partial partition battery 2°Cycle (16 st.)	

Tab.1 - In process hold-up in tanks and extraction equipments of Eurex plant, /1/.

2. The hydrodynamic modelling

The hydrodynamic model implemented within PEPSICODE derives from experimental studies carried out by means of a transparent liquid-liquid contactor with coloured phases /4/.

Mixer

Within the mixer the flow of the two mixed phases is particularly well approximated by a plug-flow. In this condition residence time is obviously given by the ratio between volume and total flow Vmix/(A+O), (Fig.2).

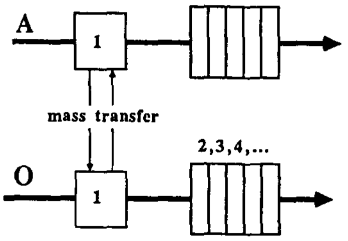

Fig.2 - Mixer model.

Settler

In the settling zone plug-flow hypothesis is reasonably acceptable only for organic phase, as a consequence its residence time is given by the ratio between the volume of organic zone and the flow of organic phase.

Aqueous phase shows a noticeable shift from plug-flow. Aqueous zone within the settler is modelled by six perfectly mixing theoretical vessels, connected in series with a recirculation flow R between the first and the last one. This model allows simulation of aqueous flow A which, in small quantity, may invert global flow from input to output. This is accounted for by the recirculation flow R. The six theoretical vessels have different volumes, ranging from 2/20 to 5/20 of the aqueous phase volume. The output of the settler is placed in the fourth vessel (Fig.3).

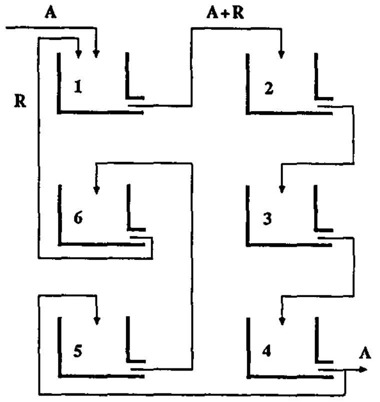

Fig.3 - Settler model.

3. The extraction process

The mathematical model of the extraction process is based on the following equations /5/:

$$V_a(X_i - X_{i_{in}}) = V_o(Y_i - Y_{i_{in}})$$

$$Y_i = \text{Eff}\, D_i X_i + (1 - \text{Eff})\, Y_{i_{in}}$$

$$D_i = K_i X_4^{p_i}\left[Y_{TBP} - \sum_{i=1}^{4} Y_i\right]^{q_i}$$

where:
i=1 indicates uranium, 2 for plutonium, 3 for fission products and 4 for HNO_3.

V_a, V_o - Aqueous and organic volumes
X, Y - concentrations in aqueous and organic phase
Eff - Murphy efficiency
D_i - distribution coefficients
p_i, q_i - coordination factors
K_i - equilibrium constants

This constitute a set of 12 equations that solved for time steps gives near continuous evolution of the concentrations in organic and aqueous phases.

The in-process inventory for any contactor is influenced by many parameters, namely:

- acid and nuclear material concentrations in the streams entering the contactor;
- organic TBP concentration;
- U(IV) reductant concentration in the feed streams;
- flow rates of all streams entering the contactor;
- number and volume of the stages;
- phase volume ratios.

In order to characterize more completely the mathematical model, input flows within PEPSICODE have not been considered fixed nominal values but random values originated from a random sampling process on a gaussian density function with nominal values as centroid. In the next future, also the other parameters will be described with stochastic distributions.

4. Description of the Code

PEPSICODE is written in FORTRAN 77 and optimized to run on PC MS-DOS compatible. It simulates dynamic pattern of a solvent extraction process of uranium and plutonium carried out by means of a battery of pump-mix mixer-settlers.

Default configuration of the stages number of the battery is 15 for scrubbing and 16 for extraction. Default configuration may be easily changed by the code user. Chemical simulation is carried out on the basis of conventional theorethical models and takes into account five components: uranium, plutonium, fission products as whole, nitric acid and TBP.

The code gives as output the time pattern of the uranium, plutonium, fission products hold-up inside the battery for the transitory from the start-up to steady state, further also solute concentrations in output flows are given.

5. Conclusions

By means of PEPSICODE an extraction campaign performed at EUREX pilot reprocessing plant has been simulated, data obtained show a good approximation with real ones. Parametric studies on the influence of the recirculation flow in the settling zone show hold-up grows up with this flow, causing slowing down of process dynamics.

Fig.4 shows the plutonium hold-up in the extraction battery of the first cycle with different recirculation flow. The input data are listed in Tab.2.

Simulation experiments using the model described in this paper are currently underway. The sensitivity of NRTMA under more realistic operating conditions will be tested (effects of process noise, inventory fluctuations, changes in burnup of processed fuel, deviations from equilibrium plant operation etc...).

In parallel, investigations on the validity of the simulation model will continue.

N° of extraction stages	8
N° of scrubbing stages	10
Mixer volume	2.5 l
Settler volume	4.3 l
Feed flow rate	0.0006138 l/s
Scrub flow rate (10th stage)	0.0004166 l/s
Scrub flow rate (1st+9th stage)	0 l/s
Organic flow rate	0.0016666 l/s
TBP concentration	1.09 moles/l
U concentration (feed)	1.0504 moles/l
Pu concentration (feed)	0.0105 moles/l
HNO3 concentration (feed)	2 moles/l
HNO3 concentration (scrub)	3 moles/l

Tab.2 - Input data for the preliminary tests.

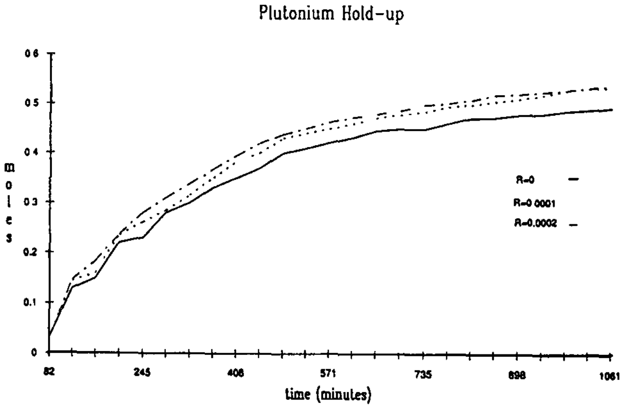

Fig.4 - Plutonium hold-up in the first extraction cycle vs. time with different recirculation flows.

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THE EURATOM INFORMATICS ARCHITECTURE

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Abstract

Open systems and standards in a multiproduct environment are the EURATOM guidelines. Consequently, the O.S.I. model, UNIX (POSIX) and X/OPEN specifications determine the EURATOM informatic strategy.

The major objectives are the development of secured telecommunications, the migration to open systems and the integration of data processing from measurements in the plants to accountancy at the headquarters.

1. Introduction

The purpose of this paper is

(i) to demonstrate to new inspectors the main information flows and verifications at nuclear installations and at Euratom headquarters;

(ii) to inform operators about the informatics strategy used by Euratom in order to improve information transfer between the nuclear installations and Euratom headquarters.

Chapters 2 to 4 discuss the information flows and verifications, the computer architecture and the informatics strategy of the Commission of the European Communities. Chapter 5 explains the Euratom objectives based on this architecture and strategy.

2. The main Information Flows and Verifications

To simplify the understanding of the different information flows, Fig. 1 shows the main information flows relevant to Euratom Safeguards.

There are 2 types of lines, indicating

(i) the information flows (thin lines);

(ii) major verifications carried out by Euratom (thick lines).

2.1. Flow of information (thin lines)

1. The operator performs measurements in order to improve his knowledge of the physical reality in his plant.
2. Euratom performs its own measurements (mainly non-destructive analysis) in order to verify:
   - the material flow in critical steps in the industrial processes (verification A);
   - the operating records (verification B);
   - the List of Inventory Items (LII) given by the operator (information flow 8) when a physical inventory is done (verifications C and D).

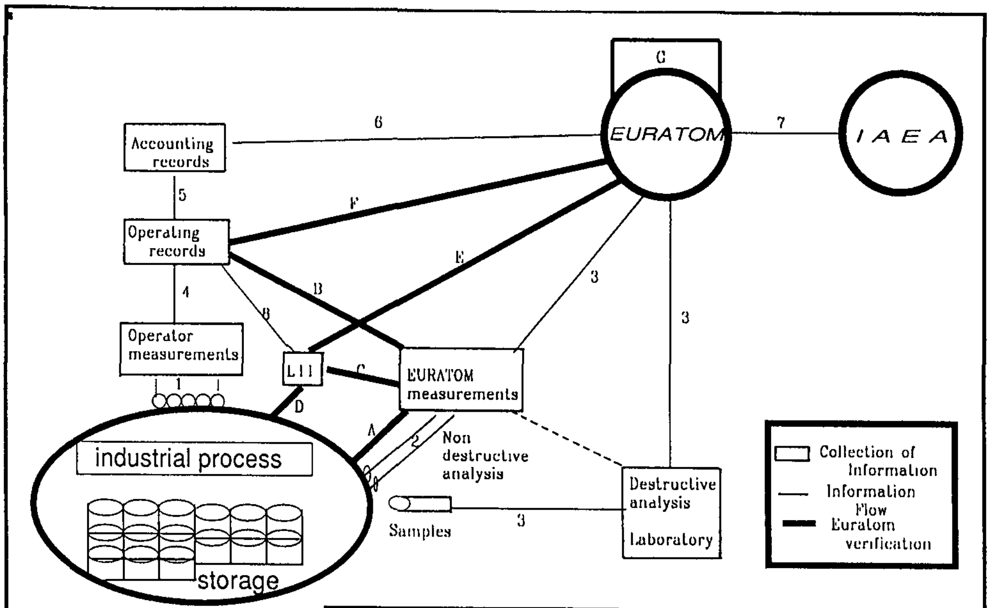

Fig. 1 : Main Euratom safeguards information flows and verifications.

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Typical items to be measured are: tank volume, weight, temperature, density, neutron/gamma spectra, images produced by a containment/surveillance camera etc.
The equipment used to collect the measurement results are automatic nuclear material flow measurement stations, logging devices, verification and transmission systems and others. The majority of these instruments are portable or at least transportable. MS-DOS PCs are used daily to collect and evaluate the data. Great effort is made to authenticate the appropriate functioning of the in-plant instruments (Ref.). The inspector can verify the quality of the data by comparing information coming from different steps of the plant or processes or by checking the consistency of data generated by different operations.

3. To improve the measurement precision, samples are analysed in laboratories. Euratom will install on-site laboratories in some major plants to have the analysis results available as soon as possible.
4. The measurement results of the operator are reflected in the operating records.
5. The accounting records of the operator are generated from the operating records. In the large plants, Euratom has installed computers to support the inspectors in verifying measurements, operating records, accounting records and in preparing the physical inventory verifications.
6. On a monthly basis, the Inventory Change Reports are sent to Euratom either directly or via an enterprise headquarters or a national organisation. After each physical inventory taking, the Material Balance Report and the Physical Inventory Listing are transmitted to Euratom (Ref. 2). 600.000 entry lines were reported in 1990 to Euratom by the operators, 74% of which were in computer-readable form.
7. With the same monthly frequency, Euratom sends the reports to IAEA (Ref. 3).
8. The List of Inventory Items (LII) is produced for a full or partial Physical Inventory Verification. This listing is later compared with the Physical Inventory Listing received at Euratom headquarters.

2.2. The verifications: (thick lines)

A. Safeguards relevant critical process steps are verified using mainly non-destructive measurements.

B. The operating records are validated with these measurements.

C. Items on the LII are verified by non-destructive or destructive analysis.

D. Based on the LII established by the operator, the physical inventory is verified.

E. The LII used for the physical inventory verification is compared with the Physical Inventory Listing received at Euratom headquarters.

F. The operating records help the inspector to validate the Inventory Change Reports received at Euratom headquarters.

G. Accountancy verifications are performed at headquarters in order to ensure the quality of the reports.

3. Hardware and Telecom Architecture

3.1. The first computerized nuclear materials accountancy system of the Euratom Safeguards Directorate became operational in 1962 after the creation of Euratom Safeguards in 1959. A second system was established before 1969. The present system became operational in 1977. After a general change in the informatics policy of the Commission of the European Communities based on the principle of distributed information processing, the Euratom Safeguards Directorate got its own computer in 1982.

3.2. The present hardware and telecom architecture at Euratom headquarters is described in Figs. 2 and 3. Fig. 2 shows the internal network, Fig. 3 the external network. These two networks are completely independent for security reasons.

3.2.1. The internal network (Fig. 2) is based on 2 Ethernet Local Area Networks (LANs) installed in 2 separate buildings. These 2 LANs are interconnected via a telephone line. All information passing through this line are ciphered automatically. Many different computers are connected to this internal network:

a. A Siemens mainframe with the BS2000 operating system. It includes:
   - a 7560-EX CPU of 1.7 MIPS and 16 MBytes real memory;
   - 4100 MBytes of disk space;
   - 2 magnetic tape units of 1600 and 6250 bpi each;
   - 2 line printers (600 lines per minutes);
   - 32 Siemens 9750 terminals.
b. A Unix Olivetti 3B2/700 of 7 Mips, 900 Mbytes of disk space and 16 Mbytes of real memory. 27 WYSE VT220 terminals are connected to this computer.
c. A UNIX NCR 600 of 1.2 Mips, 120 Mbytes of disk space and 8 Mbytes of real memory. 10 WYSE VT220 terminals are connected to this computer.
d. 70 MS-DOS PC are connected to the LAN. They can access not only the UNIX computers but also the Siemens BS2000 system.

e. 15 laser printers are connected to the UNIX computers.

The main software packages used by Euratom are:

- for wordprocessing: Q-Office on UNIX and MS-WORD on MS-DOS;
- for spreadsheets: MULTIPLAN or LOTUS 123;
- data base management systems: DBASE III+ or Clipper on MS-DOS; Oracle on MS-DOS and UNIX; ADABAS on Siemens BS2000;
- for telecommunication: TCP-IP with NFS or FTP on the LAN; Easy Access on the MS-DOS PCs to access the UNIX computers or the BS2000 computer in emulation mode;
- for electronic mail: Insem Local Server (an in-house package).

3.2.2. The external network (Fig. 3) is composed of the following equipments:

a. To exchange files or messages, a UNIX computer (NCR 600) is connected to the public packet switching data network based on the X25 standard. The confidential data is encrypted by a software package using the Data Encryption Standard (D.E.S) algorithm (Ref. 4). Presently the protocol used to ensure file transfer is KERMIT (soon X400 or FTAM).

b. To exchange telexes a software package is used. 3 terminals and 2 laser printers are connected to the telex-transmission system.

c. To exchange facsimiles Euratom uses 2 fax machines equipped with a cipher unit (OMNISEC 519B).

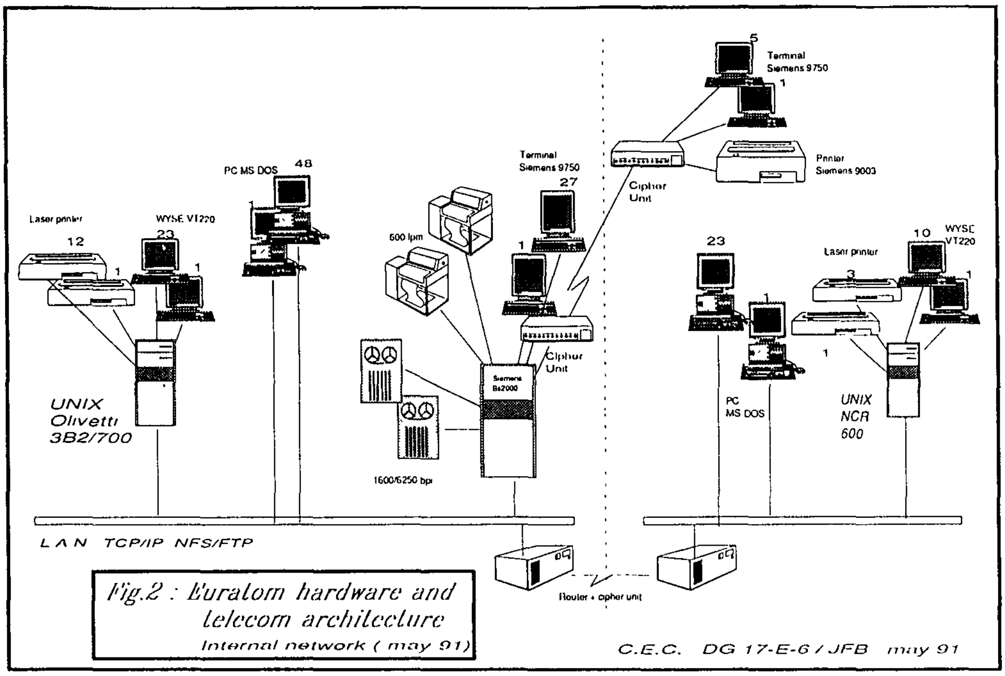

Fig.2 : Euratom hardware and telecom architecture
Internal network (may 91)

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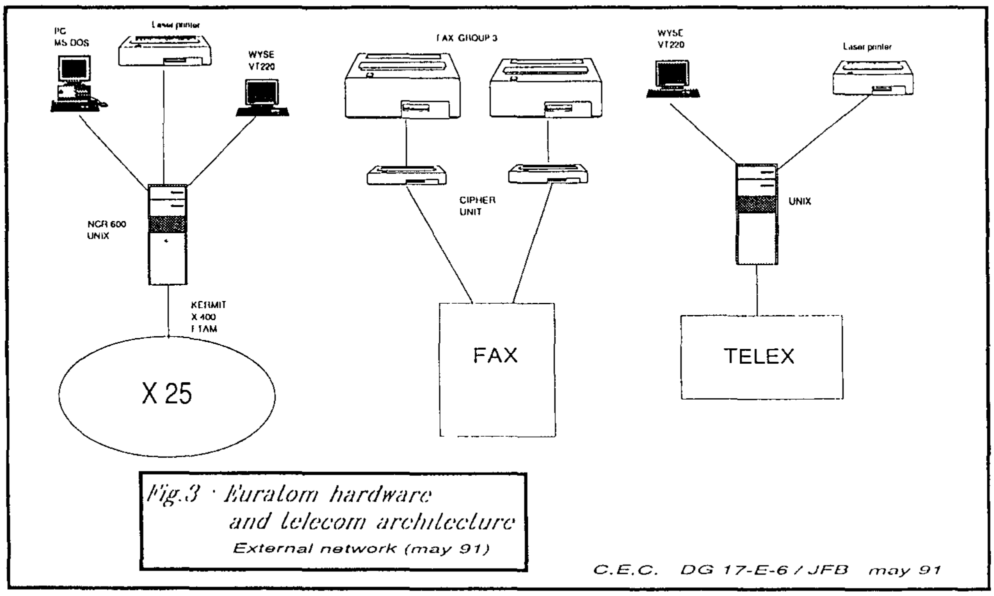

Fig.3 · Euratom hardware and telecom architecture
External network (may 91)

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3.2.3. In big nuclear installations, Euratom has installed computers to support the inspectors in their activities. Today, 8 nuclear installations are equipped with MS-DOS PCs. Moreover, 12 portable PCs are at the disposal of the inspectors. In the near future, UNIX computers will replace some MS-DOS PCs.

