COMMISSION
OF THE EUROPEAN
COMMUNITIES

JOINT RESEARCH CENTRE
Ispra Establishment
Italy

**CONFIDENTIAL
COMMUNICATION**

Category **1.3**

Nr **3584**

Copy № **226**

WARNING

The information contained in this document is communicated confidentially by the Commission of the European Communities to Member States, persons and undertakings and should not be passed on to third parties.
(Euratom-Treaty, Article 13, and Regulation (EEC) No. 2380/74 of the Council of Ministers).

Management of Nuclear Materials and Radioactive Waste

PROGRAMME PROGRESS REPORT

July - December 1978

Programme Progress Report JRC Ispra
July - December 1978

MANAGEMENT OF NUCLEAR MATERIALS AND RADIOACTIVE WASTE

Abstract

This document is the progress report of the programme Management of Nuclear Materials and Radioactive Waste of the Joint Research Centre for the period July-December 1978.
The programme consists of three projects.
The main achievements during the reporting period were the following:

Project 1 : Evaluation of the long-term hazard of radioactive waste disposal

The model developed for the evaluation of the probabilistic value of the geological barrier, is being applied to a clay formation.
The model developed for the calculation of pathways and dose rates to man has been applied to the assessment, for different fuel cycle strategies, of the long-term risk of the radioactive waste disposal in geological formations.
The examination of glass samples irradiated in a nuclear reactor, to simulate the long-term damage produced by alpha-particles in vetrified waste, have been completed.
Experiments on plutonium migration in sand columns have been carried out.
The collaborative programme for an Integral Experiment on the plutonium waste measurement system of the Dounreay Nuclear Power Development Establishment (DNPDE) has been started.

Project 2 : Chemical separation and nuclear transmutation of actinides.

Studies have been performed to determine the experimental conditions which make possible to keep plutonium in an extractable form during the operations of concentration and denitration of the HAW solutions.
Tests have been carried out on various steps of the OXAL process at fully active scale.
For the assessment on the nuclear transmutation of actinides progresses have been made concerning reactor physics calculations, fuel element design, evaluation of risk and technological implications on the nuclear fuel cycle.

Project 3 : Decontamination of reactor components

Analyses using scanning electron microscopy (SEM), Auger electron spectrometry (AES) and secondary ion mass spectrometry (SIMS) have been performed in order to investigate the physico-chemical structure of oxide layers and the mechanisms of chemical decontamination.

JOINT RESEARCH CENTRE
Ispra Establishment
Italy

Management of Nuclear Materials and Radioactive Waste

PROGRAMME PROGRESS REPORT

July - December 1978

BEMÆRK

Den viden, som rummes i dette dokument, meddeles som fortrolig fra Kommissionen for de europæiske Fællesskaber til Medlemsstater, personer og virksomheder og må ikke videregives til trediemand. (Euratomtraktatens artikel 13 og Minersterrådets forordning(EØF) N° 2380/74).

Hverken Kommissionen for de Europæiske Fælleskaber eller nogen, som optræder på Kommissionens vegne er ansvarling for den eventuelle brug af information, som er indeholdt i det følgende.

ZUR BEACHTUNG

Die in diesem Dokument enthaltenen Kenntnisse werden von der Kommission der Europäischen Gemeinschaften den Mitgliedstaaten, Personen und Unternehmen der Gemeinschaft vertraulich mitgeteilt und dürfen nicht an Dritte weitergegeben werden. (Euratom-Vertrag, Artikel 13, und Beschluss des Ministerrates (EWG) Nr. 2380/74).

Weder die Kommission der Europäischen Gemeinschaften noch Personen, die im Namen dieser Kommission handeln, sind für die etwaige Verwendung der nachstehenden Informationen verantwortlich.

NOTICE

The information contained in this document is communicated confidentially by the Commission of the European Communities to Member States, persons and undertakings and should not be passed on to third parties. (Euratom-Treaty, Article 13, and Regulation (EEC) No. 2380/74 of the Council of Ministers).

Neither the Commission of the European Communities nor any person acting on behalf of the Commission is responsible for the use which might be made of the following information.

AVERTISSEMENT

Les connaissances contenues dans le présent document sont communiquées confidentiellement par la Commission des Communautées européennes aux Etats membres, personnes et entreprises et ne peuvent être transmises à des tiers. (Traité Euratom, article 13, et règlement (CEE) N° 2380/74 du Conseil de Ministres).

Ni la Commission des Communautés européennes, ni aucune personne agissant au nom de la Commission, n'est responsable de l'usage qui pourrait être fait des informations ci-après.

AVVERTIMENTO

Le cognizioni contenute nel presente documento sono comunicate confidenzialmente dalla Commissione delle Comunità europee agli Stati membri, persone ed imprese e non debbono essere trasmesse a terzi. (Trattato Euratom, articolo 13, e regolamento (CCE) N° 2380/74 del Consiglio dei Ministri).

Né la Commissione delle Comunità europee, né alcuna persona che agisca per suo conto, è responsabile dell'uso che dovesse essere fatto delle informazioni che seguono.

OPMERKING

De kennis, die in dit document is vervat, wordt door de Commissie van de Europese Gemeenschappen vertrouwelijk aan de Lid-Staten, personen en ondernemingen medegedeeld en mag niet aan derden worden doorgegeven. (Euratom-Verdrag, artikel 13, en het besluit van de Ministerraad (EEG) No. 2380/74).

Noch de Commissie van de Europese Gemeenschappen, noch de personen die namens haar optreden, zijn verantwoordelijk voor het gebruik, dat eventueel van de hiernavolgende informaties wordt gemaakt.

CONTENTS

1. INTRODUCTION

The safe and economic management of the radioactive waste, produced in the exploitation of nuclear energy at an industrial level, requires a considerable effort of R&D.

The Joint Research Centre (JRC) started work in the field of radioactive waste management in 1973. This programme is part of the activity of the JRC in the field of Nuclear Safety which includes also the programme Reactor Safety and the programme Plutonium Fuel and Actinide Reasearch. The staff allocated to the programme for 1978 consists of 63 research men, corresponding to about 6% of the total JRC-staff. The programme is carried out at the Ispra Establishment with a participation of the Karlsruhe Establishment.

The JRC programme Management of Nuclear Materials and Radioactive Waste has been oprganized into three projects:

Evaluation of Long-Term Hazard of Radioactive Waste Disposal

comprising essentially the identification and the evaluation of the long-term hazard of the permanent storage of radioactive waste in geological formations. This type of storage is considered at present to be the most appropriate to solve the problem of radioactive waste.

Chemical Separation and Nuclear Transmutation of Actinides

The objective is to obtain a better appreciation of this advanced strategy for managing radioactive waste by separating the actinides responsible for long-term risk, from the bulk of the fission products and by their transmutation in nuclear reactors.

Decontamination of Reactor Components

The objective is to study the nature of the contaminated layers and the application of various decontamination techniques in order to optimize the decontamination procedures required for the safe operation and for the decommissioning of nuclear reactors.

The Commission of the European Communities started in 1975 an Indirect Action in this field. In this Indirect Action, which is conducted by means of contracts with national laboratories, various aspects of waste conditioning technologies are studied and a large coordinated action for the study of waste disposal in various types of geological formations was established.

The two programmes complemented each other rather well and the coordination between them and with national activities was assured by appointing one Advisory Committee for Programme Management (ACPM) for the two actions.

Projects

2. PROJECTS

2.1. The Evaluation of Long Term Hazard of Radioactive Waste

The long-term hazard of radioactive waste disposal in geological formations, which is largely due to the presence of actinides, is studied by the barrier approach based on the evaluation of the barriers provided between the disposed waste and man.

The barriers considered are the following:

- Segregation provided by disposing the waste in a deep geological formation
- Long-term stability of the waste conditioned in glass and bitumen
- Retention of actinides by geological media
- Ecological distribution pattern of actinides.

The evaluation of the long-term hazard requires the development and application of waste hazard analytical models and experimental studies for the quantification of the values of the different barriers.

In the field of models development we are passing from generic models in which the data are arbitrarily set on the basis of scientific considerations, to a more applied type of development in which the data are collected on specific experimental sites, not necessarily linked to any future disposal operation.

Concerning the experimental studies on the long-term stability of the conditioned waste, both radiation damage studies on glasses and studies on the leaching of vitrified and bituminized waste are in progress.