4. The New Informatics Strategy

Open systems and standards in a multi-product environment is the basic rule for all Commission's services and consequently also for the Euratom Safeguards Directorate.

This rule is enshrined in guidelines published by the Commission which thinks that this basic rule is relevant not only to public administrations but also to the private sector.

The most recent version of this rule is contained in the "Guidelines for an informatics architecture" (Ref. 5).

The main principles are briefly presented here to simplify the understanding and to facilite the integration and exchange of data with operators of nuclear installations.

The spirit of these guidelines is:

"Most architectures have been designed by the computer manufacturers to match their present and future product ranges. Such architectures are, generally speaking, mutually incompatible even where standards are used. A vendor-independent architecture can only be developed by the user organisation being the customer of the information technology (IT) industry - but no single customer has the power to impose a given architectural design on industry. Consequently, such an architecture must emerge from the ongoing process of supply and demand."

"Open systems are those systems and components which can be specified and multi-sourced in a competitive market place. An Open system specification must not be owned by any single vendor and must be freely available."

The switch to open systems procurement policies based on standards is an obligation for public administrations in the European Community (Council Decision 87/95/EEC of December 1986)."

It is in the interest of all users to remain free to choose the best way to integrate new technology independently of the policy of individual vendors." (Ref. 5).

A vendor-independent strategy based on open systems

(i) is economical because standards reduce complexity and training costs and because competition brings prices down;

(ii) necessary to communicate between organisations.

"Migration to open systems should be planned as a series of steps congruent with the life cycle of equipment and application in order to avoid costly conversions."

Consequently the main standards followed presently by the Commission are:

MS-DOS
UNIX V Rel 4
SCO UNIX OPEN DESKTOP
C, Cobol
SQL
X-WINDOWS
MOTIF

The Open Systems Interconnection (OSI) model has been the basis for the connectivity standards since 1980.

For portability, the UNIX operating system is the de facto standard with increasing acceptance, but the UNIX based Portable Operating System Interface (POSIX) reached the de jure status in 1989.

X/OPEN specifications for a Common Application Environment supported by a sufficiently large number of computer and software vendors have been a great help in bridging the gap.

For the external communication, the Commission follows the standards presented in Fig. 4.

<table>
<tr><th></th><th colspan="2">Terminal Access</th><th>File transf</th><th colspan="4">Message Handling</th></tr>
<tr><td>7. Application</td><td rowspan="4">VIDEOTEX</td><td rowspan="4">X3
X28
X29</td><td rowspan="3">FTAM</td><td rowspan="3">MHS</td><td rowspan="4">TTX</td><td rowspan="7">TELEX</td><td rowspan="7">FAX</td></tr>
<tr><td>6. Presentation</td></tr>
<tr><td>5 Session</td></tr>
<tr><td>4. Transport</td><td>TPO</td><td>TPO</td></tr>
<tr><td>3 Network</td><td colspan="5">X25</td></tr>
<tr><td>2. Data link</td><td colspan="5">HDLC</td></tr>
<tr><td>1 Physical</td><td colspan="5">X21-Bis</td></tr>
</table>

Fig. 4 : O.S.I. model for external telecommunication

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5. Objectives for the Next Five Years

The three main objectives which will influence the informatics development for the next 5 years are:

(i) telecommunication;
(ii) integration of in-field and headquarters applications;
(iii) migration to Open systems.

5.1. Telecommunication

a. Efforts to decrease the number of accounting reports received on hard copy must continue (see information flow 6 in Fig. 1). In 1990, in the mean, 26% of the accounting reports had to be keyed in manually. For the first 3 months of 1991, this percentage could be brought down to 10%. The objective for 1995 is 1%.

The reasons for changing from manual to automatic data entry are two-fold:
(i) to minimize human errors and
(ii) to speed up the availability of data.

The transmission medium which is mainly used to date is the floppy disk (64% of the reports transmitted in computer- readable form). To date, in the mean, 80.000 entry lines are input every month into the Euratom system.

b. The replacement of magnetic tapes and floppy disks by secure telecommunication is an important objective.
   - Secure telecommunication: three principal caracteristics for secure telecommunication must be respected which are implemented by hardware and software components:
     (i) the sender must be authenticated,
     (ii) the transmitted data cannot be modified without automatic detection of the modifications,
     (iii) the transmitted data should only be comprehensible to the receiver (the data must be ciphered).

     To implement caracteristic (iii), Euratom will use the Data Encryption Standard (D.E.S) algorithm (see Ref. 4).
   - The standards followed for the telecommunications are presented in chapter 4 above. Fig. 5 summarizes the possibilities of exchanging data or messages with Euratom.

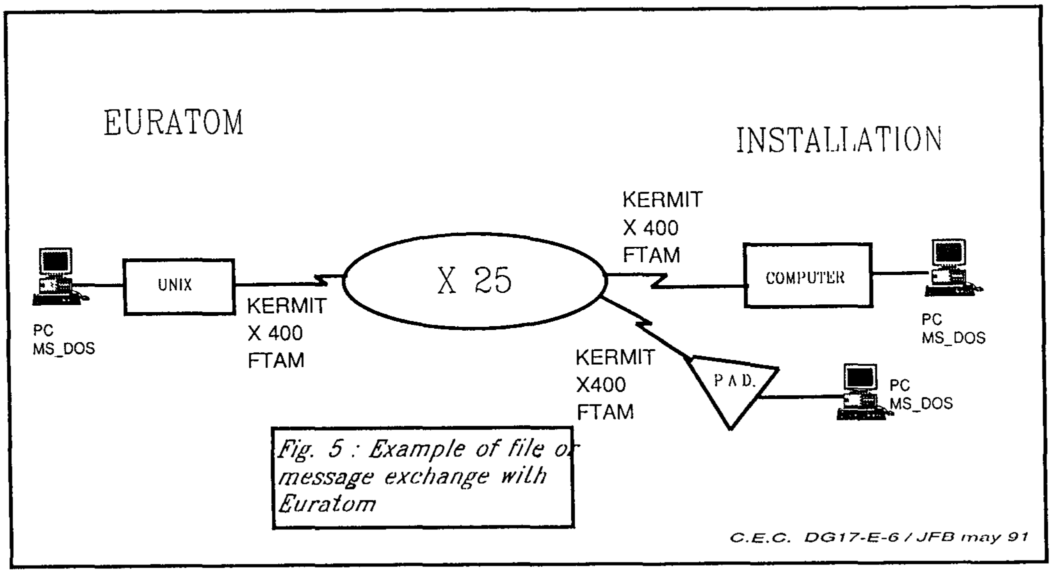

Fig. 5 : Example of file or message exchange with Euratom

In the years 1991 and 1992, X400 will be used to transfer not only messages but also ciphered files. When FTAM becomes available in a production environment, the ciphered files will be transmitted using this protocol.

Great effort will be put into the establishment of a secure telecommunication system with IAEA. This should not only serve for the transmission of the accounting reports, but also for the exchange of messages, inspection planning information, and other regular information exchanges.

5.2. Integration of In-Field and Headquarters applications.

This objective concerns large nuclear installations requiring computer processing support.

a. Confidence in the operating records must be improved by:
   - increase of the frequency and accuracy of verifications;
   - decrease of the delay between the measurements and the verification of the operating records.

b. The in-field software has to be compatible with that used at headquarters to:
   - decrease the costs for the training, maintenance and interventions;
   - standardize the interface with other software used by inspectors and headquarters (graphical user interface, etc.).

c. Euratom intends to develop secure telecommunications between headquarters and Euratom offices in the nuclear installations
   - to exchange newly acquired information;
   - to reduce inspector isolation;
   - to support the inspector in solving computer problems;
   - to avoid the need for the inspector to carry confidential information while travelling.

5.3. Migration to Open systems

Following the strategy developed in chapter 4 above, all the applications running on the Siemens BS2000 have to be converted to run under the UNIX (POSIX) environment. This requires considerable human and financial effort which is not available presently.

The main applications are described in the paper of M. KSCHWENDT. (Ref. 6)

6. Conclusions

Open Systems, secure telecommunication and development of in-field computer applications determine the Euratom informatics strategy for the next years. Close cooperation with the operators and the IAEA is essential to achieve these objectives.

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A METHOD FOR CALCULATION AND REGISTRATION OF NUCLEAR DECAY IN A MOX FUEL FABRICATION PLANT

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Abstract

The calculation and registration of the continuous decrease in quantity of nuclear material due to nuclear transformation becomes rather important in large MOX Fabrication Plants.

This is due not only to the important amounts of plutonium that are processed but also to the relative high ^{241}Pu content of the plutonium which is used in the fabrication process. The decayed quantities are already important over smaller periods e.g. one month and are to be taken into account because they can influence the MUF-evolution and consequently the inspection and verification efforts.

This paper describes how the nuclear transformation is processed at the BELGONUCLEAIRE MOX Fuel Fabrication Plant.

1.Introduction

The physical process leading to nuclear transformations is a continuous one. Consequently the time lapse between two analysis results has to be considered in the accountability system. If this is not done, the decayed quantities will be added to the MUF, with all its consequences.

In large commercial MOX plants these quantities can certainly not be neglected. In the period 1975-1980 a typical ^{241}Pu-content used in the fabrication process was 3.3% with a relative low throughput of about 100 kg per year. If a time period of e.g. 3 months was not taken into account for decay, the contribution to the MUF would be about 40 g Pu per year. At present the new MOX plants have a throughput of 2000 kg Pu per year, while one PI is taken per year. This would mean, with a typical ^{241}Pu content of 9.7% ^{241}Pu of the Pu in process nowadays, a contribution to the MUF of about 2500 g. The situation for the future MOX plants with greater throughputs will even be worse.

As an increase of the throughput does not mean a proportional increase of the MUF, it is clear that the decay has to be excluded from the MUF. At the BELGONUCLEAIRE MOX plant there is not seen any increase of the MUF related to the increased throughput.

BELGONUCLEAIRE developed gradually a real time accountability system, based on our experience, which allows us to calculate and book the decay at any time. It is based on a itemized accountability system. Per item all the necessary data for calculating the decay are registered in a file (Pu-total, isotopic composition, date on which the data are valid ...).

The quantities received from the shipper are booked as declared by the shipper with all data necessary for calculating the decay when needed. In general, the decay is calculated and booked :

- when incoming PuO$_2$ is sampled and analyzed (normally not);
- when the master blend is made;
- when the final blend (secondary blend) is made;
- when scrap is micronized and blended;
- when the rods are updated with the analyses results.

At each step the Pu-quantities of the items (batches) are updated and the decayed quantities are accounted for.

Thanks to the accuracy of our calculations at the various process steps, BN could considerably reduce the number of samples and analyses.

2.Description of the problem

In the period before 1987 several problems were encountered when dealing with Pu quantities registered in the accountability system. Below we list the main problems the safeguards service was confronted with.

- Which was the Pu isotopic composition per item, batch ?
- Which was the validity date for the mentioned Pu amounts ?
- How to update correctly the quantities per item ?
- Updating the Pu quantities was cumbersome even if the isotopic composition and analysis date were known.
- The listings or documents distributed at different periods, mentioned different quantities for the same item, especially when the material had to be stored for several years. This was very annoying when comparing balances.
- An unknown amount of MUF was due to the NT (Nuclear transformation) as it could not always be correctly calculated and booked.
- In order to compare measurement results obtained by the inspector during PIV/SIV (SIV = 4 weekly short inventory verification) and FLOW verifications the operator needed the updated values, otherwise no exact evaluation of the measurement results was possible.

3.Requirements for a correct NT follow-up system

- The exact analysis date, isotopic composition and related Pu amounts have to be known for all the incoming Pu, as well as for the actual inventory.
- Care has to be taken, that from the analysis date of the incoming Pu until the reference date of the rods leaving the facility, no time period is not taken into account for calculating the NT, notwithstanding the various

crossblendings and material flows in the plant (Fig.1).
- Certain process steps have to be defined, where NT must be calculated and accounted for.
- The system has to be computerised as far as possible to exclude human transcription errors. That means e.g. that for a blending all calculations, file updating, inventory change report, etc. no operator has to intervene.
- Per item, the validity date and related Pu-amount and composition has to be known exactly.

4.Description of the BELGONUCLEAIRE system for accounting NT. Solution choosen

4.1. General.

When BELGONUCLEAIRE started developing a new accountability program in 1985 one of the aims was to solve the above mentioned problems and certainly avoid the contribution of NT to the MUF and uncertainties when comparing book values with measured values by the inspectorates.

Based on BN experience a system was developed which allows to calculate and book the NT at any time on any item as long as it is registered in the accountability records. It is based on an itemized accountability system.

This was achieved in the following way :

1. an itemized system was developed that allowed BELGONUCLEAIRE to know the exact Pu quantities and isotopic composition per batch and per item on any date, either in the future or in the past. Per item at least the following data are registered in different files : Pu-total, isotopic composition, analysis or calculated date.
2. at certain intervals, process steps; e.g. when blending several batches/items, the decay is calculated and accounted for until the blending date and the new values are registered in a file. In this way BN avoids that the NT has an influence on the MUF.
3. listings can be made, at any time, with calculated values on the date wanted, e.g. in case of non-destructive measurements by the inspectorate listings are made which give the Pu amounts on the measurement date, without changing the accountability records.

4.2.Process steps where NT is computed and accounted for.

The following internal rules are applied (Fig. 1) :

4.2.1.On receipt of PuO_2

No decay is booked. The data as declared by the shipper, are used for accounting purposes.

The items remain in the store with the original shipper values (Pu-total + corresponding isotopic data and analysis date). The isotopic composition is registered by batch (BN-lot).

This gives the possibility to calculate at any time the actual isotopic composition.

Exception : if an analysis is done by BELGONUCLEAIRE on the incoming PuO_2, a NT is accounted for, for the period between the analysis date of the shipper and the new analysis date of BELGONUCLEAIRE. The difference between the newly calculated values and the analysis result will be declared as Shipper-Receiver-Difference (DI). For fresh PuO_2 powder, there can be a time lapse up to several years between the analysis date, after reprocessing, until the last can is used at BELGONUCLEAIRE for the master blend. This means that the NT-amount can be considerable.

4.2.2.On master blends. This means that fresh PuO_2 powder + UO_2 + scrap is micronized and blended in the process. In order to obtain the same isotopic composition for several blends, cans from different batches are used (= crossblending).

As the accountability system knows the Pu-content + corresponding isotopic composition + analysis date for each can, it is easy to calculate the NT and to update the accountability files for the blended quantities. The NT is automatically calculated until the blending date and registered in the files by the computerprogram that is used for processing the blending in the accountability files.

4.2.3.On secondary blends. These blends are made of master blend powder, diluted with UO_2 + scrap to obtain a specified ratio Pu/(U+Pu) for MOX fuel.

At the moment the blend is registered in the accountability system, the decay is automatically calculated and accounted for based on the registered values of the cans (see master and scrap blend), until the date of blending.

The various conversion factors (U+Pu/ox, U/Pu+U, ...) are calculated by the same computerprogram. These factors will be used for transfers later on.

4.2.4.On scrap blends. From time to time the scrap is collected, micronized and blended. The same procedure is followed as for the secondary blend. The decay is accounted for from the various secondary blend dates until the scrap blend date.

4.2.5.On rods. The provisional values with which the rods are created after welding, will be updated for decay starting from the blending date until the reference date, as soon as the final figures per rod are available and introduced in the accountability files. The final values per rod are calculated on a reference date (this date is usually the date of loading into the reactor) valid for a campaign or subcampaign. This reference date is imposed by the client.

4.2.6.On shipments other than rods/assemblies
- shipments of small quantities : the values on the blending date are used; no adjustment is made for the decay between blending date and shipment date;
- shipments of waste quantities : idem;
- shipments of PuO_2 cans : no general rule is

applied; case by case, it is decided whether the decay has to be accounted for by BN, prior to shipment, until the shipment date.

5. Half-lives values used

^{238}Pu	: 87,74	years
^{239}Pu	: 24119	years
^{240}Pu	: 6540	years
^{241}Pu	: 14,4	years
^{242}Pu	: 376000	years
^{241}Am	: 432,6	years

These values are agreed upon by EURATOM and IAEA(1).

6. Conclusion

Inventory changes like NT are rather complex in a MOX fabrication plant. The aim of this paper was to explain how this problem was solved at BN and why it was done in this way.

It was shown how the time lapse between the analysis date at the reprocessing plant and the reference date imposed by the reactor operator is completely covered.