Experimental studies on actinides distribution in the environment relate to the chemico-physical interactions between leached out actinides and geological media. The interactions between the actinides and the biosphere are studied essentially by means of theoretical models, taking advantage of the large amount of experimental data generated by the indirect programme Radiation Protection of the Commission. Joint actions are being set up to strenghten the links between the two programmes.

Waste Hazard Analysis

OBJECTIVES

Aim of the study is to develop a methodology for the assessment of the risk linked to the disposal of different kinds of radioactive wastes into geological repositories.
Probabilistic and deterministic approaches are used, in function of the nature of the barriers considered; for the probabilistic approach, the Fault Tree Analysis method is utilized, thus treating the geological barrier as a binary system, which can only be either in functional or in failed state; for the deterministic approach, the potential levels of environmental radioactive pollution and the corresponding dose-rates to man are calculated, through development of appropriate physical and mathematical models.

For the second semester of 1978 the planned activities were:

A. Completion of the application of the Fault Tree Analysis to a specific experimental site (Belgian clay formation).
B. Refinement of generalized Model 1[1,2]; its application to different fuel cycle strategies.
C. Application of data histogram treating techniques.

RESULTS

The methodology is now well established in its details, for both the probabilistic and the deterministic sections.

A At present, the probabilistic section of the model is being applied to a specific site (the Belgian clay formation of Boom) to test the applicability of the Fault Tree Analysis to a real case. This work is being performed in close collaboration with the C.E.N. of Mol, to qualify the failure probabilities of a repository mined into a clay formation, as a function of time.
An upper section of the «tree» is shown in Fig. 1 where releases to three main receptors are considered.

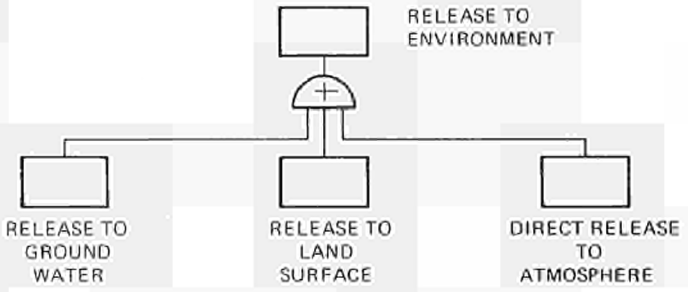


Fig. 1 — Upper Section of the Fault Tree

Each release to a particular receptor develops into a detailed «Tree», down to various sets of primary events, which are listed in Table 1.
Probability values of occurrence for three different time intervals (2,000, 30,000 and 100,000 years) are being assessed for all the primary events.
Probability data in the form of hystograms can be handled by computer codes specially developed at the JRC Ispra for fault tree treatment.

B The previous generalized Model 1 for the deterministic assessment of environmental pollution levels as a consequence of a containment failure has been improved by introducing a more detailed inventory of conditioned wastes, and by developing a leaching model for each waste type; as a rule, we have:

$$R_i = f(\sigma, l, C_i, Q)$$

where:

R_i is the release rate of isotope i,
σ is the specific surface of the waste
l is the leaching rate
C_i is the concentration of isotope i
Q is the quantity of waste undergoing leaching.

Further, a more detailed environmental system has been defined, which takes into account different pathways to man.

Table I — List of Primary Events

1) 2)	Presence of ground water above Presence of pressurized ground water below	GROUND WATER
3) 4)	Migration with retention (outside the clays) Migration without retention (outside the clays)	RADIONUCLIDE RETENTION
5) 6) 7)	3rd order Fault 2nd order Fault 1st order Fault	FAULTING PHENOMENA
8) 9)	Extrusive magmatic activity Explosive magmatic activity	MAGMATIC ACTIVITY
10)	Diapirism (2nd order only)	DIAPIRISM
11) 12) 13) 14)	Glacial overburden (loss of plasticity) Permafrost 1st order glacial action 2nd order glacial action	GLACIAL PHENOMENA
15) 16) 17) 18) 19)	Subsidence (2nd order only [loss of plasticity] 1st order stream erosion 2nd order stream erosion 1st order denudation 2nd order denudation	GEOMORPHOLOGIC PROCESSES
20) 21) 22) 23)	Pumping of contaminated ground water Drilling exposing water to aquifers 1st order human activities causing waste rise 2nd order human activities	HUMAN ACTIONS
24) 25)	1st order meteorite - causing release to atmosphere 2nd order meteorite - causing release to ground water	METEORITES

This model has then been applied to an hypothetical waste repository, to compare two different fuel cycle strategies on the basis of their long-term risks: they are the once-through strategy and the uranium and plutonium recycle strategy in LWRs.

The following conclusions were reached:

1) The long-term risk linked to the once-through strategy is about two orders of magnitude higher than that due to the recycle strategy since, in the latter option, the plutonium destruction largely exceeds the build-up of transplutonium isotopes.

2) Should the release occur within a few thousands of years after repository closure, the risk would be governed mainly by inhalation of plutonium isotopes; on the other hand, if the release should occur after more than 10^5 years, Ra^{226} will be the risk-governing isotope.
Ra^{226} if formed from the decay chain $Pu^{238} \rightarrow U^{234} \rightarrow Th^{230} \rightarrow Ra^{226}$. The conclusion can be drawn that a better control of plu- tonium losses into any waste kind could decrease substan- tially the risk at any time.
Besides, in the case of Mox-fuel, the presence in the waste of Cm^{242} parent of Pu^{238} is the major source of Ra^{226} build up in HLW.

3) These conclusions are valid only in the case of no soil retention by the geochemical barrier; otherwise, neptunium, technetium and iodine isotopes could become the dominant nuclides, but at a substantially lower level important.
A survey of the existing models describing the environmental behaviour of radioactive pollutants has been performed[3].

C The methodology of treating large data uncertainties by probability distributions has been described in the previous Programme Progress Report[4]. This methodology has been completed by establishing functional relations between risk and input data uncertainties. An analytical relationship can thus be obtained which shows the influence of data uncertainties on the results. This method could serve especially to identify those variables which have the greatest impact on the risk. A paper is in preparation[5].

COLLABORATION WITH EXTERNAL ORGANIZATIONS

- The collaboration with the C.E.N. - Mol has been already mentioned.
- A collaboration with the indirect action programme Radiation Protection has been established for the development of environmental models and the related radioecological research.

CONCLUSIONS

The methodology developed at the JRC - Ispra for the assessment of the long-term risk is ready and available. The applicability of its probabilistic section to a specific site is being verified, and preliminary results are satisfactory. A similar application for the deterministic section of the model is planned, with the objective to identify, for specific site parameters, what are the experimental studies which can better contribute to decrease the uncertainty of the risk evaluation.

PLANNED ACTIVITIES

The Fault Tree Analysis for the experimental site in the Belgian clay formation will be completed.
A site-specific deterministic analysis will be initiated during 1979.

REFERENCES

1 F. GIRARDI, G. BERTOZZI, M. D'ALESSANDRO: «Long-term Risk Assessment of Radioactive Waste Disposal in Geological Formations»
Report EUR-5902 (1978)

2 Management of Nuclear Materials and Radioactive Waste — Programme Progress Report of the Joint Research Centre, January-June 1977, N. 3440

3 G. BIGNOLI, G. BERTOZZI: «Modelling of Artificial Radioactivity Migration in Environment: ASurvey».
To be published as EUR-Report

4 Management of Nuclear Materials and Radioactive Waste - Programme Progress Report of the Joint Research Centre, January-June 1978, No. 3528

Long-Term Stability of Conditioned Waste

OBJECTIVES

The aim of this study is to obtain information on long-term behaviour of conditioned high- and medium-level waste, in the framework of studies of Waste Hazard Analysis.

The planned activities for the second semester of 1978 were the following:

- Completion of the post-irradiation examination of the glasses, irradiated in the HFR reactor, at Petten,
- Continuation of the glass leaching tests,
- Extension of the stability tests to bituminized waste,
- Verification of accelerated tests of radiation damage on vitrified high activity waste.

RESULTS

Post-Irradiation Examination of Irradiated Glasses

The irradiation carried out in the HFR at Petten on glass samples containing uranium was intended to simulate, by the damage caused by fission frangments, the long-term damage produced by the decay of alpha-emitting nuclides in vetrified HAW.
A maximum of fission density of 4×10^{17} fissions/cm^3, which corresponds to 2×10^{22} displaced atoms/cm^3 was calculated for the irradiated samples.
This fission density corresponds to a damage period for vetrified HAW of about 10^5 years.