This way of operating guarantees that :

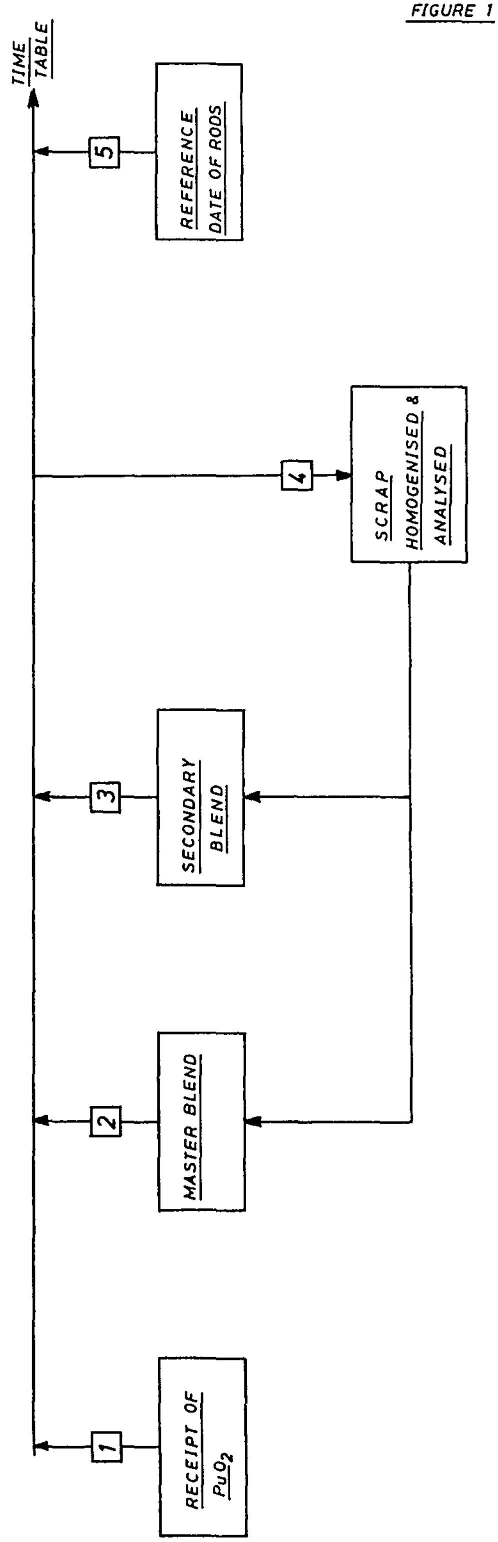
1. all Pu-decay is accounted for correctly and easy to verify;
2. the MUF is not influenced by decayed Pu amounts;
3. the obtained measurement results during PIV/SIV and FLOW inspection by the inspectorate can be perfectly compared with the values declared by the operator.
4. excludes that for the same item different values come into circulation;
5. for any item, at any time, the operator can reproduce the actual value, updated for decay.

7. References

/1/ Publication of December 1983 INDC (NDS) - 149/NE with "Proposed Recommended List of Heavy Element Radionuclide Decay Data" by the International Atomic Energy Agency.

Main flowstream for NT accounting at Belgonucleaire

FIGURE 1



COMPUTER ASSISTED HUMAN MATCHING,
A SOFTWARE TOOL FOR INTERNATIONAL TRANSIT MATCHING

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Abstract

One of the responsabilities of the IAEA is to confirm the receipt of international shipments of nuclear material by matching shipments reported by one Member State against receipts reported by another Member State. When sufficient information is available, this matching of receipts against shipments is straight forward. However, because of non-uniform reporting practices in different countries a significant number of reports must be investigated manually. In these cases the matching reports are often hidden among a large number of other transactions. To identify matchings involves considerable work in studying lists of declarations and using expert judgement. An expert system, Computer Assisted Human Matching (CAHM) has been designed and implemented to reduce substantially the manpower required in the Human Transit Matching phase.

This new system is completely integrated within the IAEA Safeguards Information System (ISIS) and since January 1989 has completely replaced the previous manual matching system. The current implementation of the system covers the area of international transfers and domestic transfers as well.

The CAHM system:
- extracts data (un-matched records) from the accounting file,
- generates a new data structure,
- activates the automatic matching algorithm,
- displays the results for human acceptance,
- updates the matched records in the accounting file.

1. Introduction

The CAHM software package was developed to help achieve the safeguards goal of verifying the information contained in the reports of State concerning international and domestic shipments. The term "transit matching" is used for procedures performed in the process of identifying which "receivers reports match shippers reports" that nuclear material has been transferred to another State when the reports are received at IAEA headquarters in Vienna, they are loaded into the ISIS database. These data are then checked for validity and consistency with relevant requirements. After this quality control process, an interrogation is made to find a corresponding report from the country from which it has been indicated the shipment was made. When such a correspondence is found, the shipment and the receiver reports are marked as "Machine matched". From the data, it can be observed that on average about 75% of all shipment records must be manually treated in order to achieve a matched status. This process is called the Human Transit Matching phase.

The final aim of this system is to reduce considerably the manpower required in the Human Transit Matching phase, trying to replace the knowledge of human experts with a program which simulates their behaviour while at the same time allowing human experts to validate the results of the system. The system under examination does not pretend to replace human activity completely throughout the process of searching and validating shipper and receive records (Human Matching phase), but to reduce it considerably, so that the human expert is only involved in the "Suggested Matched Group" validation phase. This paper is divided into two parts. In the first part, section 2 gives an overview of the expert system techniques from the methodological point of view. In the second part, section 3 presents the development of the system from the first analysis and design to the last version used at present by the IAEA in Vienna, while section 4 describes the expert system which allows the creation of the associations (Matching) between shipment and receiver records and section 5 analyses the results obtained using CAHM and considers the future development of the system.

2. Overview of Expert System Techniques

The Expert System is a computer program which uses an intensive collection of heuristic knowledge based on experience, observation or measurement to solve a specific problem, usually through a process of inductive inference. The knowledge is unlikely to be derived from scientifically provable theorems, but may be derived by analogy with similar experiences or by iteration from an initial estimate. Many knowledge-based systems have concentrated on simulating the activities of a human expert who has a long record of achievement as well as a thorough educational training in his field of expertise. As a result, the programs have come to be known as Expert Systems /1,2/.

The Expert System has evolved through practical experimentation rather than theoretical research. In early work the knowledge was embedded in the program. As a result problem solving was efficient but the systems were useful only for one task and were difficult to update and maintain by staff not participating in the system design. They were unpopular with experts as a result of the rudimentary user interfaces and reasoning procedures that were seen to be clumsy and inappropriate.

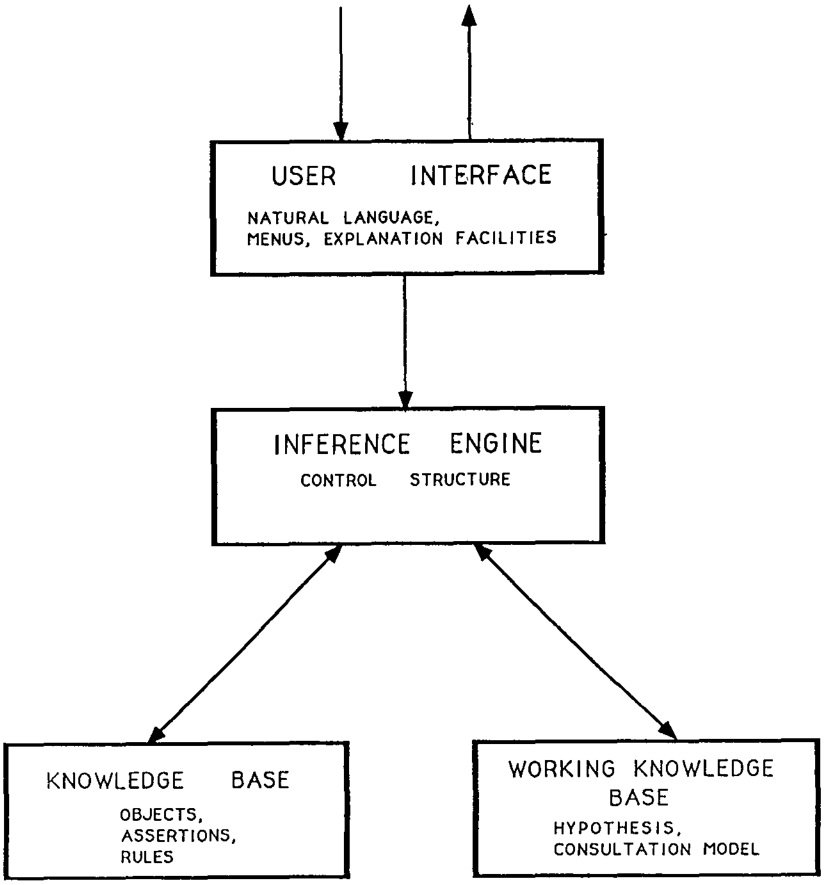
Recently a modular approach based on the three distinct elements shown in Fig. 1 has been adopted. The elements are:
- the inference engine,
- the knowledge base,
- the user interface.

Using the modular approach the system control is independent of the knowledge and the user interface. The program is less sensitive to a particular application and it is feasible to develop commercial software packages for a limited range of applications which can be filled with alternative collections of knowledge. This type of commercial software is known as an expert system shell.

The Knowledge Base.

The knowledge base is a collection of information which contains the following:
- the program variables which represent conceptual units of information,
- information in the form of assertions about the program variables,
- heuristic information in the form of conditional rules which describe relationships between program variables.

The form in which information is stored in the knowledge base is known as the method of knowledge representation. Knowledge representation can affect the conflicting requirements of extensibility of the knowledge base and search



SIMPLE SCHEMA OF THE EXPERT SYSTEM

Fig. 1

efficiency. The principal methods for knowledge representation are either as a semantic network or as a list of production rules. In the semantic network form of representation the conditional relationships are compiled into the computer memory from an explicit graphical representation of conceptual relationships. The search efficiency of this type of representation is good, but the system is difficult to maintain and concepts cannot be evaluated according to a number of different criteria. In the production system, rules of thumb can be represented easily, making modification simple; this factor has contributed to the popularity of production systems. The major drawbacks are that system performance is often dependent on the ordering of the rules and can require extensive search procedures. Moreover, unexpected side effects can be experienced during inference.

The knowledge base is filled during a process known as knowledge acquisition. Knowledge acquisition is often carried out by a "knowledge engineer", who is a computer scientist conversant with the expert's knowledge representation methods, working together with an expert. Their joint task is to translate the expert's knowledge into the format required by the software system.

The Inference Engine.

The inference engine is the system which carries out logical inference by processing information in a computationally efficient fashion. Its task is to select the conditional information, or rule, which is most likely to lead to a solution from the knowledge base and apply it to a working data base to produce a modification to that data base. The working data base contains the information that is known or inferred about a specific problem. The process may be iterative so that several rules are chained together producing many intermediate states in the working data base.

Inferential problem solving requires that it be possible to discriminate between alternative solutions, or courses of action. The simplest method is to decide between alternatives based on a two-valued logic. This type of system can work well although it has been found that human experts are reluctant to describe their knowledge in a deterministic fashion. Instead, various methods of approximate reasoning have been proposed and implemented. Traditionally approximate reasoning has been based on Bayesian statistics which works well for independent and well-defined data, but is less satisfactory for vague and interacting propositions.

The User Interface.

The user interface has two main functions; to provide a vehicle for knowledge acquisition and testing, and to provide a consultant-like dialogue during problem solving.

The user interface can have a major influence on acceptance of the Expert System. Whilst performance of the Expert System can be analysed for particular case studies, it is not possible to device a test methodology that can demonstrate complete system reliability. Instead, the user interface can increase user confidence by providing a detailed explanation and interrogation facility through which the inference procedures can be monitored and assessed. Explanation and interrogation facilities are based on the chain of inference rules. Normally, a temporary record of the reasoning chain is maintained and the inference paths can be displayed to the operator, who can judge whether the intermediate steps are reasonable. The textual content of the rules can be used to display simple explanatory information.

3. Design Evolution

The CAHM system as a whole is made up of a suite of programs which, starting from the raw data which are normally found on the ICR-File, (Inventory Change Report ADABAS File 105), extracts all the Shipment and Receiver records, generates a new data structure which is more suited to the phase of selection of country pairs and then, after having selected two or more country pairs, by the use of an expert system, generates a file which contains all the "suggested matches" ready for the validation and update phase /4/.

The development of the CAHM system was anomalous, as compared to the design of a normal software system, because the central nucleus (expert system) was designed first and then all the series of modules which allow the logical connection between the raw data contained in the ICR file and the "Suggested Matches" file was created /2/.

This mode of procedure is justified by the fact that the practical running of the whole system depends exclusively on the quality of the results of the central nucleus (Expert System).

This in the first place we tried to understand the behaviour of a human expert when tackling a problem of matching a certain shipped batch with one or more receivers, or viceversa, i.e. in other words we tried to understand if it was possible to simulate or better codify the mental process of the human expert with a logical series of conditional rules.

This first phase of work was particularly complicated because in a first analysis it seemed that the rules used for the Matching process were contradictory, in other words we could not formalise the problem. Considering that it was impossible to find general rules which would allow formalisation of the reasoning which the expert uses in the active sense, i.e. why "make a certain Match", we tried to use the cognitive process in the passive sense, i.e. try to formalise the rules which determine the reasons why the expert "does not make a certain Match".

Using this approach it was possible to formalise general universally valid rules. The application of this procedure was extremely useful in the first phase of drawing up rules for the expert system.

Once this preliminary analysis was finished a very simple prototype was made which could be used to test the validity of the rules determined with the method described above.

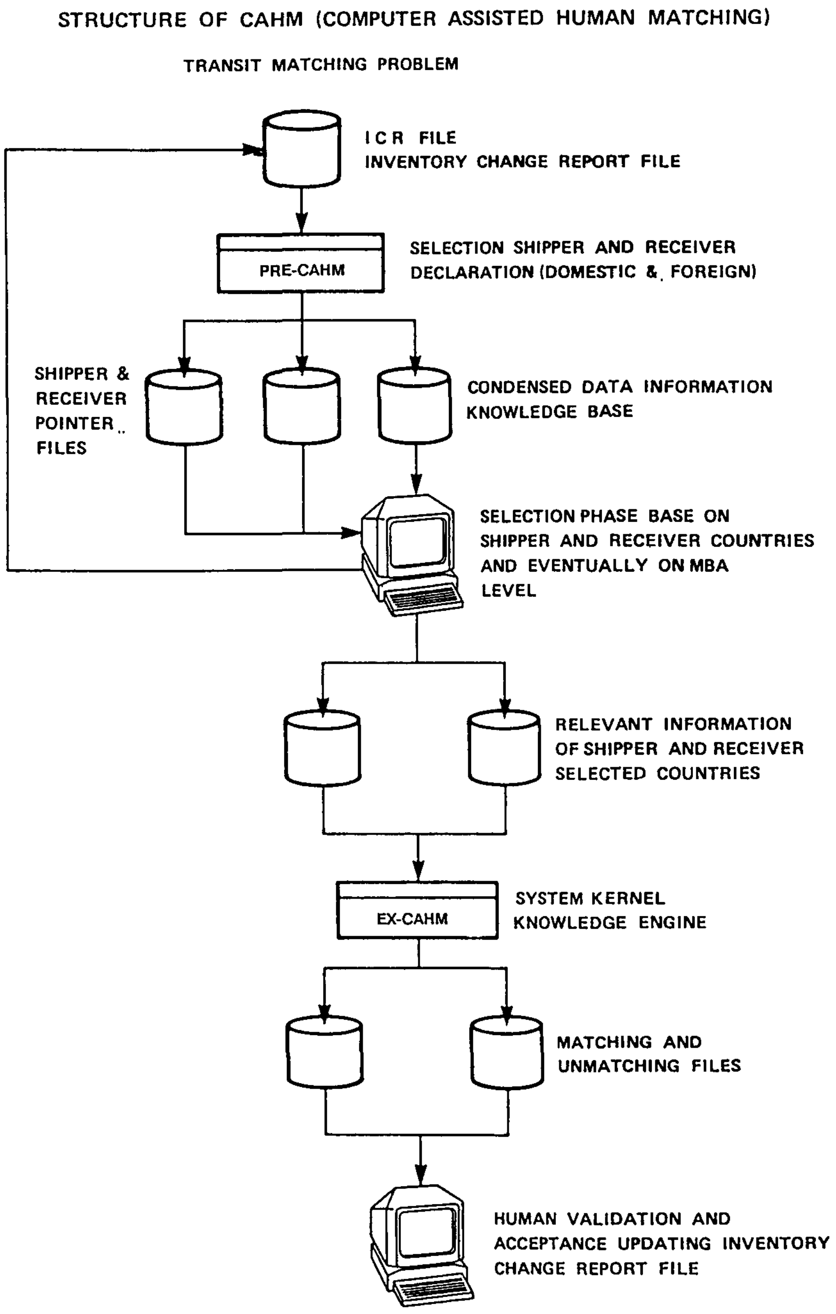


Fig. 2

The results of this first prototype were not very encouraging, or rather the prototype did not create invalid Matches, but the number of valid Matches was very small.

The result of this first experiment should not be considered completely negative because, on analysing the results in detail, it became clear that the number of rules used was too small to give good results. The problem to be resolved was how to determine the best matches from a large class of possible Matches. To determine this type of rule we analysed the Match results which were available in the ICR file. To do this a set of about 10000 shipment and receiver records, which had already been treated by human experts, was extracted at random from the ICR file and we tried to analyse the Human Matching results trying to extract general rules from this mass of data.

The results of this analysis were very encouraging because we were able to understand the rules most commonly used for the Matching process. Once these new rules were determined we then made a second prototype using all the rules which had been already formalised.

The results obtained were very encouraging because not only did the system manage to make most of the Matches but in some cases it gave the expert new combinations which had not been previously considered. The experience acquired in the making of the second prototype was fundamental in producing the final system because not only could we make a software system which was capable of formalising the cognitive processes of the human expert, but also, through the use of practical cases, we understood how to proceed to acquire new knowledge from experts.

In fact because of a normal psychological process the expert is led to give more importance, in his description, to the more complicated Matching case than to the normal cases. This is why in the first analysis the rules seemed contradictory whereas using the analysis of the practical Match results identified the normal cases, i.e. the rules which were valid for most cases. Having available all the necessary rules we then implemented a first version of the "Stand Alone" system /1/, using data sets extracted "ad hoc" from the ICR file. In this first file we extracted unmatched records from the ICR file with a simple sequential reading program, and then the expert system was used to find the possible matches.

The results which the expert system gave were compared with the results obtained by the human experts in their normal working procedure and when necessary some new rules were introduced, or some existing rules were modified to adapt the expert system to a new situations which was not considered.

This expert system "Tuning" phase continued until optimum results were obtained and, having obtained approval from the people in charge of the Safeguards activity of the Vienna Agency to move on to the production phase, we then produced a series of programs which allowed complete integration of CAHM with ISIS (IAEA Safeguards Information System).

4. General System Description

The main aim of the whole CAHM system is to start from a given file of unmatched shipped records and find the corresponding partners in the corresponding file of unmatched receiver records. In other words the system matches the shipped records declared by a certain country with the corresponding receiver records of the receiver country.

The problem, which at first sight would seem easy to solve, becomes much more complicated because the mode of defining a single record depends on the country to which it belongs when it is declared. This means that the same entity may be defined in a different way depending on which country it belongs to. This is why the module was designed with an expert system, in that it is composed of rules which define the "knowledge" with which a Match can be made, and by an "inference engine", which scans all the defined rules and gives the results, which in the specific case are all the possible "matches".

Unlike a traditional "Expert System", where there is a phase of dialogue between the user and the "Expert System", for efficiency and because of a specific request made by those responsible for the work, the module works like a "Black Box", i.e. the CAHM user does not interact with the Expert System, another module being responsible for the decision part, the approval or refusal of a certain Match Group.

The two input files, shipped and receiver, are sorted depending on whether homogeneous record groups can be created, to make the process of consulting "matching" rules by the "inference engine" easier. Homogeneous groups are defined, records which have the following information in common:

Country FROM
Country TO
Class of Element Code
MBA Code
DATE of Matching
Element Code
Batch name

Obviously if all the values of the fields specified are equal we have an ideal situation, but the system considers the various degrees of aggregation of these homogeneous sets. In other words the system assigns levels of similarity to the sets and processes them on the basis of the types of rule which are appropriate for their state of similarity.

We will now examine the rules which define the generical case of "Matches". These rules are in relation to the type of link which can be found between an unmatched shipped and receiver record. The following types of link may be considered:

a) 1 Shipped v. 1 Receiver,
b) 1 Shipped v. M Receivers,
c) M Shipped v. 1 Receiver,
d) M Shipped v. M Receivers,

Below we give a brief description of the rules used:

We consider for a possible Match only records which belong to the same class of element code.

The element weight is equal to or less than a certain threshold which is a function of the element itself.

The date of the shipped record must be less than or equal to that of the receiver. The difference must be less than the time expressed in days needed to transport a certain batch from one country to the other. Thus this value is a function of the country pair.

Degree of similarly between the shipped record and its corresponding receiver. When the two batch names are the same the "match" is generated even if none of the other rules hold. This can be done only in the case of the 1 Shipped v. 1 Receiver relationship.

Before generating a match one must check if the batch name of the two records being examined has a high degree of similarity with any other batch name. This can be done only in the case of the 1 Shipped v. 1 Receiver relationship.

For each group of shipped or receiver records which have the same data create sub-totals, the sum of the weights and then check whether any corresponding batch has the same weight.

To generate a group of homogeneous batches for a possible match, each of the group components must belong to the same MBA.

Batch groups can be created with different element codes only when all other possibilities have been examined.

Below is the table of the threshold values applied to the various Elements and Isotope Codes for unmatched shipped and receiver records. This table represents the threshold values applied each time the program makes a Match. Sometimes the system also considers the possibility of creating a match even when the element codes are not the same but they always belong to the same class, in which case the threshold value applied is the smaller of the two considered.

Element Code	Isotope Code	Unit of Meas.	Rounding Difference
Enrch. Uranium	U-235, U-233	gr	1.0 gr
Depl. Uranium	Depl. Fissile	kg	500.0 gr
Natur. Uranium	Natur. Fissile	kg	500.0 gr
Unif. Uranium	U-235, U-233	gr	1.0 gr
Plutonium		gr	1.0 gr
Thorium		kg	500.0 gr

When the Expert System has consulted all the rules for all the unmatched records it generates two files which join the associations found (Matches) and the pointers to the ICR file.

One to One Matching Phase.

It is the objective of this phase to try to find all the shipment and receiver records that can satisfy the one type of link. The system scans the data structure of the receiver and for each unmatched record found it scans the shipment unmatched record directory. When the date of the shipment record is earlier than or equal to the receiver date the inner set of rules, which check the validity of the matching suggested, are activated.

The first rule says that the similarity between the two batch names is checked. To do this the program runs a procedure which assigns, on the basis of the degree of similarity of the two batch names, a value between 0, when the two batches are completely different and 100, when the batches are identical.

The first operation performed on the two names is to eliminate special characters and non-significant zeroes. The similarity value is then assigned on the basis of the number of equal characters and the position which two or more equal characters occupy inside the two batch names.

When the degree of similarity is not greater than the threshold value, one checks if the isotope is declared in the two batches. If yes, if the two isotope codes are equal, the rule is considered to hold. The second rule to check is the difference in weight between the two element codes, thus the program checks if the threshold value is less than or equal to the difference of the two weights.

The third rule checked is the equality of the element codes, but in the case of Depleted uranium, Natural Uranium and Unified Uranium this rule is considered to hold even if the two element codes are different.

The fourth rule checked is that the difference in days between the receiver date and the shipment date must be greater than or equal to the Transit Times already calculated.

When a shipment and receiver combination satisfies all the rules mentioned it is saved in an array which contains all the candidate shipment records for matching with the corresponding receiver.

When the scanning of the data structure of the shipment records is finished all the combinations found, which have been saved in the array already mentioned, are analysed and "weighed".

The procedure of "weighing" any combination means, in technical terms, assigning a certain value on the basis of certain parameters. This values represent the probability that the right Match has been found.

One thus takes the rules already mentioned analysing them again, but this time from the quantitative point of view, assigning a certain "weight". These "weights" are then summed.

Obviously the most probable combination is that with the highest sum of "weights". Before passing to the phase of writing any matching, the system checks if there is a more probable combination between the receivers which has not yet been checked, because the shipment/receiver pair which the system chooses at this point of the run represents the most probable between the current shipment and the set of receivers which satisfy the preceding rules. This does not however exclude the fact that there may be a receiver which has not yet been examined, which, coupled with the current shipment, could give rise to a new pair with "weight" which might be greater than that of the current pair.

To check this situation the following notion should be remembered. Of a set of candidates, which have conformed to all the rules cited above, in most cases, the one chosen is the one with batch name closest to the current receiver. Thus if there is between the shipment or the receiver records which have not yet been examined a record with batch name more similar than a fixed threshold, the system decides that there may be a combination with possibly greater "weight", thus it releases the combination being examined and goes on to examine the next receiver.

Matching One to Many Phase.

In this section we consider the one to many relationship which may occur in a match. This type of relationship is two-way, because there may be one shipment vs. many receivers.

Both the relationships are described in this section because the procedures are very similar from the software point of view.

The system checks both the relationships, but the order of execution depends on the number of unmatched records.

When the number of unmatched receiver records is greater than the unmatched shipped records the system executes the rules starting from the data structure of the shipment records, because the one shipment v. many receivers is more probable than the opposite relationship. In the opposite case it runs the rules starting from the data structure of the receiver records for the same logical reason, with the problem terms inverted. In this case also the induction engine runs a certain number of rules which are very similar to those run for the relationship considered above, the difference being due to the given priority and to the difficulty of running the rules themselves. This can easily be understood if one considers the rule.

Element weight of the unmatched shipment record equal to or less than a certain threshold, to the sum of the weights of the elements of the unmatched receiver records.

In mathematical terms this means calculating all the possible combinations for a certain set of elements n, for all the possible classes k, when k assumes the value from n, n-1, n-2, ... until k = 2.

The formula is the following:

$$\binom{n}{k} = \frac{n!}{k!(n-k)!}$$

Thus the inference engine runs all the other rules so as to reduce to the minimum possible the number of set elements to work with. As in the previous example, we will consider the case of one shipment v. many receivers. In this case the system saves the element weight, the isotope weight if declared and the inventory date of the current unmatched shipment record. Starting with these parameters one can examine all the rules which define a possible matching.

Using the directory which points to the MBA of the receiver records data structure, one runs the first rule which is:

1) *All the records belonging to the same match group must have the same MBA Code.*

In the next phase the inference engine runs the following two rules:

2) *The shipment record date must be less than or equal to the group of receiver records.*
3) *The Transit time expressed in days must be less than or equal to that of the group of records considered.*

On the basis of the number of records generated with the running of the previous rules, the inference engine decides whether to run the following rules.

4) *All the records which form part of the same group must have the same inventory date or a date within the same period (4-6 days).*
5) *The similarity of the batch names of the records of a certain group must exceed a certain threshold.*

The running of one rule does not necessarily imply the running of the second; the inference engine decides on the basis of the number of unmatched records. If this number is considered too high, the last rule is run cyclically increasing the threshold value incrementally each time until the inference engine considers the number of elements to be reasonable.

In effect the running of the rules by the inference engine acts as a "filter", reducing the number of unmatched records and making the remaining records more homogeneous.

On this last set of records the system runs the final rule.

6) *Weight of the element of the unmatched shipment record equal to or less than a certain threshold, to the sum of the weights of the unmatched receiver records.*

In cases in which the number of combinations is very large, the use of rules 4 and 5 serves to identify a subset of the set of all combinations. The final search for a match is carried out on the subset using rule 6. This kind of rule based heuristic search is necessary to prevent the system becoming bogged down in exhaustive computations which are unlikely to be more successful then the heuristic search.

Many to Many Matching Phase.

This section describes the Many to Many matching phase. The Expert System considers this phase as the last attempt to establish a relationship between two groups of unmatched shipment and receiver records.

This type of relationship occurs fairly rarely, if the preceding phases have been carried our correctly and exhaustively. For most cases this type of relationship (matching) occurs when the information available is not sufficient to find the relevant relationships in the preceding phases.

Let us consider the case of the country pair (Japan France). In this specific case the quantity of nuclear material, sum of the batch weights, declared as shipped from France is equal to the quantity of nuclear material which Japan declares it has received. As the number of batches is different, it is intuitive that there are one to many or more to one types of relationship (Matching), but because the information available to the expert is insufficient to verify these types of relationship the matching established is of the many to many type.

The rules which the inference engine runs are the same as those run in the preceding phase, but the approach is different, because this type of relationship says that there are many shipments and many receivers. Thus all the records which belong to the same MBA are agin subdivided on the basis of the same inventory date, and then on the basis of the country pairs the Transit Time is calculated. Again from this set we eliminate all the records with Transit Time greater than that given by the expert system.

All these operations have the aim of obtaining various more homogeneous aggregates of records (batches) from one non-homogeneous set of records and of reducing the number of records which define an aggregate. Once this operation is performed the inference engine dynamically generates an array which contains all the possible combinations for each previously generated aggregate.

The inference engine considers the array so generated as a directory. Each row of the array is a possible combination, thus for each line one sums all the weights of the records whose address corresponds to each element of the same line. Once the total weight is obtained the system also saves the inventory date, the highest in the case when the records which form part of the aggregate being examined have different inventory dates.

The operations which the system has performed until this moment have allowed the transformation of many to many relationship into a one to many relationship because the inference engine has available an inventory date and a weight, the same attributes which define a batch. Thus the system performs the same logical operations which were performed to analyse the preceding relationship. In the case when no valid matches are found the inference engine analyses the next combination. In other words a new weight with a new inventory date is generated and the process is repeated recursively until all the combinations for the aggregate being examined are exhausted.

5. Conclusion and Plan Future Work

The CAHM system was installed on the IAEA Vienna computer in October 1988 and for the first three months the system operated in parallel with the old manual system.

The results obtained by the expert system, following this first experience, were analysed and compared with those supplied by the human experts. As a result of the success of the comparison, it was decided to replace "Human

Matching" with the CAHM system. For international transfers this decision was operative at the beginning of 1989. The use of the system has led to a considerable saving in manpower and greater reliability in treating the matching of the data of nuclear (transfer) transport. The advantages thus obtained can be summed up below:

a) A Safeguards Analyst was given another task with no detrimental effect on the quantity and quality of the activity under examination.
b) The Safeguards Analysts considered more than 90% of the CAHM "Suggested Matches" output as valid.
c) As a result of the implementation of CAHM the Safeguards Analysts have more time to solve more complicated cases.

Considering that the CAHM system is a logically independent application a distributed processing system could be implemented where the kernel of the system could be accessed from several workstations which are connected to a Local Area Network (client-server architecture). The migration from a centralized system to a distributed processing system should be discussed and eventually approved by the IAEA Vienna Safeguards authorities.

6. References

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MANU - ICR ACQUISITION AND TRANSMISSION THROUGH MINITEL

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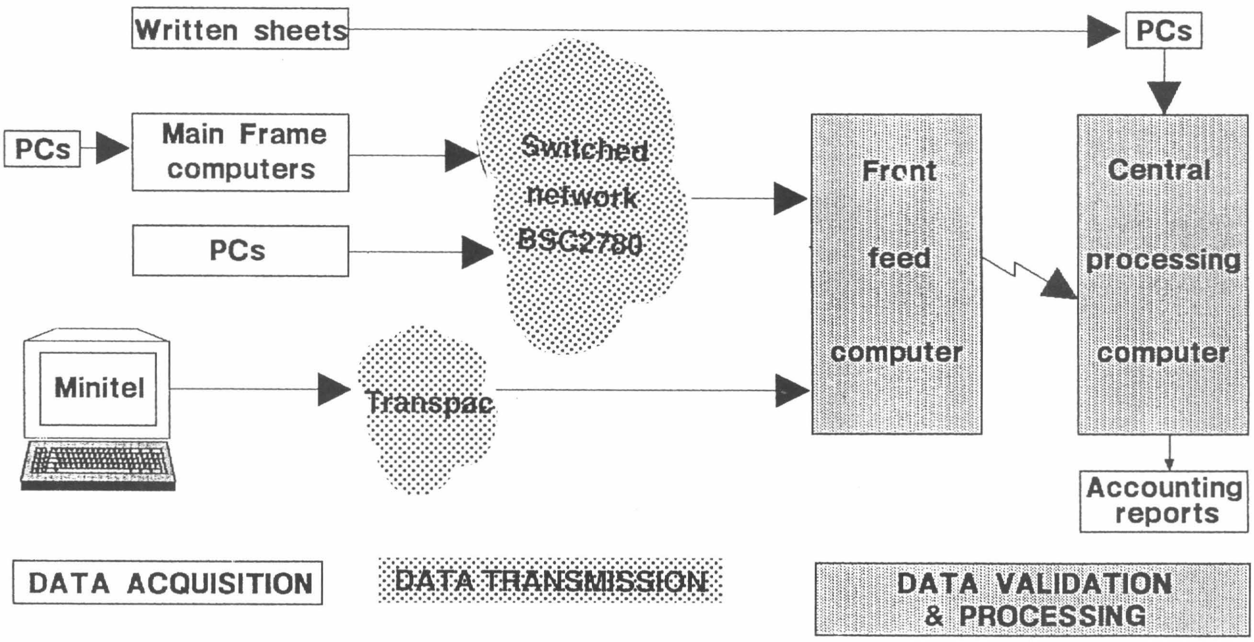
Abstract

Since last September a new telematic transmission mode for domestic accounting reports is available to correspondents that used paper and the usual postal services for sending their reports to IPSN. It is called MINITEL. This type of link is made in conversational mode with IPSN's front feed computer and makes use of the Transpac network. It is accessible from a micro-computer, a terminal or a minitel. A minitel is a device consisting of a telephone set linked to a printer and a screen with a keyboard similar to that of a typewriter.

This system is perfectly suitable for the transmission of small amounts of data and should thus be used by small facilities which often make few reports with, logically, numerous erros. For less than 10,000 FF, this device makes possible the following: the acquisition of reports by means of data acquisition masks, strictly identical to the nuclear materials operation report form; the consulting of reports by relating to previous accounting reports; the retrieval of reports issued by other units (in the case of shipper/receiver relations); the correction of inaccurate reports; the issue of accounting reports by copying the screen on which there is an image of the single form; assistance for operators.

SCHEMATIC OF COMPUTERIZED SYSTEM FOR ACCOUNTING DATA CENTRALIZATION



NB Also : data authentication

April 1991

" MANU " MENU

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- GET BACK ICR RELATING TO SHIPMENT OR RECEIPT ACKNOWLEDGEMENT
- GET HELP
- PRINT ICR

FISSILE MATERIAL ACCOUNTANCY ON PERSONAL COMPUTER

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Abstract

Recently, a new accountancy programme for nuclear material has been installed on a PC. Being programmed in dBaseIII, it represents the advantage of being user-friendly and having easy access. The programme treats the problems met in a diversity of installations such as fuel fabrication laboratories, research laboratories, PWR and research reactors with dismountable fuel assemblies. Examples of this flexibility in use are given.

1. Introduction

For almost fifteen years, our fissile material accountancy has been treated on the central computer, of our institution (different types of IBM computers). However, as a result of the constant increase of the number of users and realization of the different accountancy tasks, in a reasonable lapse of time, it has become more and more difficult. This has been the reason why we decided in early 1989, to change our policy and to keep the fissile material accountancy on an independent personal computer.

Taking into account our experience acquired by the different successively used programmes, we have made a complete analysis of the overall problem and have asked the software company HAVE (IBM dealer at Mol), to rewrite the programmes required for the realization of our accountancy.

The accountancy is done on the following configuration :

- a personal computer : PS2 model 80 with a hard disk unit of 111 Mb and 4 Mb fast memory
- a 5.25" floppy disk drive
- a 3.50" floppy disk drive
- a tape streamer for the back-up
- an IBM model 4216 laser printer

For writing the programme, use was made of the Clipper software from the company Nantuket, allowing management of the files and offering moreover possibility of compiling the realized programme, thus presenting the great advantage of an accelerated data processing at the level of the final user.