During the reporting period the post-irradiation analyses of the glass samples have been completed with the measurement of the stored energy. Measurements of leaching rates and densities had been previously carried out on the irradiated samples.
The stored energy values are reported in Fig. 1.
These values are in good agreement with those measured in radiation damage experiments carried out in other laboratories using glasses loaded with alpha-emitters.
Thus it can be concluded that the simulation of alpha-damage using fission fragments gives realistic results.
A final report on the experiment is in preparation.

Glass Leaching Tests

The aim of the glass leaching tests is the evaluation of the long-term weight loss and the systematic study of the surface layer composition in order to clarify the leaching mechanism.

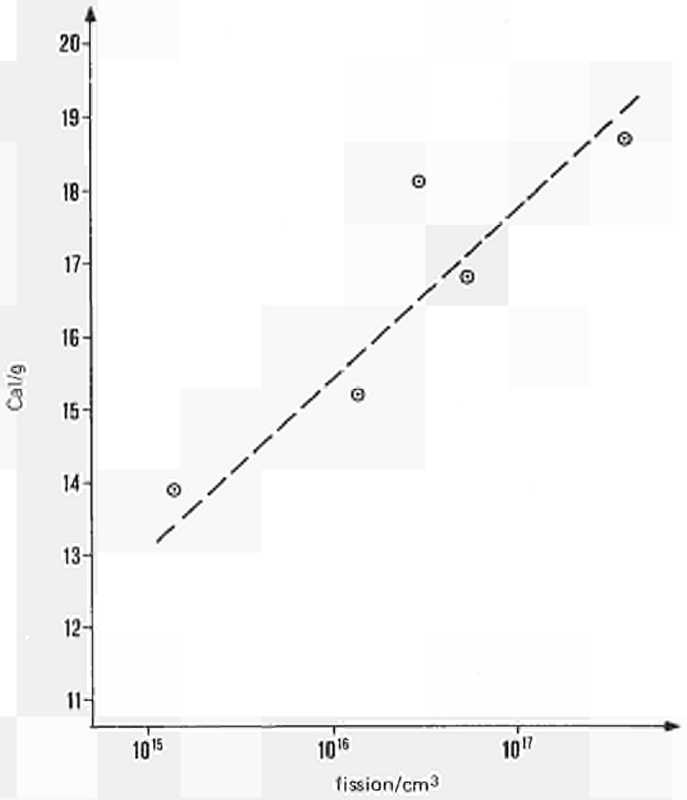


Fig. 1 — Stored Energy Measured in Glass Samples Irradiated in the HFR Reactor (Energy Reased up to 550 °C)

During the reporting period the analysis of the leached surfaces has been completed, in connection with leaching tests in pure water of 1 year duration at 80°C and 50°C.
The results of the analysis of the leached surface show that in the initial period most of the leaching is due to a solution mechanism while in a second time the weight loss due to release of microcolloids is predominant.
It can be concluded that, due to this mechanism, the assumption of an homogeneous leaching of the glass seems confirmed.
A final report on the experiment is in preparation.
Experiments are in progress to study the influence of the water composition on the leaching rate.

After the completion of the leaching tests using water conditioned by percolation through a silica sand, a new series of tests has been initiated using water which was previously contacted by clay.

Stability Tests on Bituminized Waste

Stability tests on simulated bituminized waste are in progress in order to verify the dependence of leaching on the solubility of the bitumen-incorporated salt and to clarify the mechanism of the long-term leaching.
The tests have been delayed by difficulties encountered in the preparation of homogeneous salt-bitumen mixtures.
Fig. 2 shows the surface of a sample loaded with 40% $NaNO_3$ after leaching with HCl for 24 hours. White dots represent small cavities from which the crystals of $NaNO_3$ were leached. The distribution of the crystals appears to be quite homogeneous.

Verification of the Validity of Accelerated Tests of Radiation Damage on Vetrified High-Activity Waste

Measurements of optical absorption on silica glasses irradiated with protons or heavy ions have been initiated, as planned, in order to gain a better knowledge of the defects introduced by irradiation and of their annealing behaviour.

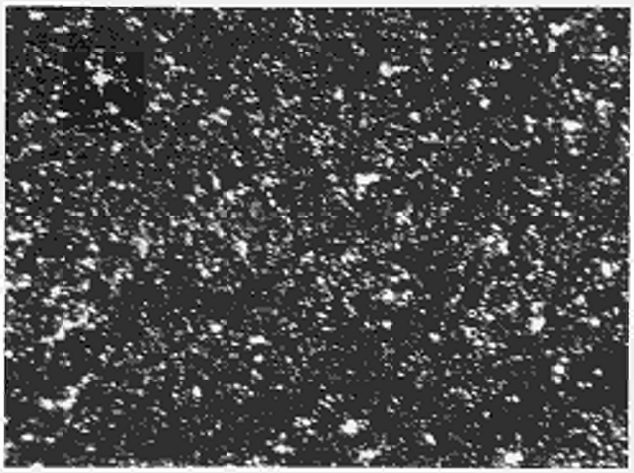
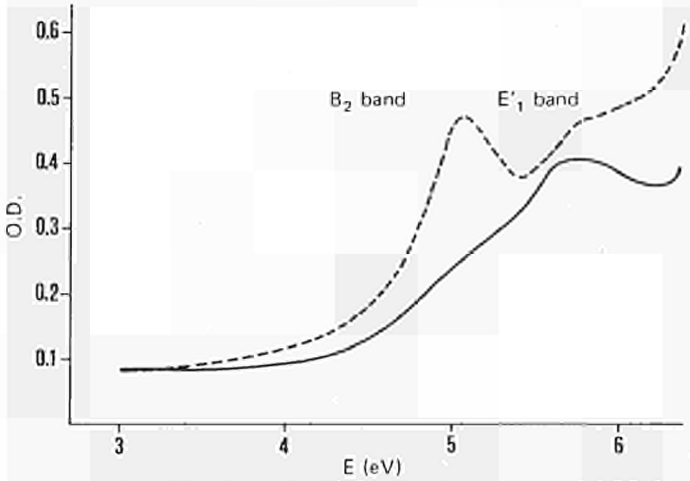


Fig. 2 — Metallographic Photograph of the Surface of a Leacted Bitumen — 40% $NaOH_3$ Samples — Showing the Homogeneity of the Distribution

The specificity and the greater sensitivity of spectral absorption determination provide the necessary complement to microcalorimetric studies, for detection and monitoring of various types of damage.

Absorption measurements cover systematically the range from ~1900 Å to ~25,000 Å, with special attention for the u.v. region, where the dominant products of damage are known to give more specific effects.

Interesting results have been obtained by comparing optical effects in samples of pure SiO_2 irradiated with light or heavy ions. The effects are illustrated in Fig. 3, where the intensities of the so called B_2 - and E'_1 - bands obtained with proton or Ni^{6+} irradiations of identical samples are shown. The first of these bands, which is ascribed to an electronic vacancy centred around a real atomic displacement, is strikingly different in the two cases, being almost negligible in the proton-bombarded samples where the total damage content in D.P.A. is greater. This effect confirms previous findings, i.e. the displacement products are stabilized in a form that depends drastically on the way of production for equal numbers of displacements. In the case of protons such displacements are formed by small cascades; the opposite applies with respect to heavy ions. These considerations point to a possible role of complex self-annealing and radiation-annealing effects, of obvious importance for better evaluation of simulation tests.

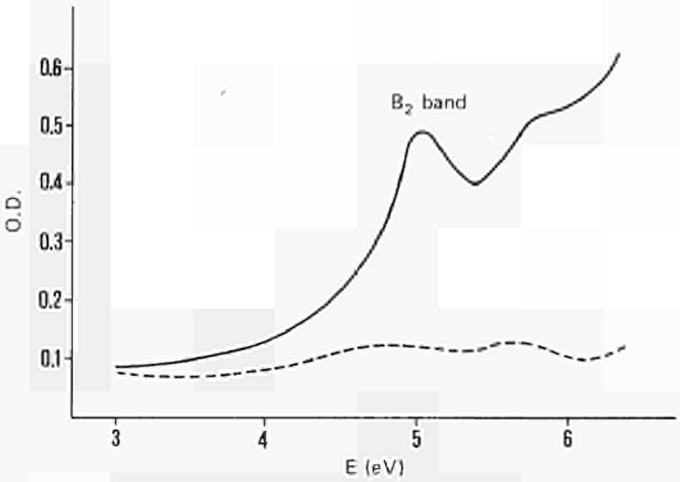


– dotted line: heavy ion Ni^{+6} (46.5 Mev, total dose 0.1 DPA)
– full line: protons (1 Mev, total dose 0.2 DPA)

Fig. 3 — Optical Absorption Measurements on Heavy Ions and Protons Irradiated SiO_2

A preliminary series of post-irradiation annealings, gave also rise to interesting observations.