2. Description of the software

All data are stored in databases which can be divided into two groups : the main files and the auxiliary files, helping the user while executing the accountancy.

I. MAIN FILES (Each table gives the contents of the file)

This group includes the files containing the data for the different batches and reports. The group comprises 7 files.

1. Smbatch1.dbf : contains in 1 record per batch all general data for this batch.

Field	Field name	Type	Space	Decimals
1	Name	Character	8	
2	MBA	Character	4	
3	Place	Character	10	
4	Seal	Character	8	
5	Description	Character	4	
6	Use	Character	2	
7	Measurement	Character	1	
8	Item	Numerical	4	
9	DOC	Character	10	
10	Date	Date	8	
11	Info	Character	20	
12	Type	Character	2	
	Total		81	

2. Smbatch2.dbf : contains per batch the data concerning the nuclear material contained in this batch from Smbatch1.dbf. This file contains as much records per batch as there are different types of nuclear material in this batch.

Field	Field name	Type	Space	Decimals
1	MBA	Character	4	
2	Name	Character	8	
3	Post	Numerical	4	
4	Metweight	Numerical	10	3
	Total		26	

3. Smic.dbf : contains all records for establishing an ICR.

Field	Field name	Type	Space	Decimals
1	MBA	Character	4	
2	Date	Date	8	
3	KMP	Character	1	
4	Measurement	Character	1	
5	Tbook	Character	2	
6	OMBA	Character	4	
7	Name	Character	8	
8	Item	Numerical	4	
9	Descr.	Character	4	
10	Elt	Character	1	
11	Eltweight	Numerical	10	3
12	Eltunit	Character	1	
13	Iso	Character	1	
14	Isoweight	Numerical	10	3
15	Isounit	Character	1	
16	Obligation	Character	1	
17	Use	Character	2	
18	Oinfo	Character	8	
19	Book	Numerical	1	
20	Cor	Character	1	
21	Odate	Date	8	
22	Doc	Character	10	
23	Note	Character	30	
	Total		121	

4. Smba.dbf : contains the totals per element and per obligation for the different MBA on the declaration data.

Field	Field name	Type	Space	Decimals
1	MBA	Character	4	
2	BAdate	Date	8	
3	Elt	Character	1	
4.6.16	Obl. Met.	Numerical	12	3
5.7.17	Obl. Fiss.	Numerical	12	3
18	Code	Numerical	1	
	Total		168	

5. Smpb.dbf : contains the totals per element and per obligation for the different MBA on the date of the physical inventory.

Same table as sub 4., except second line : BAdate read : PBdate

6. Smdat.dbf : contains per MBA the date of the last declaration to Euratom and of the last control by Euratom.

Field	Field name	Type	Space	Decimals
1	MBA	Character	4	
2	Declaration	Date	8	
3	Inspection	Date	8	
	Total		20	

7. Sminbd.dbf : contains the totals per element and obligation at the date of inspection for the different MBA.
Same table as sub 4., except second line : BAdate read : Inspdate

II. AUXILIARY FILES

This group consists of the statical files containing the required information which can be asked by the programme when realizing the different bookings. This group comprises 5 files :

1. Smickode.dbf : contains per MBA the possible admitted IC-codes with their KMP and a conversion factor.

Field	Field name	Type	Space	Decimals
1	MBA	Character	4	
2	ICCD	Character	2	
3	ICCF	Numerical	2	0
4	KMP	Character	1	
	Total		9	

2. Smmater.dbf : contains the codes imposed by Euratom concerning the material description, measurement and use.

Field	Field name	Type	Space	Decimals
1	Form	Character	2	
2	Packing	Character	1	
3	Situation	Character	1	
4	Use	Character	2	
5	Measurement	Character	1	
	Total		7	

3. Smmbg.dbf : contains all MBA codes of the Community, known to us, with their address and a code whether they belong to SCK/CEN or not.

Field	Field name	Type	Space	Decimals
1	MBA	Character	4	
2	Address	Character	45	
3	INT-EXT	Numerical	1	
	Total		50	

4. Smpla.dbf : contains per MBA all possible locations where nuclear material is allowed to reside with their KMP.

Field	Field name	Type	Space	Decimals
1	MBA	Character	4	
2	Place	Character	10	
3	KMP	Character	1	
	Total		15	

5. Smpost.dbf : contains, per post, the data concerning element, obligation, isotopic composition.

Field	Field name	Type	Space	Decimals
1	Post	Numerical	4	
2	Elt	Character	1	
3	Obl	Character	1	
4-17	Isol-14	Numerical	8	
	Total		118	

3. Application of the software

This programme is used via programmed screens which, per selection, allow to realize the desired accounting operations : entry, modification or erasion of data in an auxiliary file are carried out by means of the programme and have only influence on the selected file. All data of the auxiliary files may be consulted on the screen or on the document printed on request.

Recording of the accountancy movements of the batches in the main files influences the different files, the relations and functional connections of which are programmed.

This programme allows not only recording of all accountancy movements to be obligatory declared to the international control bodies, but also to realize recording on diskette of the monthly report transmitted to EURATOM (Inventory Change Report - Physical Inventory Listing - Material Balance Report), to obtain quickly inspection manuals required during the inspections (about 40 per year for the whole of the 7 installations of our research centre) and also the Physical Inventory Listing and the Material Balance Report, foreseen in the regulation. By drawing up, as a Working Paper, a recapitulatory list of all inventory modifications brought to the inspected installation since the last control, we facilitate the necessary verifications thus allowing decrease of the duration of. the inspections.

We can also retrace the complete history of a material batch with its divisions, present locations (all data since 1983 are recorded on the hard disk of the PC, the former data remain accessible via the central computer, if necessary).

The possibilities for modifications which must not be declared to the international control bodies are also included in this programme, such as : change of the description, quality, use, form or quantity of items or affixture, change or removal of the seals on a batch or on a location.

Owing to the geographical dispersion of our installations and the dimension of some (the whole of the laboratories comprises officially 12 key measurement points of which 7 are really active), it is very important, in the scope of safety, to be able to locate, at any moment, the different materials and their movements even when we are not legally obliged to declare the latter to the international control bodies.

This control on the movement of the materials is also of prime importance in an installation such as reactor BR2, where shift work is carried out 24 hours on 24 all year around and where each shift carries out its part of the manipulations during each loading - unloading period (every month); these are thus entered directly by the shift responsible in the specific system of the installation.

To avoid transcription errors, the central accountancy can, via a central computer, have access to the internal lists of BR2 and ask a list of the modifications carried out since the last consultation (receipts, shipments, change of the key measurement point, change of the storage location inside a same measurement point ...).

Since the reactor BR3 has been stopped, we are carrying out the last movements of the pins such as to be able to reconstitute a certain number of complete dismountable fuel elements which are to be stored in a container during a transitory period which is still to be determined; for this reason, it has not been judged necessary to establish a link similar to that of the reactor BR2.

4. Conclusion

We can say that this programme is largely wider and more powerful than required by the international control bodies.

It allows to cover certain aspects of our safety system by a continuous follow-up of the material batches staying in our installations. It also allows to realize certain economies at the level of time required to execute the accountancy and a reduction of the duration of the inspections at the level of the accountancy control.

OPERATION OF THE ADVANCED MULTI-LANGUAGE PC SYSTEM FOR SAFEGUARDS REPORTS AND INSPECTION SUPPORT FOR SINGLE-MBA OR MULTI-MBA

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Abstract

The new advanced multi-language PC version of the system for safeguards reports and inspection support is now in operation at the Risø National Laboratory for all Danish installations.

The system can be used on PCs in other Community countries, because the language of the display text can be selected on a menu. The programs are being delivered to Petten for use for 3 material balance areas.

The system has been improved by significant increased capacity and speed. A sub-batch system has been integrated in the general programs.

1. Introduction

The new advanced DOS version for PCs of the system for production of safeguards reports and inspection support documents is now in operation in Denmark. It is used for all Danish installations.

ICR and BA reports have been submitted on diskettes to Euratom with respect to reports since 1 February 1991 for all these installations. Future PIL and MBR reports will also be sent on diskettes.

Since 1984 the high quality inspection support produced by the previous CP/M version has been appreciated by Euratom and IAEA inspectors, because their tasks were eased. The system has eased significantly the tasks at the safeguards office and at the various installations.

The new DOS version for PCs includes additional possibilities for inspection support.

The system can be used in other Community countries. All display texts can be shown in English or in another language selected on a menu. For ICR printouts the form can be printed in English or in another selected language.

The flexibility of the system makes the operation easy for a single MBA, for a group of MBAs at an installation, for a number of MBAs (regional use) or for all installations in a country. It can also be used for central safeguards administration of all Catch-All MBA installations.

The programs are being delivered to ECN Petten in the Netherlands for use for three material balance areas. Various improvements of the programs have been made in order to ensure that the general system will comply also with all safeguards and operational needs at Petten.

2. New Advanced PC System

Multi-Language Version

The new advanced version for PCs is designed for use in the various countries within the Community.

The standard language for all display texts is English, but another language for all display texts can be selected on a menu. Hereby, operation in these countries is eased.

The printout form for the inventory change reports is in English. However, it is possible to print the form in another selected language. Copies of ICR printouts can be used as official document to the receiver, e.g. at international transfers.

The programs can be operated on a large group of personal computers comprising models from IBM, Olivetti, and other manufactures producing IBM compatible PCs using PC-DOS or MS-DOS operative systems.

The advantage of using a PC, which is not connected to any external line, is that unauthorized persons can not interfere with or extract safeguards data. The security aspects are limited to the room, where the PC is located.

Purpose of System

The system can be used for e.g. light water power reactors, research centers as well as bulk handling installations having rather simple safeguards accountancy needs. Special modified versions can be developed on request. However, it is not the intention of the program designers to solve the problems at large bulk handling installations requiring complicated accountancy.

Safeguards tasks are rather standard, while operational tasks are individual for the various types of installations. The operational tasks depend also on the size of the installation.

Therefore, it is an advantage to separate safeguards tasks from the operational tasks. Hereby, the user of the system is not obliged to comply with special requirements for other types of installations.

The new PC system is designed for the standard safeguards tasks and for the production of inspection support documents.

The system can produce reports:
- Inventory change reports. Concise notes can be entered.
- Monthly reports on the book inventory sorted by obligation.
- Physical inventory listings.
- Material balance reports.

The reports required by Euratom can be submitted on diskettes for direct use in the Euratom Safeguards Directorate.

The system can also produce various types of high quality inspection support documents. They ease the inspections very much. The various types are described in this paper.

A new sub-batch system has been integrated in the general programs. The user can enter batch data or sub-batch data in the PC. In both cases the system produces the safeguards reports at batch level as required by Euratom.

The new version of the system could be useful to operators not only with respect to the safeguards tasks but also with respect to operational tasks. Supplementary programs can be added.

Single-MBA/Multi-MBA Operation

The system is very flexible. The operator of a single material balance area can easily use the system as a SINGLE-MBA system.

If more than one MBA exist at the installation, e.g. at a power reactor site, then it will be an advantage to use the system as a MULTI-MBA system for the MBAs.

Also at a research center benefits are achieved, if a central safeguards office uses the system as a MULTI-MBA system.

It is also possible to use the system as a regional system for a group of installations.

Finally - as in Denmark - the system can be operated as a national system for all MBAs and for all Catch-All MBA installations in the country.

The harddisk contains all inventory change reports and book inventory information on particular days for all MBAs covered by the system.

For each material balance area, the updated inventory information is stored on a separate diskette. The diskette contains batch data as well as sub-batch data.

Fast and Easy Operation

The new advanced version for PCs replaces the CP/M version that has been used in Denmark since 1984. Improvements have been included where possible on basis of extensive research performed by a safeguards expert and a programming expert since 1984.

Direct access to the mostly used programs is very easy from DOS at start of the PC. Access is also possible through the main menu and the menus for the various parts of the system, but direct access to programs is very fast. Direct access from DOS to the menus for the various parts is also possible. Therefore, it is not necessary to call the main menu.

The display colours can be changed by the user. Hence, each user can select the colours he prefers. This provision is similar to the provision that the language of all display texts can be selected on a menu by the user.

At operation of the programs, it is possible to benefit from the individual MBA guide text (5 pages), which is shown automatically, when needed. The user can change this text at any time.

The operation of the various programs is also eased by fixed guide texts on the display.

At a transfer between 2 MBAs covered by the system, it is possible to produce shipper's inventory change report as well as receiver's inventory change report from one input to the PC.

The input tasks are also reduced due to standard information stored in the PC. Repetition of data are performed automatically or by pushing the key ENTER.

Enrichment information can be used for calculation of U-235, if wanted. Then the quantity of U-235 is not entered.

In the batch field it is possible to enter 15,149 for a batch 15/149. When the key ENTER is pushed, then 15,149 is converted to 15/149. Therefore, it is not necessary to use the key SHIFT. One hand can enter data, while the other hand can be kept at the line in the source document.

Concise notes can be entered in the inventory change reports. Experience has shown, that the tasks in the safeguards office are performed more effectively, if useful information is entered.

It is very fast and easy to produce safety copies of data on inventory changes, on book inventories on specific days, as well as on the batches and sub-batches in the various material balance areas.

New Integrated Sub-Batch System

In order to limit the accountancy tasks in the situation, where many similar items are handled, it may be useful to consider these items as sub-batches within a particular batch.

The new advanced PC system includes a sub-batch system integrated in the ordinary programs. The user can enter information on batches or on sub-batches. In both cases, the system produces e.g. physical inventory listings and inspection support documents at batch level. The data are used also at the production of material balance reports.

The sub-batches within a batch may concern one or more material category, one or more enrichment, and one or more obligation. However, the program calculates information at batch level for each material category, enrichment and obligation.

A batch may consist of a number of sub-batches located in various places. Here, it is easy to update the location information in all lines for a sub-batch, if it is moved to another place, or transferred from the batch.

The sub-batch consolidation listing shows the data for each sub-batch as well as the totals for the batch. Therefore, it is very useful at inspections.

An item listing on all items, or on selected items, sorted in sequential order, can be useful to the operator.

A batch may also consist of a number of sub-batches, which all are located in the same position. Here, it is easy to update the position for all sub-batches. All lines can be deleted in case of shipment.

An example is a fuel assembly consisting of rods, which can be replaced by other rods. Each rod is a sub-batch, while the fuel assembly is the batch. Movement of the total fuel assembly can be entered in the PC. It is also easy to update the PC, if a rod is moved to or from the assembly.

The sub-batch consolidation listing shows the data for the various rods as well as the totals for the assembly. It is very useful at inspection of such assemblies.

An item listing can show the rods in a sequential order and the assemblies, they concern.

Inspection Support Documents

The inspection of accountancy is eased by the following support documents:

- Book inventory updating document showing the inventory at the previous inspection, consolidated information for each type of change on a particular day, and the book inventory at the new inspection. Inventory information is also shown according to IAEA specifications.
- Inventory change report printouts with and without obligation.
- PE/BA printouts for the physical inventory and the book inventory, both sorted by obligation.
- Physical inventory listings with and without obligation.
- Material balance reports.

The inspection of materials is eased by the following support documents:

- Inventory distribution document showing the total inventory within each KMP and AREA. It also shows the number of batches and items.
- Updated inventory listing showing the various batches sorted by KMP, AREA, POSITION. It does not contain obligation information.
- Sub-batch consolidation listing showing the various sub-batches within each batch. The lines for the total quantities for a batch are equal to the lines for the same batch in the updated inventory listing.

It should be noted that more than one KMP can be used for the batches within the inventory part of the system in order to ease the inspections. If only one KMP is accepted by Euratom in physical inventory listings, then the system can convert all KMPs to A at the production of physical inventory listings for Euratom.

3. Operation in Denmark

Safeguards Administration

In Denmark, the new advanced DOS version for PCs replaces the CP/M version that has been used since 1984 at the Risø National Laboratory for all Danish installations.

It will be used for 8 MBAs at Risø, 5 MBAs elsewhere in Denmark and 18 Catch-All MBA installations. The MBAs at Risø comprise now a fuel fabrication plant, two reactors, a hot cell facility, a storage MBA, a waste installation, a uranium ore MBA and research laboratories.

The conditions in Denmark are such that it is not necessary to use regional administration (all MBAs at Risø and near Risø). It is convenient to include all other installations in Denmark.

The efficiency of the safeguards administration was increased significantly by use of the CP/M version. Many tasks - such as production of printouts and production of the monthly diskette for ICR-BA reports to Euratom - are performed by the secretary at the national safeguards office.

The increased speed of the new DOS version for PCs leads to further improved conditions at the safeguards office.

Since March 1984 Euratom Safeguards Directorate has received diskettes containing more than 25,000 records concerning reports. There have been no problems at the transfer of data.

Procedures

The safeguards office administers all safeguards tasks for all installations in Denmark. Standard procedures ensure that reports comply with Euratom requirements. Errors in data are also avoided.

The operators can concentrate their efforts on operation of the installations. The local, individual registration system for nuclear material can be developed in such a manner that the operation of the installation is most effective without any unnecessary safeguards administration. However, link to the accounting system at the safeguards office is necessary.

The advantage of the individual local systems is that operators at small installations are not obliged to comply with the complicated accountancy requirements for the large installations.

The safeguards office produces the inventory change reports on basis of minimum information from the operators as well as safeguards information in the ledger sheets kept in the safeguards office. The information is entered in the accounting system by the secretary, and the safeguards officer performs then an audit.

At a transfer between two material balance areas covered by the system, the safeguards office is able to produce the inventory change reports for the shipper as well as for the receiver from one input to the PC.

The safeguards office updates the inventory information at physical inventory takings on basis of information from the operator. The physical inventory listing and the material balance report are then produced.

The tasks at the safeguards office are limited by concentration on changes of the total inventory at each MBA. Below is described the preferred administration.

The new, advanced PC system is very flexible and can be operated in various manners. There may be circumstances, where the operator e.g. by using the inventory part of the system himself can achieve significant benefits at the operation and at the inspections.