Fig. 4 shows an example of complete annealing of the B_2 displacement centre which is obtained after heating at 650°C. Intermediate observations showed that partial destruction is already effective at 300°C. The relatively low range of temperatures in which displacement damage is removed gives a hint for further investigations of thermal effects.



– full line: after heavy ion irradiation (46.5 Mev, total dose 0.1 DPA)
– dotted line: after annealing at 650 °C

Fig. 4 — Annealing Effect at 650°C in Ion-Bombarded SiO_2

COLLABORATION WITH EXTERNAL ORGANIZATIONS

The collaboration with AERE, Harwell, concerning irradiation experiments and theoretical investigations, has been continued.

PLANNED ACTIVITIES

For the first semester of 1979 the following activities are planned.

Radiation Damage on Glasses

The results obtained in the irradiation experiments in the HFR have shown the validity of the damage simulation by fission fragments.

This method has the advantage to make possible the simulation of extremely long damage periods without increasing considerably the experimental effort.

Thus a new irradiation experiment has been planned (BONI III) where a fission density of 2×10^{18} fissions/cm³ will be reached.

The fission density corresponds to a damage period for vetrified HAW of about 10^6 years, value which seems very difficult to reach with the loading of glasses with alpha-emitters.

Leaching Tests

The leaching tests on glasses and bitumen will be continued.

Verification of the Validity of Accelerated Tests of Radiation Damage on Vitrified High Activity Waste

Another series of heavy ions irradiated samples (SiO_2 and borosilicate glasses simulating HAW) will be measured by means of optical and calorimetric techniques with the following aims:

1) To correlate the calorimetric results previous obtained with the optical measurements to get a sight of all defects formed.
2) To study the annealing as a function of temperature and impurities present in the samples.
3) To study the dipendence of stored energy on the type of

Applying the above relation we extended the original conduction model by

1) first computating the Rayleigh number associated with the pool from equation (1)
2) in case convection occurs, i.e. $Ra_{k_e} > 580$, the Nusselt number Nu_1 is evaluated from equation (4)
3) substituting the conductivity value λ in the upper layer by a new effective value according to equation (9).

Interaction of Actinides with the Environment

OBJECTIVES

The objective of this study is to obtain an understanding of the interaction of actinides with geological media and groundwaters following their eventual leaching from vitrified high level waste stored in geological formations.

For the second half of 1978, the planned activity was to pursue the experiment on column migration and to analyze the size and charge of the chemical species of actinides liberated from the leached glass. Also experiments under high lithostatic and hydrostatic pressure were foreseen.

RESULTS

Experiments have been performed to evaluate the consequences of an accidental release from a clay formation taking as an example the geochemical conditions existing around the Boom formation in Belgium.

A water pathway has been established which flows over the alpha-bearing glass prepared in the laboratory, and then through columns containing soil samples.
The set-up adopted is illustrated in Fig. 1. The elution behaviour in the case of a pure sand column is shown in Fig. 2. No retention (e.g. filtration or absorption) seems to occur in this case.

The experiment with the Belgian subsoils are still in progress with water composition typical of the aquifer of this zone.

The soil contamination profile of the Pu and Am activity leached over several months from glass will be determined and presented in a next report.

Parallel to these experiments, investigations of the chemical species of actinides produced by the leached glass have been initiated. The Fig. 3 shows the set-up adopted: in this case a water pathway has been established which flows over crashed and sieved Pu-bearing glass and then through an ultrafiltration system followed by cationic-anionic resins.

During a 34 days experiment the total activity measured was 1.58μC, of ^{238}Pu, corresponding to a leaching rate of 4.8 x 10^{-7} g/cm^2 x day. The ultrafilter retained 20.1% of the leachate activity while 60,6% and 19,0% were found on the cation and anion exchange resins respectively. 0,3% was retained on the fraction collector.

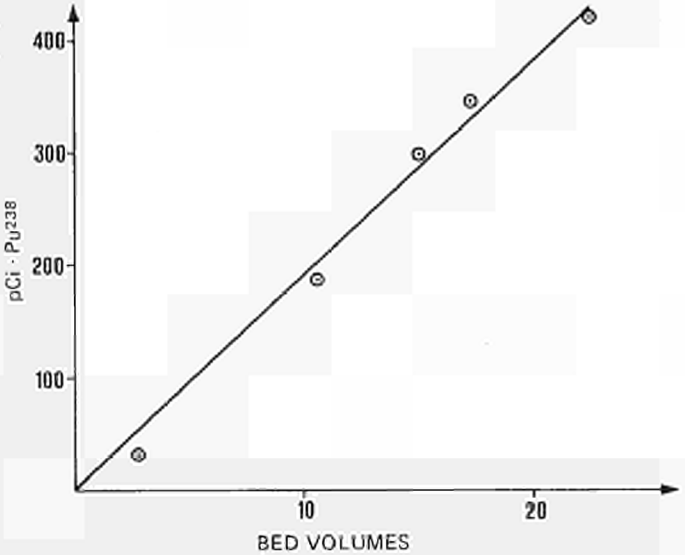


Fig. 2 — Elution Behaviour of a ^{238}Pu-Doped Glass Leachate on a Sand Column Glass Leaching Rate 17 · 10^{-7} g/cm^2 Day

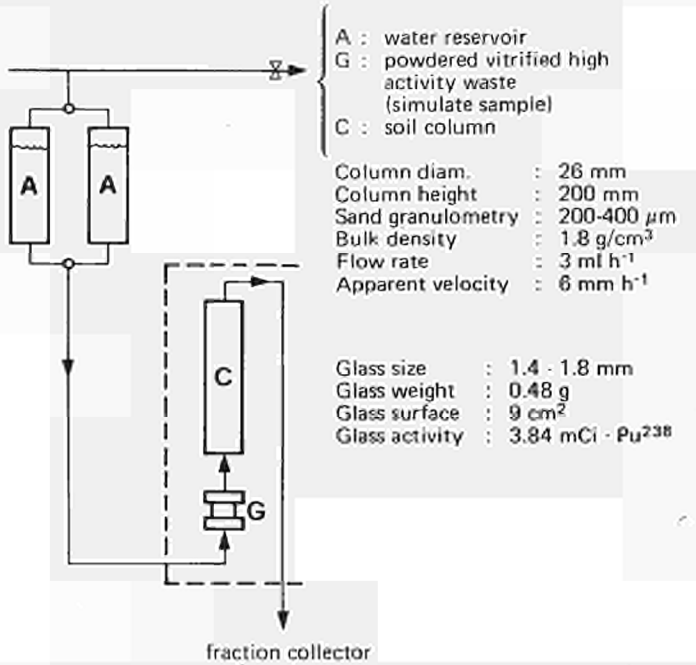


Fig. 1 — Experimental Set-Up for Studying the Interaction of Glass Leachates with Deep Soil

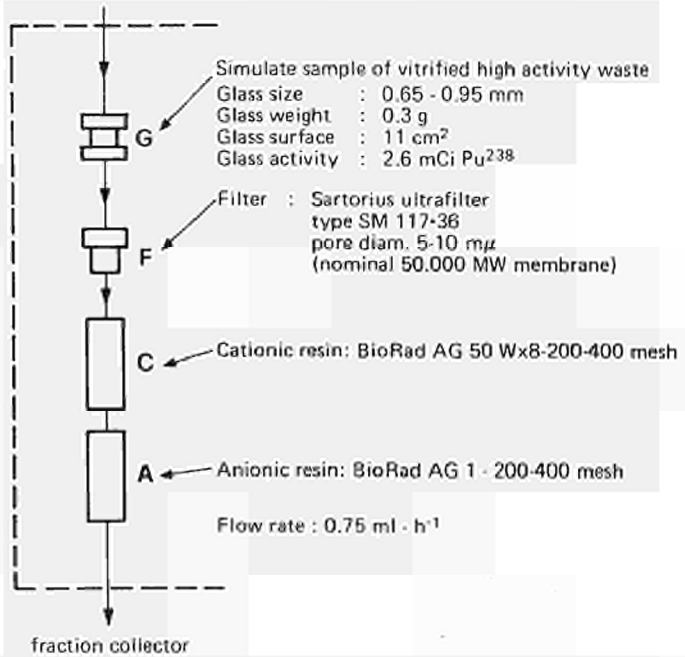


Fig. 3 — Experimental Set-Up for Studying Chemical Forms of Glass Leachates

CONCLUSION AND PLANNED ACTIVITIES

The long term experiments in progress will be continued with plutonium and americium isotopes.