Accounting System at Safeguards Office

For process material undergoing daily physical changes, the safeguards office uses a consolidated batch-account for material of a particular enrichment and obligation. The daily processes are not recorded in the ledgersheet for this account. Receipts, shipments, and transfer to another batch are recorded promptly. Rebatching to ordinary batch accounts are performed at physical inventory takings and just before inspections in accordance with the physical situation. Consolidated process differences and roundings are then reported and recorded for each consolidated account. Hereby, the assessment of the inventory differences is eased.

Each fuel assembly at a reactor constitutes a batch, for which the safeguards office keeps a ledger sheet. Just before inspections the PC is updated with respect to the location of the various assemblies.

If many similar items are handled at an installation, then it is convenient to use a sub-batch system. Hereby the number of ledger sheets is reduced, because one ledger sheet can concern a number of items.

For a batch consisting of a number of items, that normally are not changed or moved, a sub-batch listing is kept at the safeguards office. This listing has information on location of the items. The total quantity is recorded in the ledger sheet for the batch.

The ledger sheet for a batch includes information on the dates for all changes that have been reported. The corresponding MBA or batch is also recorded.

The PC contains the updated safeguards information on batches and consolidated batches corresponding to the data in the ledger sheets. The sub-batch system, which is integrated in the new PC version, makes it possible to enter sub-batch data instead of batch data. Location information can be updated just before an inspection. Then the inspection support documents can be printed rapidly. Copies are ready for the inspectors, when they arrive.

Accounting Systems at Installations

The individual accounting systems at the installations should comply with the operational needs. Experience has shown that some operators prefer that unnecessary safeguards information should not be entered by the operator. There is a high risk of errors, if persons without sufficient safeguards education enter data. Wrong data at the installations could give rise to confusion at inspections. Instead, documents produced by the safeguards office are kept as correct safeguards information.

The starting document is the inventory change report for the receipt of the material. The safeguards office ensures at the production of the report that the data correspond to the official safeguards information from the shipper. Frequently, different data are declared in the various transport documents.

A copy of the inventory change report is checked and signed by the operator. This copy is returned to the safeguards office, where it is used as a source document for the safeguards accounting. Another copy is stored at the installation.

Subsequent reports are produced by the safeguards office on basis of minimum information from the operator. A copy is signed by the operator and returned to the safeguards office. Another copy is stored at the installation.

Furthermore, the operator receives copies of the monthly BA reports on the book inventory sorted by obligation, as well as the physical inventory listings and material balance reports.

The local accounting and identification system should comply with the operational and safety requirements. It should also include convenient provisions for fast identification of the various materials of a particular batch or consolidated batch recorded by the safeguards office.

At least one convenient parameter concerning the quantities (e.g. the gross weight) should be recorded for use at the verification of the total quantity declared at inspections. By use of supplementary standard information (e.g. enrichment and U-factor) for the particular batch, it should be possible to verify the safeguards data on weights of U and U-235 declared in the inspection support documents.

If e.g. the weight of U_3Si_2 is used as operational parameter for certain fuel fabrication processes, then these quantities can be used for establishment of a material balance for the batch based on this parameter. Conversion to safeguards data leads then to the data in safeguards reports and inspection support documents.

As mentioned previously, the operator may have advantages by operating e.g. the inventory part of the advanced PC system.

The accounting and identification systems have been developed in close collaboration with the safeguards office.

Inspections

The inspection support documents are produced by the safeguards office after updating of the PC data with respect to the situation just before the inspection.

The inspection support has eased the tasks of the inspectors not only during the inspections but also at subsequent evaluations.

The inspectors perform accountancy checks in the safeguards office as well as inspection of materials in the installations.

When four inspectors perform the inspection, then two inspection teams are established. The secretary in the safeguards office assists the team concerning accountancy checks, while the safeguards officer assists the team performing verification of materials. When the accountancy checks have been completed, then both teams check materials.

The inventory at the previous inspection is updated at the accountancy check with respect to changes since that inspection, and the new book inventory is calculated. The PC produced inspection support document Book Inventory Updating and copies of inventory change reports - with and without obligation information - ease this task very much.

The inspectors also check that the changes declared in the inventory change reports signed by the operator have been recorded in the ledger sheets. They also check relevant transport documents, which are kept as source documents.

Printout of the physical inventory listings - with and without obligation information - and material balance reports are ready, when the inspectors arrive.

The printout of PE/BA on the total physical and book inventory - sorted by obligation - is given to Euratom inspectors only.

At the installations the staff from the safeguards office accompanies the inspectors and advises them with respect to various aspects. Furthermore, the operators are advised with respect to rights of inspectors and operators. One staff member from the installation is present, but he does not need to be familiar with safeguards aspects. The operators have appreciated this assistance that has reduced the work load of the operators.

The various inspection support documents ease the tasks of the inspectors very much:

- The document on the inventory distribution can be used to assess the number of items to be selected for inspection.
- The updated inventory listing shows the batches sorted by location. The inspection time is reduced by using this document.
- The sub-batch listings are also very useful at the inspection of batches. The consolidated quantity is equal to the information in the updated inventory listing.

The staff from the safeguards office also checks identification and weight data for the batches and items. Improvements are organised, if needed. Hereby, the intended safeguards quality is ensured.

4. Conclusions

The new advanced multi-language PC system for safeguards reports and inspection support is used for all installations in Denmark. It can be used in other Community countries, because the language of display texts can be selected on a menu.

The system can be operated for a single MBA, a group of MBAs or for all MBAs in a country. The new effective sub-batch system, that is integrated in the general programs, may be very useful to operators.

The system constitutes an important improvement of the CP/M version that has been used for all Danish installations since 1984.

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CLOSING SUMMING-UP BY THE CHAIRMAN OF THE SYMPOSIUM

B H PATRICK
UKAEA, Harwell, UK

This meeting has covered the complete spectrum of safeguards topics, ranging from the political and regulation aspects, and through all the technical areas which contribute to the subject. To attempt to summarise such a diverse range would be a very daunting task and one which is beyond my capabilities.

The Secretaries of the Sessions have kindly provided me with summaries but to go through each one would take too long and I am sure you do not wish to hear a long speech. Instead therefore, I propose to pick out a few highlights, selected on a personal basis, but in the proceedings I shall include the summaries by the Session Secretaries.

To me, the highlights have been as follows:

> In the initiatives which are beginning to be brought forward on energy policy in the European Community, nuclear power is expected to continue to play an important role and safeguards are an essential ingredient in that. They will also be of high importance in the increase in nuclear trade which could result when the open market comes into being at the beginning of 1993.
>
> An important landmark was reached recently when fresh MOX fuel was verified for the first time under water by the Euratom Inspectorate. The anticipated increase in the flow and use of MOX in the future makes it essential to devise and demonstrate successful safeguards measures which can give the necessary assurance. The ability to verify under water is a key part of that.
>
> A number of organisations are working on schemes for randomisation of inspections or zone approaches. Mr Gmelin's remark that while the elegance of the techniques is unquestionable, the practice is not so encouraging. I suspect this is the first shot in a discussion that will last for some time and I look forward to seeing the outcome of the argument.
>
> The advances in NDA techniques in recent years have been remarkable. In some areas, they are beginning to approach capabilities so far achieved only by DA methods and this can only be good for safeguards. I would not be surprised if this infringement of the prime position held by the DA community stirs them into even greater efforts, resulting in further improvements in their armoury. This rivalry is a great stimulating influence which will inevitably lead to improved accuracy and therefore improved safeguards.
>
> Recent developments in C/S techniques seem to have eliminated the Cinderella image which it has had. The application of modern, sophisticated techniques has brought about a revolution and raised the status of C/S very significantly. Data are now of higher quality which leads to greater assurance. This is a much-needed development because, with the limitation on inspection resources making it difficult to keep pace with the increasing number and complexity of nuclear installations, greater reliance on C/S is required. C/S seems to have risen to the challenge and those responsible for the advancement are to be congratulated.
>
> The participation of Eastern European countries in this Symposium is a very welcome addition. Both sides have much to offer and can learn from each other. I hope their participation is a trend which will continue, to the benefit of all.
>
> If I had to select only one highlight, it would have to be the trend towards integration. Integration has been with us for a long time but I lost count of the number of times the words "integrated system" were used by presenters. More and more, complete systems are called for, requiring contributions from several technical areas. Besides producing better, more reliable, safeguards systems, integration brings together personnel involved in different disciplines and this must be an additional benefit. Using the word "integration" in a looser sense, I have been struck by the number of papers involving authors from different organisations, not only R&D ones but also the inspection agencies. Safeguards is an international business and collaboration adds to this dimension and is therefore to be encouraged.
>
> I am sure that each of you, if you had been in my position, would have selected an entirely different set of highlights; and that is how it should be; discussion, collaboration, different ideas, all have an important role to play and it would be dull if there were no disagreements.
>
> The programme for this Symposium held out the prospect of an interesting meeting and we have not been disappointed. I believe it has been an excellent gathering, with stimulating papers and exciting developments. The future for safeguards looks very bright and we can look forward to facing challenges with confidence and the prospect of making a major contribution to a safer world.

Acknowledgements

We come now to the point in the proceedings when the (metaphorical) bouquets are given out. On behalf of all the participants, I should like to express grateful thanks to:

Hosts of the dinner, EDF, COGEMA & CEA. A splendid evening that will be remembered with pleasure for the rest of our lives. Excellent

food, superb entertainment in a truly magnificent setting, what more could one ask.

Professor L Stanchi - I cannot imagine what it would be like to have a Symposium without the presence and hard work behind the scenes of Professor Stanchi. He is the personification of the ESARDA Symposium and, as we have come to expect, he has organised another outstanding meeting. I am sure he will once again finish it all off by producing the proceedings in a very short time, adding maximum value to the contributions. Assisting Professor Stanchi as Scientific Secretary was Mlle Neuilly. Her hard work and contribution have also been instrumental in ensuring the success of the meeting and for that we acknowledge our gratitude.

The Commissariat a l'Energie Atomique (CEA) of France - our hosts are to be congratulated on their choice of location. The facilities have been excellent and we are grateful for the effort they have put into ensuring the success of the meeting.

The Technical Programme Committee - selection of papers is a thankless task but an important one. We are indebted for their wisdom in selecting a worthwhile and stimulating programme.

The Executive Secretariat - Mrs C Tharaud, Mr P Marmonier and Mrs M F Morier, all of the CEA. Helped by the secretarial and administrative personnel Mrs D Saxby, Mrs S Lutz, Mrs A Cipolletti and Mrs M Leconte. Also personnel taking care of accommodation and security. They have provided valuable support to participants throughout the week.

Authors of papers (both oral and poster) - without their presentations, the Symposium would be a nonentity. Thank you for the trouble you have taken to inform us of the work going on in safeguards and nuclear materials accountancy around the world.

Chairman and secretaries of sessions - for their efficient conduct of the sessions and for ensuring that they ended close to time.

Participants - for providing such an interested audience and stimulating questions.

Interpreters - who have coped with a wide variety of technical jargon admirably and managed to keep up with presenters, keen to tell us about many aspects of their work.

Projectionists, technicians and all others who have contributed to the efficient running and success of this Symposium.

Session 1: Invited Papers

In the first session four speeches were given by invited speakers. Two of them concerned the French approach to nuclear safeguards, both technical and political; the third one presented the energy policy of the Commission of the European Communities; the fourth reviewed the support programmes to the IAEA by the Member States.

The presentation of Mr Laureana, of the French Ministry of Industry, highlighted the French approach to safeguarding of nuclear materials. Safeguards in France has much broader scope than international safeguards, including among its objectives materials which are not considered by international safeguards (e.g. D, T, Li-6, and military materials), consider such issues as physical protection of materials and installations, and also addresses such problems as the verification of good technical management by operators.

Mr Felten, Deputy Director of the International Relations Department of the CEA, pointed at the need of reconsidering certain aspects of international safeguards, since the recent events in the Middle East showed that some of the political assumptions on which the present system is based might not be verified in the future. In particular: (i) there is the possibility that nuclear weapons are considered for actual use in local conflicts rather than for deterrence, and (ii) there is the fear that weapons grade materials might be obtained through an undeclared fuel cycle rather than diverted from the civilian cycle. These facts might require a revision of certain limitations now imposed on safeguards activities, more intensive use of all existing safeguards provisions and the possible introduction of appropriate non-quantitative criteria in safeguards implementation.

Mr Gmelin, on behalf of the Director General for Energy of the Commission of the European Communities, presented the main lines of the energy policy of the Commission. Having stated that energy is a major economic issue in the Community and that the Commission aims at achieving a "secure energy supply to satisfy demand at reasonable cost and low environmental impact", Mr Gmelin said that this target may be achieved through a Single Energy Market. The Commission is working to this objective by issuing appropriate regulations to harmonize national energy policies, by promoting common industrial strategies, by strengthening relations with countries of the EFTA and of the former East European economic block. Specific collaboration programmes are now developed for East Europe. The importance of nuclear power in Europe is high, since 35% of the EC electricity is now produced in this way by 134 reactors. Public acceptance of nuclear power is a major concern for the commission, and safeguards is one of the means to make the nuclear option credible. In this context, the Commission is giving considerable attention to the recent statement of Mr Blix on the strenghtening of safeguards.

Mr Naito, Director of the Division of Development and Technical Support at the IAEA, highlighted the importance of the Support Programmes of the Member States to the IAEA for the development of safeguards instruments, procedures and techniques. The number of these programmes has steadily increased from the original one in 1976 to the present 14, and covers now 150 tasks for a total budget of 22 million dollars. The programmes practically cover the needs for R/D of the IAEA and are coordinated through national coordinators' meetings. Diffusion of information is achieved through the SPRICS Informatic System.

Session 2A: Safeguards Systems and Implementation I:

The first session (2A) on Safeguards Systems and Implementation was entirely devoted to new concepts for safeguards implementation.

In presenting his paper, "Contributing to Goals concepts for Safeguards Implementation", Mr Gmelin emphasised the fact that by the end of the 1990s Safeguards in the European Community will have to cope with an increased availability and use of plutonium, on increased use of MOX in LWR, with the presence of large commercial fuel fabrication and reprocessing plants, and of plants to condition irradiated fuel before repository. Consequently and in order to continue to improve the effectiveness and the efficiency of the safeguards operation, with necesasrily limited resources, safeguards concepts and approaches need new thinking

- Relating to the technical goals, i.e. the tryptic of characteristic amounts, times and probabilities. In this line the IAEA Safeguards Criteria for 1991-1995 are under review in the perspective of the Unique European Common Market in 1992.
- For large fuel cycle installations in the line to gradually replace manpower by machine power.

Mr Lu and Mr Canty, in spite of different approaches, presented stimulating thoughts on the use of randomization for inspection. Application to IAEA safeguards practices shows for the two approaches an improvement of the efficiency in the use of inspection resources and a reduction of intrusiveness in plant operation. For Lu, unpredictability and increased intensity of the inspection results in a strengthening of inspection effectiveness. According to Canty guaranteed detection times comparable or shorter to those obtainable without randomisation are achieved.

Because of the importance of this new concept the authors are warmly encouraged to pursue their studies on the use of randomised inspection scheme. Werkoff presented the first results obtained in France for the development of a knowledge-based software system for inspection strategy. The use of such software is expected to assist the IAEA in improving and optimising its inspection strategy, particularly in taking into account the whole fuel cycle of a state.

Session 2B: Safeguards systems and Implementation II

Five papers were presented in this session.

Two papers (2.5 and 2.6) described the State system for nuclear material accountancy and control in Czechoslovakia and Poland.
The nuclear facilities and the materials on which these systems are applied were described together with the inspection activities performed both by the national authorities and by IAEA.

The State office accountancy system was computerised in both countries and in the framework of the international agreement, a good cooperation exists between State and IAEA inspectors.

Some examples of the measurement techniques and instrumentation used were given together with the research and development activities carried out in order to improve the accuracy of the measurements and the effectiveness of the controls.

A nuclear material accountancy system in the industrial MOX fuel fabrication plant of Belgonucleaire was presented in paper 2.7.

In order to solve the problem of separate accountancy systems, for safeguards and clients, Belgonucleaire is using a system called "current account" in which materials are recorded in terms of Pu total, Pu fissile, Pu equivalent, U total and U_{235}. Advantages of this system are: traceability between fabrication and recycled materials from different customers; allowing a wider choice for cross-blending operations; avoiding interruptions of fabrication in case of temporary technical or administrative problems.

The data obtained by this "current account" system, referred to about 6 tons of Pu processed in the last five years, were used satisfactorily both for safeguards and commercial purposes.

The problems associated with the safeguarding of large plutonium stores presently under construction or recently commissioned were examined in paper 2.8.

Several features were analysed and evaluated from the safeguards point of view: reduction of personal access, improved remote operation, transmission of data to a central office, use of unattended safeguards systems, etc.

The conclusions were: an accurate installation of a properly designed NDA system, interlinked with the overall C/S system to maintain continuity of knowledge and in such a way that loss of continuity of knowledge is greatly reduced.

Furthermore, in order to have a reliable safeguards system, more development activities are required in particular in the area of authentication techniques and advanced surveillance equipment.

A survey of NDA instrumentation used routinely by the Euratom Safeguards Directorate was presented in paper 2.9 with the purpose of outlining the present and future needs for NDA instrumentation by comparing the types of nuclear materials and facilities in the European Community with the capability of existing NDA instrumentation.

The different NDA techniques based on passive and active neutrons and gamma measurements, were described and evaluated.

Further improvements were envisaged for irradiated LEU and HEU assemblies, U_{235} content of UF_6 cylinders and for techniques for measurements on wastes and scraps and for more sophisticated portable instrumentation.
Regarding large-scale facilities, it is necessary to include automated NDA stations able to measure 100% of the material flow at various parts of the plant. This automated approach requires a large amount of preparatory work before the equipment becomes operational.

Session 3: Measurement Techniques

A major change in the way that independent analysis of verification samples will be accomplished in the future was identified in a paper describing the design concept for an on-site laboratory for the Euratom Safeguards Directorate at the BNFL Sellafield site. Due to the vast increase in samples of special fissile materials required to be analysed for Safeguards purposes from the new large reprocessing facilities in France and the UK and the increasing difficulty of transporting such samples, it has become necessary to set up on site laboratories to carry out independent analysis at these plants. Of the many additional advantages that will be gained from on-site operation the most important is probably the considerable improvement in timelines of verification, with results obtainable within 48 hours.