Parallel to columns experiments, an exploratory programme will be started on the use of several chemical analytical techniques which appear promising for valence state determination of plutonium in very diluted solution.

Experiments will be also undertaken to confirm the applicability of such methods to simulated aquatic surface ecosystem where chemical speciation may play an essential role in altering the biogeochemical behaviour of these elements, thus affecting their fate and distribution in the environment. As previously reported, this development is being carried-out in close cooperation with the indirect programme Radiation Protection.

Preliminary experiments under high lithostatic pressure have been undertaken in order to determine the types of problems that will arise on application of these techniques for radioisotopic migration studies. An experimental set-up designed to withstand operating pressures up to 20 - 25 atm. is being constructed.

Actinides Monitoring

OBJECTIVES

The study aims at the development of a methodology for plutonium waste monitoring. For the reporting period theoretical and experimental work was planned concerning development of reference instruments and methods in the framework of an Advisory Laboratory.

RESULTS

Reference Monitor for Passive Gamma Assay

This monitor is described in Chapter III of our guide[1]. The assembling of the apparatus was completed and its text is in course. The experimental verification of the interpretational model for the reference monitor as outlined in the guide, will be started after the completion of the test.

Upgraded Reference Monitor for Passive Neutron Assay

This monitor is described in Chapter IV of our guide[1].
Upgrading of the existing reference monitor consists in:

a) increasing the efficiency of the neutron detector assembly
b) flattening the spatial response of the neutron detector assembly
c) providing variability of the energy response of the neutron detector assembly
d) reducing the dead time of the signal processing uni (computerized system)

Substantial progress has been achieved only for the signal processing unit. The circuits have been developed and tested. Commercially available units such as data processor with peripheric units have been purchased.
The new signal processing unit allows for dead time reduction by an order of magnitude.

Active Neutron Assay

Chapter V of our guide[1], dedicated to active neutron assay, was drafted.
In the first section an insight is given (by means of one group, one dimensional diffusion theory) into the problems of neutrons transport from an external source into a multiplying and scattering medium.
This section deals also with other fundamentals of the active neutron assay technique concerning instrumentation.

In section 2 the reference monitor is described.
This monitor is composed of a (Sb-Be) neutron source located in a cylindrical lead pile, which also incorporates the detectors for source and induced fission neutrons.
Discrimination between source and fission neutrons is accomplished by neutron energy biasing. The mathematical model is based on the Nordheim theory of heterogeneous multiplying media[2].

Integral Experiment

The collaborative programme for an Integral Experiment on the Pu waste measurement system of the Dounreay Nuclear Power Development Establishment (DNPDE) was agreed upon.
The objectives of this Integral Experiment are:

a) determination of statistical data (as defined in ref.[1] Chapter II) related to the probable errors in radiometric assay of waste streams;
b) determination of systematic errors in radiometric assay of waste streams.

A schematic illustration of the method is shown in Fig. 1.

The experiment consists in 100% throughput radiometric assay of waste streams and sampling for chemical analysis of selected items from the streams. The chemical analysis results are confronted with radiometric assay results. In the collaborative agreement the work load is divided as follows:

- DNPDE Dounreay provides for the historical data and experimental data (as defined in ref.[1] Chapter II) for both the radiometric assay and the chemical analysis;
- JRC Ispra provides for interpretation of radiometric assay using historical and experimental data from DNPDE, evaluates statistical data and determines systematic errors in radiometric assay.

The experimental programme was initiated recently by:

a) installation of our computerized analyzer for passive neutron assay in DNPDE
b) calibration of DNPDE passive neutron monitors, e.g. response to a bare Pu source and to Pu distributed in matrix materials. The method is outlined in Chapter IV of our guide[1].
c) bench mark measurements in order to check the theoretical predictions, performed by means of computer codes (ANISN and MONK), concerning the neutron spectrum in the thermal region in active neutron assay by the 14 MeV neutron generator of DPNDE.

First results concerning points b) and c) are expected in November 1978.
The collaborative agreement foresees a report in August 1979 on the first results from passive neutron assay and active neutron assay by the 14 MeV active neutron monitor.

Sample Preparation

Experiments are in progress to minimize the measurement uncertainty caused by heterogeneity of waste samples.
First experiments were done with a mock-up simulating a waste drum containing contaminated cellulose in which compact plutonium materials (sources) are embedded. The aim of this experiment is to set up methods for the detection

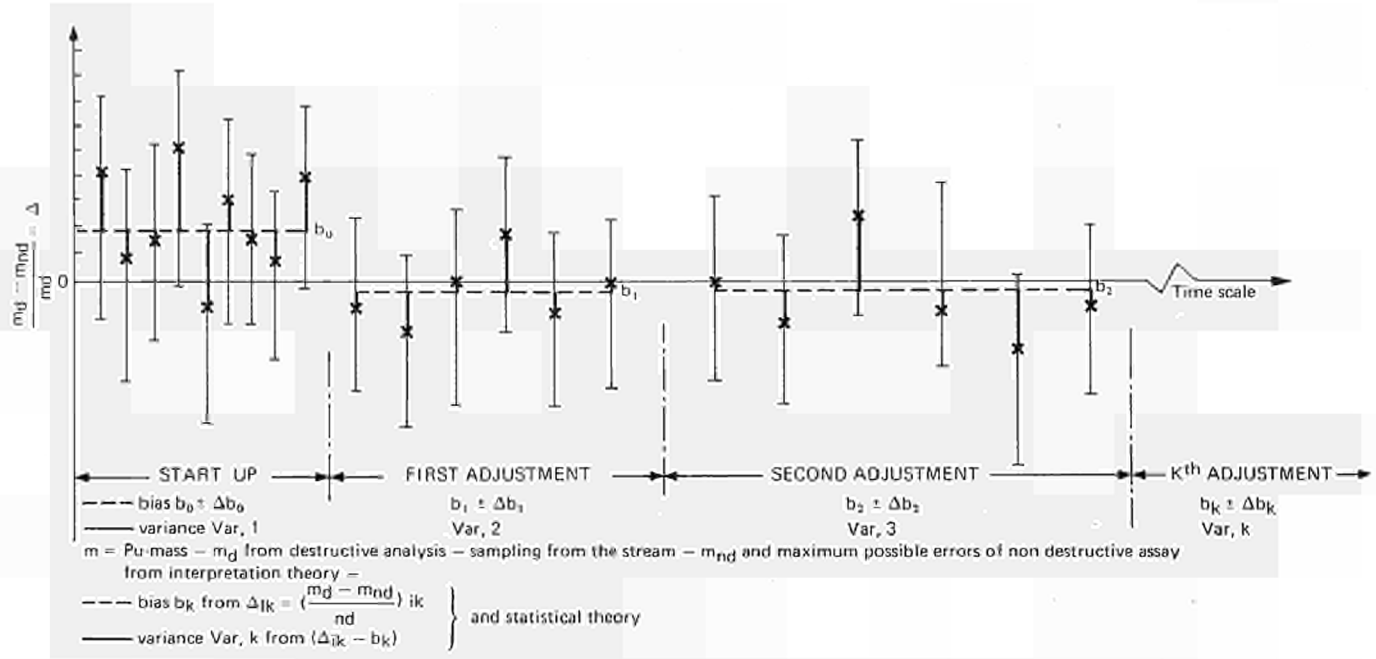


Fig. 1 — Sampling from Waste Stream Assayed 100% by Non Destructive Radiometry

of the presence of compact Pu sources and for the evaluation of their influence on the monitor response.
To this purpose the following procedure is adopted:

- measurement of spontaneous fission neutrons emerging from the waste drum
- centrifugation of the waste drum
- repetition of the spontaneous fission neutron measurements
- comparison of the results obtained before and after centrifugation.