A variety of techniques will be required for the analysis of input solutions, plutonium nitrate solutions and plutonium oxide samples. Well established titrimetric methods will be used for assay analysis and NDA techniques will be increasingly used where applicable. It will be essential to use automatic equipment and robotics to maintain a high throughput of samples with a minimum of staff.

The ECSAM laboratories will continue to have an important function in the future in analysing inspector samples taken from nuclear facilities in the EC and the current and future work of the ECSAM and PERLA laboratories were described in two further papers. Certain activities were highlighted; the production and certification of reference materials at JRC-Geel together with the responsibility for implementing the REIMEP programme, the development of automatic robotised methods of analysis at JRC-Karlsruhe and the development and testing of in-field methods of analysis for uranium at JRC-Ispra. PERLA has provided invaluable training courses for EC and IAEA Safeguards inspectors in the use of currently used NDA techniques as well as assessing the performance of a variety of NDA techniques employed in Safeguards. Further workshops for PNCC, calorimetry and software for NDA are planned for 1992.

The final paper given in the session described the features and possible applications of the highly sensitive time-resolved laser-induced spectrofluorometric techniques for the determination of elemental uranium, curium and americium. The technique is very rapid and suitable for such applications as; mapping uranium concentrations around nuclear sites, medical surveillance of staff and for the determination of uranium in TBP solutions from the Purex process.

Session 4: Measurement Techniques:
Destructive Analysis

G Bortels (4.1) This presentation covers the situation in alpha spectrometry. The most important points are: experimental parameters, peak distortion. The application to $^{238}Pu^{239+240}Pu$ was presented.

V F Kositsyn (4.2) The results of preparation and certification of three reference materials were presented. The certification of ^{233}U, ^{235}U, ^{238}U ratios to ^{236}U was done by analytical methods and by IDMS. The R.M. will be used in mass spectrometry as well as for calibration of mass spectrometers.

C. Nichol-Rostaing (4.3) Titanous sulphate and ferric nitrate have been proposed as substitution of titanous (III) chloride and ferric choloride. Experimental conditions to synthesize titanous sulphate have been optimized.

T K Li (4.4) The determination of Pu concentration and isotopic distribution has been carried out by gamma-ray spectroscopy. The results obtained by this study agree well with results by IDMS.

P DeBievre (4.5 & 4.6) The framework of REIMEP UF_6 measurement was presented. The results obtained by M.S. and gamma ray spectrometry have been compared. Calculated examples of both isotopic selective processes were presented.

Final Remarks In this session it is important to note the comparison of different techniques used for the determination of U and Pu. Alpha particle spectrometry gamma-ray spectrometry and mass spectrometry: results obtained with those techniques can be compared. Each one of them presents some advantages or inconvenience in relation to the others.

Session 5a: Measurement:Techniques Non
Destructive Assay I

This first session on Non-Destructive Assay concerned gamma spectrometry techniques (4 papers) and calorimetry measurements (1 paper).

The first paper presented the conclusions of the ESARDA interlaboratory PIDIE exercise on plutonium. The results showed the very important progress of the last years in the field of the gamma spectrometry methods, for detectors, electronics and software with the analysis possibilities in the X,γ region. Now, capability is near the DA performance for the determination of plutonium isotopic composition. Nevertheless, ^{242}Pu is not measurable by this technique and has to be determined by calculated correlations; improvements are necessary to obtain reasonable accuracy. A benchmark exercise on this subject in the framework of ESARDA NDA Working Group could contribute to improve the actual situation.

The second paper of SCK MOL concerned a large measurement campaign by gamma spectrometry on about 500 waste drums. Although this technique is interesting for safeguards measurements (simplicity, rapidity) and progress have been realised during these last years (additional gamma segmented scanning, interpretation software) its use stays limited for the homogeneous and low density material in drums.

The presentation by Canberra and the Safeguards Directorate of EURATOM (Paper No.3) showed the interest of the unattended gamma systems to improve the security and to decrease the cost of safeguards inspections. The development of counting electronics, software and computer architecture and networks must be actively followed in this direction.

In Paper No.4, the USSR team of Riga Scientific Research Institute presented recent improvements for the performances of Cd-Te detectors. These very compact detectors, which do not require a cooling system, have attracted a very high interest for gamma spectrometry measurements in the framework of safeguards control. It is now indispensable to obtain quickly industrial detectors with a resolution consistent with the measurement needs.

The last paper of this session presented the recent developments on the calorimeter with the aim of reducing the measurement time of this technique which is always attractive by its very good accuracies for Plutonium mass determination. Progress has been significant and the evidence of preheating and reduction of the measurement time below two hours makes the method acceptable for more safeguards applications.

Session 5B: Measurement Techniques: Non-Destructive Assay II

The session treated passive neutron coincidence techniques and, in particular, multiplicity theory and practice. The session (5B) was opened by Dr Hooton who gave a review of passive neutron coincidence techniques. The preparation of a review of this subject is a very positive initiative and it is hoped that it will be extended to other areas in safeguards.

The second paper was that of Dr Krick who discussed two different multiplicity counters constructed at Los Alamos. He showed that with very high efficiency counters (> 50%) they were able to verify the plutonium mass to better than ± 2% for a wide range of masses and for pure and impure plutonium. Practical multiplicity counters are proposed which may well indicate the way forward for future measurement systems for the verification of large plutonium masses.

A paper by Dr Carchon et al discussed an important step forward in the underwater verification of fresh LWR-MOX fuel elements. The paper discussed a passive neutron coincidence fork detector for underwater measurements. The detector is now available in Euratom Safeguards Directorate for inspection use.

In addition there were two papers from the Soviet Union, the first from Dr Janjushkin contributed to the multiplicity analysis (he attached a program listing to his paper to encourage people to provide him with feedback).

The second from A K Gorobets et al investigated the effect of the moisture on plutonium measurements by neutron coincidence counting: measurement results and interpretation were given, in this area which is of high concern in safeguards.

These last two contributions show that there a very significant amount of very interesting work being carried out in the Eastern bloc countries, which is a very encouraging sign.

In total the session showed that in the area of passive neutron coincidence counting there is a great emphasis on making big improvements in existing techniques and trying new developments, as is the case of multiplicity counters and time correlation analysis.

Session 5C: Measurement Techniques: Non-Destructive Assay III

A demonstration was given on plutonium mass evaluation in spent fuel by passive neutron measurement, with a precision of about 3% and on fissile mass content evaluation by active neutron measurements with a precision of about 5%. The method presented is a reasonable alternative to the FORK detector, for systems that can be permanently installed in spent fuel storage pools.

The development of a new thermal coincidence counter with a high efficiency, for general use in U/Pu facilities has been presented. Versatility and optimum performance were primary goals of this development.

The utility of the application of different techniques, related to NDA measurements on small U/Pu samples in various forms was described. NCC, γ-ray spectroscopy and calorimetry measurement techniques were involved, on old and less well defined materials.

Progress has been reported concerning the practical use of K-edge densitometers for the verification of product solution in reprocessing plants. Accuracies of about 0.2% - 0.3% at concentration levels typical for product solutions were obtained.

A paper was reported on the installation, the calibration, sample handling procedures and on the performance of a hybrid K-edge densitometer after 18 months of routine operation. The technique has achieved a high degree of maturity.

Session 6: C/S Posters

In the Surveillance field it can be seen that: the transfer from film cameras to video techniques has been successfully achieved and the next stage in this area, namely the use of digital video looks to be very encouraging, with the capability to achieve automatic data reduction, on site review and easy transfer of data to headquarters. Euratom's support and encouragement in these developments deserves recognition.

A further trend in the surveillance field was illustrated in the CONSULHA project with an integrated system using optical surveillance and NDA techniques to monitor nuclear material movements. Integrated systems become more feasible and more credible safeguards techniques and this trend will accelerate as a consequence of digital processing developments.

The limited contributions on seals technology could lead me to conclude that seals technology does not require further development and that all is well in this area. I cannot believe that to be the case.

Comments on the C/S Session Although limited to only four papers, they all illustrated that significant technological advances are being made in C/S. These will ensure that it will be possible to provide the assurance required in the future as C/S plays an increasingly important role in safeguards.

The development of the LASCO test and training facilities will enable C/S devices to be tested under laboratory and simulated-field-conditions in order to evaluate their performance. Additionally, training facilities will be available for safeguards inspectors. There is the opportunity here to improve the interface between inspectors and developers.

The two papers on integrated optical surveillance systems served to reinforce the comments made on the poster session concerning the major advances in surveillance recording and review techniques.

It was interesting to hear that not only have the fortunes of the ultrasonic sealing bolt been revived but also to know that there are encouraging signs concerning their performance in an operating environment.

Session 7: Spent Fuel Strategies

The contributions to the Spent Fuel Session as well as for posters on the subject demonstrate clearly that Spent Fuel Strategies which do not take reprocessing into account already play a deceptive part in safeguards work today.

Not only were design questions pertaining to a final repository discussed, but also the attempt was made to undertake a diversion path analysis for final repositories on the basis of a probabilistic analysis.

The paper presented on spent fuel conditioning discussed primarily systems-analytical questions especially the application of NDA and the introduction of so-called integrated verification systems.

The topic of reprocessing featured in two contributions. The successful completion of the benchmark exercise, undertaken by the RIV Working Group demonstrated an excellent of the ICT with the data acquired analytically.

With regard to input tank calibration it was reported that a considerable number of parameters still remain to be examined more closely before it can measure up to the standard achievable by analytical input determination.

Session 8: Materials Accounting and Data Evaluation

The papers presented in this session fell into three general classes:

(1) those papers describing conventional accounting systems.

(2) papers that sought to use accountancy information to provide plant management and the international safeguards authorities with additional assurance.

(3) papers exploring ways of applying advanced mathematical techniques to the problem of retrieving as much information as possible from a given data set.

Conventional accountancy is following the development of computer hardware and software in a most encouraging manner, the larger systems taking advantage of developments in operating systems and relational database management whilst in the smaller systems IBM PCs and clones, dominate. Near real time nuclear materials accountancy has matured with little further work in the development of statistical tests, the emphasis now being on the practical implementation of the technique.

GAMMA AND X-RAY MEASUREMENT OF NUCLEAR MATERIAL CONCENTRATION AND QUANTITIES

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Abstract

Gamma and X-ray spectrometric measurements of nuclear material concentrations and quantities are restricted because of the significant and unknown attenuation of gamma and X-ray radiations in the measured sample itself. However the measured gamma and X-ray spectra, used for isotopic composition determination contain also quantitative information about nuclear material concentrations and for "thin" samples, nuclear material quantities. The possibility to extract more information from the measured spectra has been successfully used for wide range of materials. Two methods are considered:

i. The use of normalized intensity of self-excited X-rays in order to determine the corresponding nuclear material concentration [1, 2].

ii. Spectrometric Gamma Absorption Method (SGA), when different attenuation of gamma rays with different energies is used as a measure of their effective attenuation [3, 4, 5];

1. Introduction

Nuclear radiation from nuclear materials excites X-rays radiation, which can be used to determine their concentrations - passive X-ray fluorescence analyses. The intensity of these X-rays is proportional to the total element concentration of the measured sample. However this intensity depends also on the intensities of exciting radiations, which are determined by the isotopic composition of the sample. As in most of the cases now, the main purpose of gamma and X-ray spectrometric measurements is to determine the isotopic composition of the measured sample, the same measurement can be used to normalize the intensity of self-excited X-rays and finally to determine the total element concentration of the measured sample as well.

The second possibility to measure the total element concentration of nuclear materials is through intensity ratio measurements of gamma and X-rays with significantly different energies [3, 4, 5]. If the thickness of the measured sample in the direction of measured gamma and X-rays is smaller than the quasi-infinite thickness for corresponding energy this measurement can be used to determine the effective attenuation of gamma rays in the sample and total quantities of nuclear materials.

2. Gamma and X-ray Measurement of U-total Concentration and U-235 Enrichment

This method is described in more details in IAEA report STR-264.

The 98.44 KeV X-ray line is the strongest one in the measured spectra of low enriched U materials. Its intensity determination is relatively easy because the line is well isolated from other gamma and X-ray lines. Therefor the normalized intensity of the self-excited 98.44 KeV X-rays is used as a measure of U-total concentration.

Only one measurement is required. The isotopic composition of the material and its radiations are determined by U-235 isotopic abundance (enrichment). The enrichment of the sample is determined either from the intensity of 185.7 KeV gamma rays of U-235 - enrichment type of measurements, or through intrinsic calibration measurements. The determined value of the enrichment is used to normalize the intensity of 98.44 KeV X-rays.

Simple linear dependence between the intensity of X-rays and corresponding U-235 enrichment or the intensity of 185.7 KeV gamma rays was established [2].

$$I(98.44) = K0 * I(185.7) + K1 \qquad (1)$$

The same function is used for normalization. Parameters of the function are determined through measurements of suitable standards of materials with the same U-total concentration (e.g U metal samples), with different enrichments and under the same geometry of measurements. The used measure of U-total concentration is determined by equation (2):

$$R = I(98.44)\mathrm{meas}/(K0 * I(185.7) + K1) \qquad (2)$$

A second degree polynomial is used for the dependence of the U-total concentration C(U) as a function of R:

$$C(U) = K00 * R\hat{\ }2 + K01 * R + K02 \qquad (3)$$

Our restricted experience to date shows that this dependence (3) is very close to linear. The parameters of the equation (3) are determined using suitable standards with different concentrations values covering the range of measured concentrations. Measurements under reproducible geometry are required. It is recommended also (optionally) to measure the thickness (or at least the uniformity of the wall) of the container at the place where measured gamma and X-rays are passing, using an ultrasonic gauge.

The results from concentration measurements done during our tests of the method are collected in table 1. We have measured four types of samples: four large U3O8 (about 2 Kg) samples ; four small U3O8 (about 0.2 Kg) samples; five (1 Kg) enriched UO2 standards; and five natural uranium concentration standards prepared in SAL (0.5 Kg).

The four large U3O8 samples covering U-235 enrichment range (0.2 - 20) % were used for determination of normalization function

$$I(98)n = K0 * I(186) + K1$$

In this case it was for U3O8 samples with 84.8 % U-total concentration. Using this function the normalization values for all samples were calculated. The ratios of measured I(98) values divided by normalization I(98)n values are given in column 3. The average values for different materials and corresponding standard deviations are expected ones

Table # 1

U-total CONCENTRATION MEASUREMENT RESULTS

SAMPLE	Enr	R	Cum/Cuk
U3O8(s)	0.708	0.9868	0.9809
U3O8(s)	1.94	1.0006	0.9952
U3O8(s)	2.95	1.0163	1.0123
U3O8(s)	4.46	1.0219	1.0165
Aver	1.0064	1.0012	
StDev(%)	1.37	1.42	
U3O8	0.225	1.0072	1.0005
U3O8	0.708	0.9870	0.9754
U3O8	3.105	1.0046	0.9968
U3O8	19.82	0.9999	0.9945
Aver	0.9997	0.9918	
StDev(%)	0.78	0.98	
UO2	19.88	1.0343	0.9984
UO2	35.00	1.0317	0.9925
UO2	45.55	1.0375	1.0051
UO2	60.06	1.0382	1.0030
UO2	92.42	1.0407	1.0034
Aver	1.0365	1.0005	
StDev(%)	0.31	0.46	
	Conc		
ST-1	82.35	0.9830	1.0028
ST-2	81.31	0.9726	1.0043
ST-3	73.44	0.8775	0.9960
ST-4	69.66	0.8307	0.9901
ST-5	60.38	0.7393	1.0070
Aver			1.0000
StDev(%)			0.61

The concentration standards were used for determination of concentration calibration function. It can be seen from the data that in some cases even linear dependence can be a suitable function. Then this calibration function was used to determine concentrations of all of the measured samples. The results divided by known concentrations are presented in column 4. Again average values for the types of materials and standard deviations for single measurements are given. Particularly interesting are measurements of UO2 materials from Ispra for two reasons:

i. The U-total concentration of this material is significantly outside the range of concentrations of the materials used for calibration of concentration measurements. The U-235 enrichments of these samples are outside the range of the enrichments of the samples used for determination of normalization function.

ii. These measurements were done in a different location and the calibration of concentration measurements from Vienna was used.

In spite of these unfavorable conditions the measurement results are quite reasonable.

One of the strong reasons for good results is the fact that the intensity ratios are used as measure of U-total concentration. This makes both measurements and analysis more reliable.

The following conclusions are results from measurements in Seibersdorf IAEA Laboratory, where different mainly low enriched U materials have been measured and in PERLA Laboratory in Ispra, JRC, where mainly high enriched samples have been measured.

i. The method is simple and reliable. The attainable accuracy for U-total concentration should be better than 1 % RSD at the one sigma level.

ii. The largest errors in the final results of these measurements and analyses (U enrichment and U-total concentration of the sample) usually come from the corrections for attenuation of gamma and X-rays in the wall of the measured material container. It is recommended to use gamma rays ratios for these corrections as general procedure, but it will be very useful to verify these measurements with a very simple ultrasonic gauge measurement of the container wall thickness at the measurement spot.

3. Intrinsic Calibration Gamma and X-ray Measurement of Pu Isotopic Composition and Pu-total Concentration

This paragraph describes preliminary results of development of a new method, which we consider as important.

Measurement of Pu-total concentration is of even more interest for international Safeguards, than U-total concentration. Pu is accounted in elemental quantities. If

Pu-total concentration is measured then for homogeneous samples the weight of the sample determines the total quantity of Pu. Again the normalized intensity of self excited X-rays has to be used. However in this case the strong intensity of X-rays is in a complex cluster of other strong lines. This is the first feature of Pu concentration measurements. Until recently it was not possible to measure the intensity of the X-rays. Due to the significant improvements of both the available hardware (Ge detectors and new amplifiers) and the software (the pioneer work of R. Gunnink [7] for analyses of the X-ray region), recently we succeeded in developing a computer program which extracts the intensity of 99.53 KeV X-rays of Pu reliably and accurately. This creates the possibility of a single measurement for both Pu isotopic composition and Pu total concentration.

The second feature of Pu concentration measurements is that the isotopic composition of Pu-Am-241 spectrum is much more complicated. Even more complicated is the spectrum of the Mixed Oxide (MOX) materials. Naturally its influence on Pu X-ray intensity is more complicated than in the case of U materials. Fortunately not all of Pu isotopes emit intensive radiations exciting Pu X-rays. The contribution of Pu-242 e.g. is definitely negligible. The specific exciting power of Pu-241 (exciting power per percent Pu-241 abundance) is by far the most significant. For high burnup Pu materials the total contribution of Pu-241 is in the range of 90 % from the total exciting power of all isotopes. As in addition it is relatively easy to measure Pu isotopic ratios with high precision it is possible to determine the normalization function with high precision.

The third feature of Pu concentration measurements is that the intensities of its gamma and X-rays in X-ray energy region are very strong. The Overall Relative Efficiency Dependence (ORED) on gamma and X-ray energies ORED(En) in this region is determined very precisely. Fortunately very close by in energy to the 99.53 KeV X-rays is the strong 101.07 KeV Np X-rays which is excited directly by Pu-241 and Am-241 decays. The contribution of Am-241 is equal to about 0.1 multiplied by Am241/Pu241 ratio. Therefor the intensity of this line, after small well known corrections accurately represents the Pu-241 abundance of the measured sample. And corrected ratio of 99.53 KeV line to 101.07 Kev line is a measure of the Pu-total concentration measured by intrinsic calibration method.

$$R = \frac{K * I(99.53) * (1 + .101 * R6) * ORED(101.07)}{I(101.07)*(1+K8*R1+K9*R2+K0*R3+K6*R6)*ORED(99.53)} \qquad (4)$$

Where R1 is N(Pu238)/N(Pu241) isotopic ratio
R2 is N(Pu239)/N(Pu241) isotopic ratio
R3 is N(Pu240)/N(Pu241) isotopic ratio and
R6 is N(Am241)/N(Pu241) isotopic ratio

K8, K9, K0, K6 are corresponding coefficient determined through measurements of standards.

Initial results from laboratory measurements demonstrated that the precision of these measurements can be in the order of 0.2 % - one sigma relative standard deviation for single measurements.

4. Simple, Reliable and Accurate NDA Method for Measurement of U-235 and U-total Quantities in MTR Fuel Assemblies

This method is described in more details in IAEA report STR-263.

The following features of the MTR fuel assemblies to be measured have to be taken into account, when considering suitable methods for measurements of U-235 and U-total quantities:

i. Material Test Reactor (MTR) fuel assemblies are large size items, length about 60 cm. and width about 10 cm, of highly enriched uranium (enrichment 20 - 93 %), but the quantity of nuclear material per unit length of fuel assembly and its area density (projection on the plane passing through the axes of fuel assembly) is relatively small (about 0.5 grams per cm^2). Most of U-235 gamma rays and U X-rays emitted by the assembly's nuclear material escape it without severe attenuation.

ii. There are several types of MTR fuel assemblies: parallel plates elements, concentric cylindrical elements, concentric square cross section elements and some variations of these types.

iii. It is difficult to have the range of suitable standards (the same type,size quantity and enrichment of nuclear material, cladding and so on) required for direct calibration of the measurements.

There are two existing methods of measurement of U-235 quantity and one method of measuring U-total quantity.

a. Gamma spectrometric measurements of U-235 quantity using the intensity of 186 KeV gamma-rays and gamma rays absorption measurements of U-total quantity, using the intensity attenuation of 186 KeV gamma rays from external assembly, by the measured assembly [8]. Because of the size of measured assembly another fuel assembly is used as a source of irradiating gamma rays. For U-total quantity measurements by this method three measurements are required:

i. "source gamma rays" assembly alone;
ii. measured assembly alone; and
iii. both fuel assemblies together.

Making three measurements is not convenient. In addition differential measurements are not generally accurate. These type of attenuation measurements are not applicable for some type e.g. cylindrical fuel assemblies.

The direct measurement of U-235 quantity through intensity measurement of 186 KeV gamma rays is more convenient when there are suitable standards for calibration, or other methods of taking into account the attenuation of gamma ray intensity by the material of the sample itself. However, the self attenuation measurement of 186 KeV gammas can be difficult.

b. Active neutron coincidence measurements [9].

Measurement of induced fission rate of U-235 nuclei through neutron coincidence counting rate measurements using Active Well Neutron Coincidence Counter. In this case:

- Only U-235 quantity can be measured relatively conveniently.
- Because of the large size of MTR fuel assembly only part of it can be measured at once. This complicates measurements and makes them less accurate.

Because gamma-ray, passive X-ray fluorescent and spectrometric intrinsic absorption measurement, when performed in a single measurement is significantly simpler, and potentially more reliable and accurate, there is little justification to use more complicated active neutron measurements for MTR type of fuel assemblies.

We propose a new method of direct measurement of the intensity attenuation of the gamma rays used for measurement of U-235 and U-total quantities. The intensity ratio of two strong gamma ray fluxes with significantly different energies is an precise measure of the effective attenuation of gamma rays by the measured sample.

The proposed method is to use the ratio of intensities of 186 KeV to 144 KeV gamma rays of U-235 as a measure of the effective attenuation of 186 KeV gamma rays intensity by the assembly material on their way to the detector. The 98.4 KeV X-rays to 185.7 Kev gamma rays intensities ratio can be used as additional useful (redundant) measure of U total quantity and 186 KeV intensity attenuation.

The reasons to propose this method are:

i. There is large difference in mass attenuation coefficients of 186 KeV (1.516 cm^2/g) and 144 KeV (2.869 cm^2/g) gamma rays in the uranium material of the assembly and their ratio is a sensitive and precise measure of 186 KeV gamma rays attenuation - see fig 1.

ii. This ratio can be calculated accurately using suitable model and numerical integration calculations using in addition some parameters from measurements of similar standards - see fig 2.

iii. The 98.4 K U-X rays are emitted by, and characteristic, for all U atoms. The intensity of this fluorescence line is a reliable quantitative measure of uranium presence.

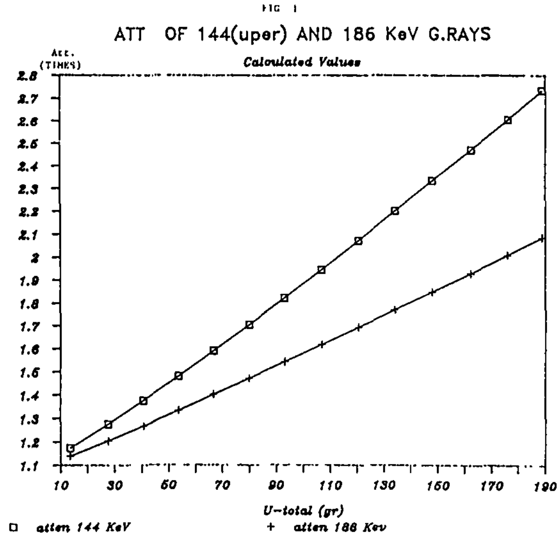

Figure 1

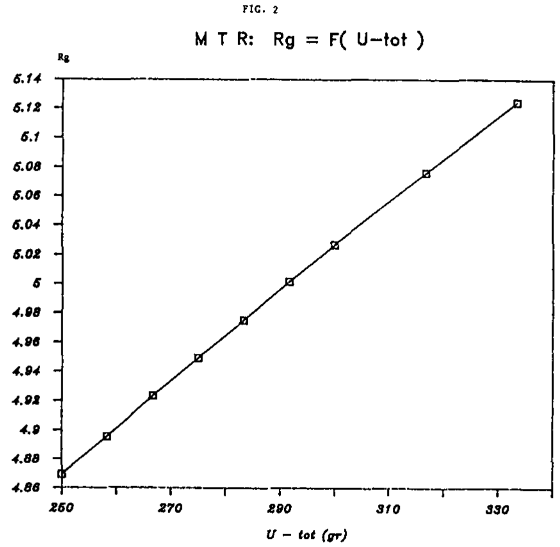

Figure 2

iv. The values of mass attenuation coefficients of 98.4 KeV X-rays and 186 KeV gamma rays in U materials are close. So the ratio of intensities of these two lines is also a measure of U-total quantity, area density, and effective attenuation properties of the whole assembly for 186 KeV gamma rays.

The measurement geometry should be such that registered gamma and X-rays pass the measured assembly by the shortest way. As the thickness of the material in this direction is significantly less than corresponding quasi infinite thicknesses, the intensities of gamma rays with energies higher than 84.2 KeV gamma rays depends strongly from the quantity of U-235 and U. Naturally from U-235 gamma rays the intensity of 186 KeV gamma rays was used as a measure of U-235 quantity. The corresponding equation is:

$$M235 = K235 * I(186) * Ka(Rg) \qquad (5)$$

where I(186) is the measured intensity of 185.7 (186) KeV gamma rays;

K235 is a calibration coefficient determined mainly by geometry of measurements and the efficiency of the used detector;

Ka(Rg) is a calibration function determined by the effective attenuation of 186 KeV gamma rays by the material of the measured assembly. For the method of measurements it is very important that Ka(Rg) is a simple function of the intensity ratio of 186 KeV and 144 KeV gamma rays of U-235;

M235 is total quantity of U-235.

FIG. 3

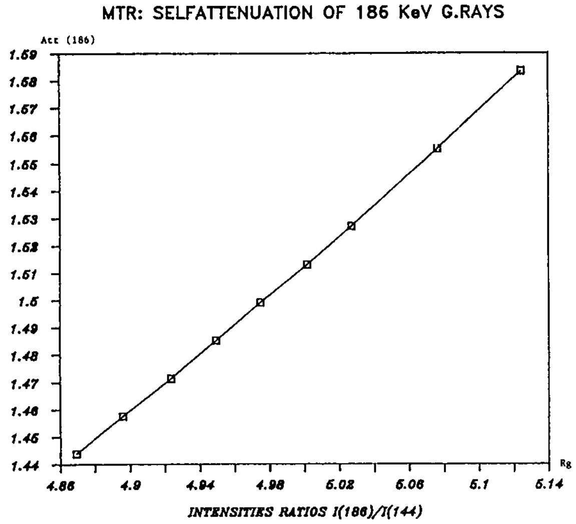

Figure 3

Calibration function Ka(Rg) can be determined using suitable standards or through numerical integration calculations. Calculated calibration attenuation function is shown on fig 3. Suitable function for interpretation of the calibration data is:

$$Ka = A0 + A1 * Rg + A2 * Rg \qquad (6)$$

Coefficients A0 , A1 , A2 are determined by calibration with suitable standards or by standards and calculations with computer programs based on models of measured and standard assemblies. We have developed such programs for parallel plates fuel assemblies and cylindrical assemblies.

As the intensity of 98.4 KeV U X-rays depends strongly on U concentration, and coefficients of attenuation of 98.4 KeV X-rays and 186 Kev gamma rays in U are practically equal, the total mass of U and the calibration function are simple functions of the intensity ratio Rx (Rx = I(98.4)/I(186)) values:

$$Ka(Rx) = B0 + B1 * Rx + B2 * Rx^2 \qquad (7)$$

$$M235 = K * I(186) * Ka(Rx) \qquad (8)$$

For samples with equal enrichment the total quantity of U is proportional to the total quantity of U-235. So the equations for total U will have the same general appearance:

$$Mu = Ku * I(186) * Ka(Rg) \qquad (9)$$

$$Mu = Ku * I(186) * Ka(Rx) \qquad (10)$$

Ku is a calibration coefficient for determination of U-total;

Ka(Rg) and Ka(Rx) are above described calibration functions determined by effective attenuation of 186 Kev gamma rays in the material of measured assembly;

Mu is the quantity of U-total in the assembly.

The calibration constants Ku and calibration functions Ka(Rg) and Ka(Rx), which are functions correspondingly of Rg and Rx can be determined using the results of measurements of suitable standards or results of computer calculations using corresponding models and programs.

Calibration functions Ka(Rg) and Ka(Rx) are similar but different functions. Calculation of the function Ka(Rx) is a more difficult task. At present it is more reliable to determine this function using suitable standards.

Calibration and Measurements

Calibration and measurements of parallel plates type of MTR fuel assemblies were done in Seibersdorf Safeguards Laboratory of IAEA. The available standard assembly was used for these measurements. The assembly is dismountable so it was possible to prepare many different combinations of plates in order to simulate different assemblies and to establish corresponding calibration functions. The length of plates is 60 cm, the width is 10 cm and they have slightly different but well known quantities of U, about 12 grams per plate of 80 % enriched U.

Having dimensions of the plates and assembly we have developed also a suitable model and computer program using numerical integration calculations. During the measurements of different "assemblies" the center of mass always was on the same distance from the front edge of the Ge detector.

In order to take into account the effective distance between the front edge of the detector and effective location of detected gamma-rays, special measurements were done. Using point multiple energy source and measurements at two different distances it was possible to determine the effective distances of detecting gamma rays with different energies. The parallel measurements and calculations of well known samples help to fix more accurately some of the parameters of calculations, particularly the effective detector efficiencies for different gamma and X-rays energies. As illustration of calculations and measurement precision part of the results are shown in table 2.

The first row of the table shows number of plates in assembly. The second row shows corresponding ratios of calculated to measured ratios of the 186 KeV intensities from the corresponding i- number of plates to the intensity from 7 plates.

Table # 2

No. of plates	8	9	10	11	12	13	14
(iI/17)c/ (Ii/17)m	1.001	1.002	1.010	1.007	1.006	1.004	1.004

As one can see from the table the agreement between the two groups of data - calculated and measured is quite good (differences are smaller than 1%).

Measurements of the U-235 and U-total quantities of cylindrical MTR fuel assemblies were done by IAEA inspectors in another country. Suitable standards were not available. The results from the calibration in Vienna and numerical integration calculations were used in order to take into account differences between standard and measured fuel assemblies on the site. The necessary model and corresponding program for cylindrical type of MTR fuel assemblies with corresponding sizes and parameters have been developed. Because the measurement system and particularly the detector were the same for standard and measured fuel assemblies the calculations effective efficiencies were the same. The results of calculations were very close to the measurement results. Differences were below 1 %.

Particularly careful measurements have been done on one of the assemblies. It has been accurately characterized and now is used as a secondary standard.

Having all these data and a secondary standard the standard procedure for routine safeguards measurements and corresponding computer programs for this type of MTR fuel assemblies have been prepared.

The procedure is based on recalibration measurements on the secondary standard. The measured spectrum is processed by the program F-CAL.BAS and it creates two small calibration files. One for energy calibration and second one for quantitative measurements of U-total and U-235 quantities. These files are generated at each new calibration.

The second program F.BAS automatically process the spectrum of the measured assembly and using calibration files determines U-235 and U-total quantities.

Results of the analysis including energy resolution of the system - FWHM(186) areas of corresponding gamma and X-rays and their ratios date and time of analysis are shown on the computer screen and recorded as a new result file with the same main name and extension "res".

Calibration measurements normally are done only once for given measurement system and type of samples. However if there is significant difference between measured and facility results calibration measurements should be repeated.

All results including measured spectra are recorded on magnetic cassettes and floppy disks, for additional analysis if required.

Some of the results of the last series of measurements are shown in table 3. In this case all measured assemblies were of the same cylindrical type. This allow some additional analysis to be done and to determine the overall precision of the measurements. In evaluating these results one should take into account that they are just routine field measurements. Attainable accuracy would be better.

Table # 3

Code	Rg	U5m/U5k	Utm/Utk
FG63	4.991	1.0022	1.0043
FG64	5.043	0.9986	1.0011
FG73	4.912	0.9911	0.9911
FG74	4.924	0.9903	0.9840
FG133	4.959	1.0162	1.0031
FG135	4.989	0.9985	0.9988
FG140	4.944	0.9937	0.9903
FG142	5.002	1.0076	1.0274
Aver	4.970	0.9998	1.0000
StDev(%)	0.88	0.88	1.32

The method has some general feature. It unites gamma spectrometric, passive X-ray fluorescence and spectrometric intrinsic absorption measurements. An important part of the method are precise models and computer programs for numerical integration calculations of gamma spectrometric measurements.

The method has the following advantages:

i. It is very reliable - a series of U-235 gamma rays are used for measurement of U-235, and U-total. They are characterized quantitatively by intensities and intensity ratios of U K-X rays and U-235 gamma rays, so there are no possibilities for substitution of nuclear material;

ii. The method is accurate. It unites possibilities of IAEA certified standards with possibilities of computational methods in order to take into account differences between standards and measured assemblies;

iii. The method is easy to use. It is passive, i.e. radioactive sources are not needed. Only one measurement is required in order to determine U-235 and U-total quantities. Processing of the spectra and all data is computerized.

The procedures for measurements of U-235 and U-total quantities in parallel plates and cylindrical types of MTR fuel assemblies have been prepared, tested and used by inspectors. The corresponding computer programs have been tested and used. This includes also special computational programs based on nuclear material item modeling and numerical integration calculations. Some of parameters used for calculations have been determined using measurements of standards.

5. Gamma Spectrometric Measurements of Pu-total Quantities in Solid Waste, Ash Samples and Rinse Solutions

Another good example of total Pu measurements by SGA method is measurement of Pu-total quantities in solid waste, contained in 30 liter drums, ash samples and rinse solutions. This method was described in details in IAEA report STR-56.

The intensity of 414 KeV gamma rays was used for quantitative measurement of Pu. The ratio of measured intensities of 414 KeV to 129 KeV gamma rays of Pu-239 are used as a quantitative measure of the effective attenuation of 414 KeV gamma rays by the measured sample itself. On site prepared working standards were used to calibrate the measurement system. The results of the NDA gamma measurements lie between the results of two destructive analyses results, which were available much later.

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P.S.
This paper was included in the Symposium programme as paper 3.1 but, because of unforeseen circumstances, it was not presented. The manuscript arrived when the proceedings were already being printed so that it is included at the end as an addendum.

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