If the results of the measurements before and after centrifugation are similar, we may conclude that compact Pu sources are absent or if present they are located near to the periphery of the drum.
The interpretation based on the homogeneous Pu density distribution yields in the latter case an overestimation of the Pu content.
If the results of the measurement after centrifugation are considerably higher we may conclude that compact Pu sources moved from internal positions towards the periphery of the drum. Knowing the relationship between source position and monitor response (Chapter IV of the guide) and the average migration length of compact sources due to centrifugation we may correct relative to homogeneous plutonium density distribution.
The mock up experiments are designed for the determination of the average migration length of compact PuO_2 sources as function of their geometric size, initial location in the drum, density of the cellulose matrix, and rotation speed of the centrifuge. PuO_2 is simulated by lead having almost the same mass density.
The local position of compact sources is identified from X-rays photographs of the waste drum.
An example of such an experiment is shown in Fig. 2.
The experimental programme was interrupted due to heavy vibrations of the centrifuge at high rotation speed. We have to overcome this problem before continuing the experimental programme.

X-Ray Transmission Pattern

Gamma-ray absorption by heavy matrix material and plutonium lumps is a serious problem in passive gamma assay of wastes. Experimental investigations on the usefulness of X-ray transmission techniques for visualization of gamma absorbers in solid wastes are in progress. To this purpose an X-ray machine ranging up to 400 kV high voltage was set up for X-ray photographs on waste drums. This facility was utilized for the X-ray photographs shown in Fig. 2.

COLLABORATION WITH EXTERNAL ORGANIZATIONS

Contract with EUREX (CNEN, Saluggia, Italy) concerning the testing of on-line alpha-monitor.

Contract with the Institut de Physique Nucléaire (University of Lyon, France) for an expertise on the preparation of the guide.

Collaboration contract with Dounreay Nuclear Power Development Establishment (DNPDE) on an Integral Experiment.

CONCLUSIONS

The objectives for the reporting period have been reached as far as concerns:

- Integral Experiment (Dounreay)
- Passive neutron assay (upgrading of reference monitor)
- Active neutron assay (Chapter V of guide)
- Passive gamma assay (assembling of reference monitor)

The progress of other planned activities was limited.
Testing of a liquid alpha monitor in a fuel reprocessing plant was delayed further. This depends on the conditions of the plant which are unfavourable for our testing programme.
Feasibility studies on samples preparation by centrifuging of waste drums had to be interrupted due to technical difficulties. Systematic studies on X-ray transmission pattern of waste drums were stopped due to the priority given to other points of the programme.

PLANNED ACTIVITIES

For the following semester are planned:

1) bench mark and calibration measurements in the frame of the Integral Experiment
2) design of an upgraded detector assembly for passive neutron assay
3) development of the upgraded signal processor (hardw and software) for passive neutron assay

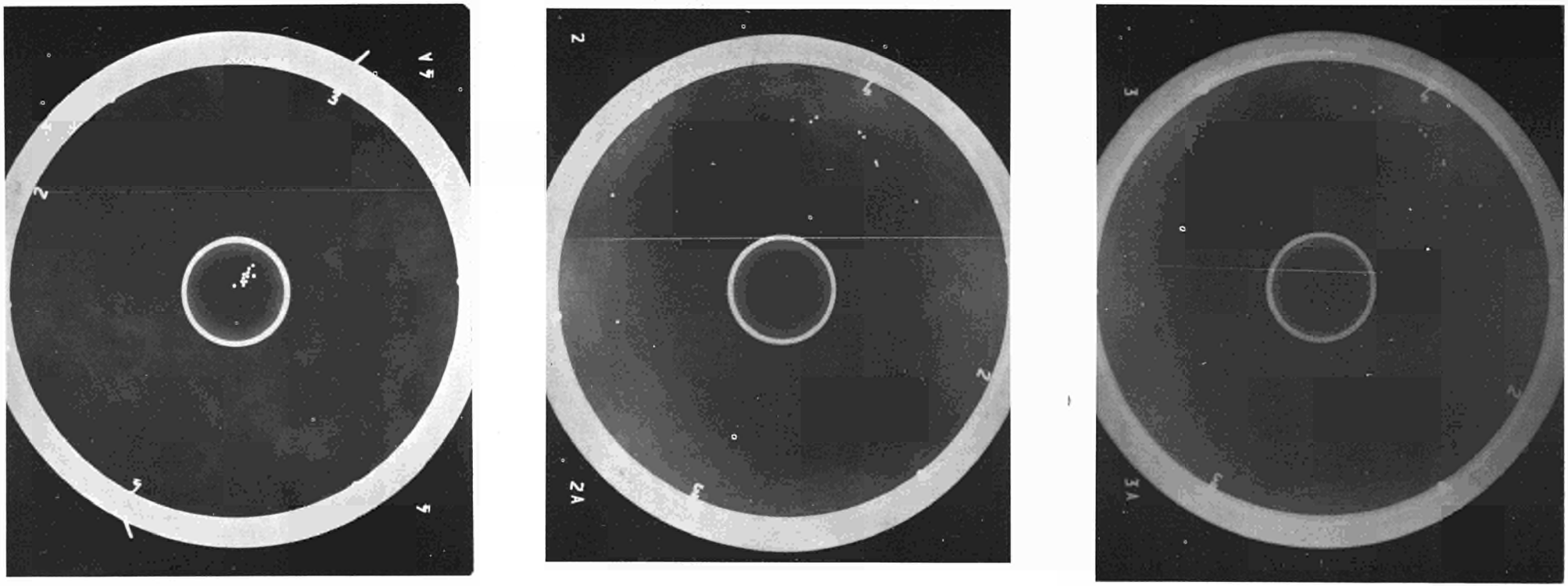


Fig. 2 – Axial X-Ray Radiography of a Waste Drum (Mock-Up) with $\phi = 39.3$ cm, the Drum is Filled with Cellulose of Mean Density $\rho = 0.15$ g/cm^3 From Left to the Right (a, b, c)

a) 10 Pb Spheres with $\phi = 0.15$ cm Are Located Near the Drum Axis
b) The Drum Was Rotated with Maximum 2300 rpm Pb Spheres Migrated to Wards the Periphery, Cellulose Has Been Compacted
c) Same Drum Was Rotated with Maximum 3000 rpm Pb Sphere Migrated Further, Cellulose is More Compacted

4) design of the reference monitor for active neutron assay
5) experimental verification of the interpretational model for the passive gamma reference monitor.

REFERENCES

1 Monitoring of Plutonium Contaminated Solid Waste Streams. A Technical Guide for Design and Analysis of Monitoring Systems.

Chapter I:

G. BIRKHOFF, A. NOTEA: «Planning of Monitoring Systems»
EUR 5635 (1977)

Chapter II:

G. BIRKHOFF, L. BONDAR, A. NOTEA: «Principles and Theory of Radiometric Assay»
EUR 5636 (1977)

Chapter III:

G. BIRKHOFF, A. NOTEA: «Passive Gamma Assay»
EUR 5637 (1977)

Chapter IV:

G. BIRKHOFF, L. BONDAR: «Passive Neutron Assay»
EUR 6027 (1978)

Chapter V:

G. BIRKHOFF: «Active Neutron Assay»
To be published in 1979

2 G. BIRKHOFF, L. BONDAR: «Application of Heterogeneous Method for the Interpretation of Exponential Experiments»
EUR 5658 (1977)

2.2. Chemical Separation and Nuclear Transmutation of Actinides

If the separation of the actinides from fission products is demonstrated to be possible, it will open up a number of alternative waste management options in which the disposal of actinides, largely responsible for the long-term risk, and fission products can be considered separately. One option which would provide an ultimate solution for actinide wastes is the transmutation to short-lived isotopes by neutron bombardment in reactors.

In the framework of the activity of the OECD Nuclear Energy Agency in the field of radioactive waste, the Commission has been chosen as leading organization for the studies on the chemical separation and nuclear transmutation of actinides.

The activity of the JRC in this field includes experiments on the chemical methods required for actinides separation from HAW and assessment studies on the possibility of actinides transmutation in nuclear reactors.

For the chemical separation of actinides from HAW, oxalate precipitation (OXAL Process) and solvent extraction by HDEHP and TBP are being investigated.

The assessment studies include, in addition to the reactor physics aspects, the implications of the nuclear transmutation on the nuclear fuel cycle (actinide fuel element design, modifications in the nuclear plants, increase of cost and risk).
In order to improve the accuracy of the reactor physics calculations a programme of neutron cross section measurements is carried out.

The JRC activities are planned in such a way as to have a maximum of information emerging in the second half of 1979. It is, in fact, intended to prepare by the end of 1979 a major report dealing with a critical evaluation of the feasibility of the chemical separation and nuclear transmutation of actinides.

Chemical Separation of Actinides

OBJECTIVES

The experimental studies on the feasibility of the chemical processes (OXAL, HDEHP and TBP) proposed for the HAW partitioning were continued.
For the reporting period, it was planned to initiate the denitration tests on fully active HAW and to continue HDEHP batch-extraction and OXAL process tests (Actinide/REs partitioning), already initiated on fully active laboratory scale. Assessment studies of the engineering feasibility of the proposed HAW partitioning processes are in progress and a first evaluation is expected by the end of 1978.

RESULTS

HDEHP Extraction Process

Denitration tests on simulated HAW solutions were continued in order to optimize the operating conditions and to clearly identify the parameters of the process.

As already mentioned[1], if suitable process conditions are applied during the HAW denitration it would be possible to minimize the coprecipitation of plutonium with other hydrolysable metal-ions (Mo, Zr, Nb, Fe) and to maintain it in extractable form even at low acidity conditions. The simultaneous co-extraction of plutonium, americium and curium can thus be attained subsequently in a single step.

Denitration tests on fully active HAW solutions were also initiated. For this purpose UO_3 samples, irradiated at a burnup of about 36,000 MWD/t and cooled for 3.5 years, were dissolved and processed. Successive batch extraction stages of this feed solution were utilized to simulate the Purex process and to obtain a 4,000 litres/t HAW raffinate.
A preliminary test on fully active HAW denitration has confirmed the possibility to maintain the plutonium in an extractable form even at low acidity conditions.

An actinide (Pu, Am) residue of a few % was however measured in the precipitate.
Further fully active demonstration tests are under way; the utilization of an improved filtration system is foreseen.

A patent application has been introduced on the use of HAW denitration process for specific purposes (i.e. inextractable Pu solubilization).

TBP Extraction Process

The TBP extraction process is applied to about ten-fold concentrated HAW in order to minimize the amount of nitrates to be added as salting agents.
During the HAW concentration and subsequent interim storage precipitates are normally formed on which plutonium is irreversibly adsorbed.
Laboratory tests carried out during 1977[2] have shown that the fraction of adsorbed plutonium can be minimized provided a nitric acid concentration above 5 M/l is maintained during the concentration process. However, Pu-bearing precipitates could form again during the necessary interim storage of the concentrated HAW solutions.

Therefore it is important for the TBP flowsheet to find process conditions suitable for re-converting to an extractable form the plutonium fraction coprecipitated during the HAW concentration and/or subsequent interim storage, and this is undoubtedly a challenging problem.
For this end an optimization of the final HAW denitration step is presently being studied for making still soluble and extractable the plutonium fraction previously coprecipitated and adsorbed on precipitates during the concentration and thestorage of HAW.
Preliminary denitration tests carried out on a synthetic concentrated HAW solution gave encouraging, although still non conclusive results.
Further experiments will be carried out in order to confirm these results and to better define the operating conditions.

OXAL Process

Experimental tests on fully active laboratory scale, have been continued.
To this purpose a fully active HAW solution, prepared[2] by dissolution and process of UO_2 fuel, irradiated at a burnup of 25,000 MWD/t, was utilized.

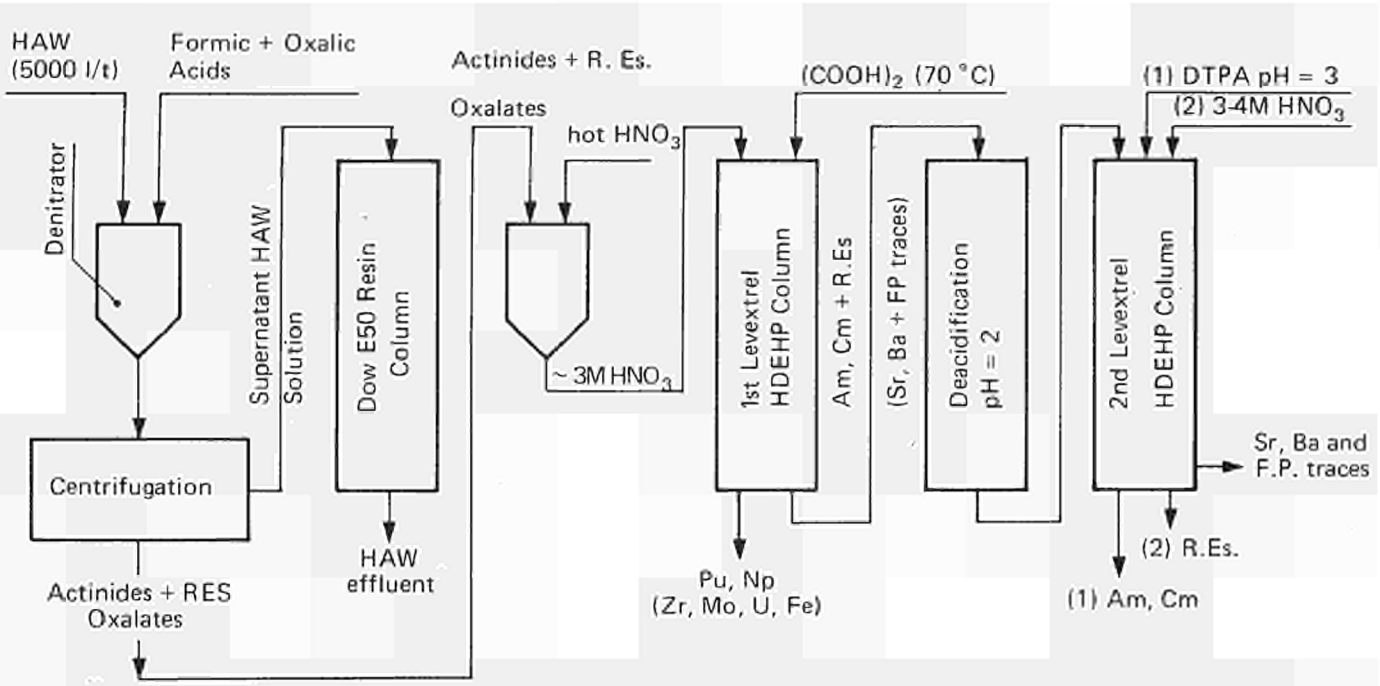


Fig. 1 — Process Scheme for the Actinide + Rare Earths Oxalate Separation and Successive Separation of Actinides from Rare Earths by Column Extraction Chromatography

The operating conditions were the same already applied on simulated HAW solutions[1].

According to the process scheme shown in Fig. 1 the following steps have been carried out in hot cell on the fully active HAW solution:

- Acidity reduction (from ~ 4 M HNO_3 to pH ~ 0.5) and precipitation of the actinide + REs oxalates by simultaneous addition of formic and oxalic acid.
- Centrifugation of the precipitate
- Flowing of the supernatant through a DOWEX 50 resin column to remove the residual actinides
- Dissolution of oxalates by hot concentrated nitric acid
- Flowing of the actinide + REs nitrate solution (3 M HNO_3) through the 1" HDEHP/LEVEXTREL column and subsequent elution of the adsorbed plutonium and zirconium by oxalic acid at 70°C.
- Acidity reduction of the collected column effluent (containing Am, Cm and REs) to pH ~ 2
- Flowing of this solution through the 2"" HDEHP/LEVEXTREL column to adsorb trivalent actinides and rare earths on it and to carry out subsequently their partitioning by selective elution.

Table I — Percent Distribution of Actinides, R.Es and Some F.Ps Measured for Different Streams of the OXAL Process

% ELEMENT DISTRIBUTION (1)				
	Oxalate precipitate	**HAW effluent from DOWEX50 resin column**	**1st HDEHP/ /LEVEX chr. column**	**Oxalic acid ELUATE 2 from 1st column**
Am	>99.9	<0.02	n.f. (3)	n.d. (2)
Cm	>99.9	<0.02	n.f. (3)	n.d. (2)
Pu	>99	n.d. (2)	>99	>99
R.Es	>95	<0.01	n.f. (3)	n.d. (2)
Sb	3.6	71.9	n.f. (3)	n.d. (2)
Ru	1.8	83.6+12.6 (4)	n.f. (3)	n.d. (2)
Cs	0.9	7.6+89.2 (4)	n.f. (3)	n.d. (2)

(1) Referred to the amounts present in the original HAW solution
(2) Not defected (3) Not fixed
(4) HAW effluent + column washing (HNO_3 solution, pH ≃ 0.5)

The results, reported in Table I, show that high actinide separation yields can be obtained by oxalate precipitation.

A further reduction of the actinide content is obtained by means of the DOWEX 50 column.

Table I shows also that high yields can be attained for the plutonium adsorption on the 1" HDEHP/LEVEXTREL column and for the subsequent elution by means of oxalic acid.

The experiments on the actinide/REs partitioning using the 2"" HDEHP/LEVEXTREL column are in progress.

Engineering Feasibility Assessment of Actinide Separation Processes

A study has been started to assess the engineering and feasibility aspects of the separation of the actinides from HAW generated in nuclear fuel reprocessing.

For the chemical separation processes much relevant information is available in the literature, but adaptation to actual HAW arisings is generally lacking; there is a need to propose conceptual flows sheets and to determine whether the separation processes can be modified and adapted to achieve the necessary separation factors and in turn can be integrated into a functional waste processing facility.

Three conceptual flowsheets have been proposed based on TBP, HDEHP and OXAL processes.

The assessment of these flow-sheets is performed assuming that only established and proven technologies will be utilized.

For this purpose a review of the separation equipments used in the nuclear industry (mixer-settlers, pulsed columns, filters and centrifuges) is underway.

Work has proceeded in close co-operation and collaboration with the radiochemistry section enabling detailed discussion on the process chemistry and problems encountered or anticipated for e.g. decontamination factors, precipitation processes, types of precipitates etc. The majority of the bench-scale experiments have been performed on simulated waste make-up solution with only a small amount of experiments being performed up to now, on fully active waste solution. This introduces at present difficulties and uncertainties into the assessment which is greatly dependent on good chemical data for real waste processes.

CONCLUSIONS

The project activities are in progress generally according to the planning. However, some experiments on fully active HAW are proceeding more slowly than scheduled due to the fact that a significant part of the experimentally involved staff (3 chemical technicians) has left the Ispra laboratories. New recruitments are under way. In the mean time a partial replacement has been accomplished by a temporary transfer of personnel from other programmes.

PLANNED ACTIVITIES

During the next six months it is planned:

- to conclude HDEHP batch-extraction tests on fully active laboratory scale.
- to conclude denitration tests under simulated conditions (5,000 l/t and concentrated HAW) and on fully active laboratory scale 5000 l/t HAW).
- to continue the experiments on the separation of the actinides from rare earths at fully active laboratory scale
- to continue the studies for a conceptual design of a HAW partitioning plant.

After the completion of the HDEHP batch-extraction tests, similar tests on the TBP process or, alternatively HDEHP countercurrent extraction tests, under simulated conditions, will be initiated.

REFERENCES

1 Programme Progress Report «Management of Nuclear Materials and Radioactive Waste», JRC-Ispra Establishment, January-June 1978
No. 3528

2 Programme Progress Report on «Management of Nuclear Materials and Radioactive Waste» JRC-Ispra Establishment, July-December 1977
No. 3486.

Assessment Studies on Nuclear Transmutation of Actinides

OBJECTIVES

The aim of this activity is to evaluate the neutron-physical and technological feasibility and cost/risk implications of the trasmutation of actinides other than fuel in fission power reactors. An overall strategy for transmuting those actinides, produced by the European Community power generating system, should be proposed giving indications as to the reactor type to be preferred as transmutation device and the way of introducing the actinides in the reactor. Taking account of the results for risk and cost analysis, a choice between the various possible recycle strategies should be carried out.

RESULTS

Corrected Isotopic Concentration for FBR Pins

In the previous Programme Progress Report[1], isotopic concentrations and geometries for minor actinides-containing fuel pins to be irradiated in the NA 1 FBR were given. These values had to be adjusted due to the following reasons:

- a radial gap of 150 μ is now assumed in order to allow swelling. Consequently, the pellet diameter was reduced from 6 to 5.7 mm;
- since curium is expected to occur mainly in form Cm_2O_3, a theoretical density of 10.66 g/cm³ instead of 11.1 g/cm³ has to be used for the oxide;
- the depleted uranium, proposed as diluent, is a fertile material which increases eventually the specific power generation. Therefore, the initial minor actinides content has to be reduced more than in the case of an inert diluent.

The correct isotopic compositions required to obtain a maximum power rating of 566 W per cm of pin length, at the end of the irradiation period, are reported in Table 1.
The stechiometries assumed for the oxides are NpO_2, $AmO_{1.8}$, $CmO_{1.5}$.

Table I – Initial Isotopic Composition in Grams of Metals and Oxides for FBR Fuel Pins

	ALL ACTINIDES		WITHOUT Np		WITH DEPLETED URANIUM	
	g/cm metals	g/cm oxides	g/cm metals	g/cm oxides	g/cm metals	g/cm oxides
U	–	–	–	–	0.681	0.772
Np	1.235	1.402	–	–	0.834	0.947
Am	0.377	0.422	1.663	1.860	0.254	0.284
Cm	0.0997	0.109	0.441	0.485	0.067	0.074
$\epsilon =$	1.712	1.933	2.104	2.345	1.836	2.077

The dependence of the burn-up on irradiation time and fluence is shown in Fig. 1. The variation of the linear power rating as function of the same parameters is given in Fig. 2.
The relative small increase of the linear power rating of the pin where by-product actinides without Np are recycled represents an argument for recycling only Am/ Cm through the FBR.

Temperature Profiles for FBR Pins

Radial temperature profiles for different geometric configurations of FBR fuel pins are being calculated at the Karlsruhe Establishment of the JRC (Transuranium Institute) by means of computer codes. The results will be published in the next Programme Progress Report. The scope of this work is to guarantee that the maximum temperature in the pin does not exceed the melting point of the fuel material.

Collection of the Performance Data of LWRs

Neutron physical calculations have indicated the LWRs as possible devices for the actinides transmutation. An additional enrichment in fissile material is required when self-generated actinides are blended with normal fuel in order to compensate the low fission-to-capture ratio for by-product actinides to be transmuted in a thermal reactor. An even higher enrichment is necessary when actinides are introduced in LWRs in form of target pins. As a high enrichment can influence the thermal properties of the fuel element, the performance data for LWRs, collected for large power plants (~ 1000-1300 MWe) are reproduced in Table II.

The data will be employed in the design work for by-product actinide-bearing LWR fuel pins in line with the results concerning the FBR.

Table II – Characteristics of LWRs

Country	United States of America								FRG	Japan	France
Name	E. Fermi 2 (2)	G.E.		Peach Bottom 3(1) 2-3(2)		Brown Ferry 3(1) 1-2-3(2)		Westinghouse	Indian Point 3(1)	Biblis B (3)	Ohi(1)	Tricastin 4 (1)
Type	BWR	BWR6	BWR	BWR		BWR		PWR	PWR	PWR	PWR	PWR
Reactor power (MW) thermal	3293	3833		3293	3293	3293	3293		3025	3733	3423	2775
Reactor power (MW) electric	1152	1265		1098	1098	1098	1098	1000	1000	1300	1175	957
Temperature °C core inlet	278	280		277	275	275	275	293.4	283.7	290	989	284
Temperature °C core outlet	285	288 IN OUT			286	286	286	331.7	315	323	325	325
Temperature °C clad max		362 a) 297 425 b) 314 445 c) 387			358		313	350	347	347	350	335
Temperature °C fuel max	1750	1871		2457	2460	1810	2415	1200 (av.)	2260	2.226	2400	2500
Dimensions core height mm	3800	4820			3660	3700	3660	4267	3650	3900	3660	3660
core ϕ eq. mm	4750	3700			4750	4800	4750	3040	3370	4170	3370	3040
clad o.d. mm	12.3	12.26		14.3	14.3	12.5	14.3	9.50	10.7	10.75	9.5	9.5
clad i.d. mm	10.60	10.64		12.67	12.7	10.78	12.7	8.36	9.46	9.35	8.36	8.36
radial gap (μ)	140	116		162	150	90	150	170	80	1607	80	80
clad thickness (μ)	810	810		813	800	860	800	570	620	700	570	570
pellet ϕ mm	10.40	10.41		12.35	12.4	10.6	12.6	8.19 (8.02)	9.3	9.057	8.2	8.2
pellet density %	93	95						95				
Clad material	Zy-2	Zy-2		Zy-2	Zy-2	Zr-2	Zr-2	Zy-4	Zy-4	Zy-4	Zy-4	Zy-4
Fuel material	UO_2	UO_2		UO_2	UO_2	UO_2	UO_2	UO_2	UO_2	UO_2	UO_2	UO_2
Enrichment	2.5	2 2.19 2.44		2.19	2.19	2.2	2.19	3.22	2.78	2.6	2.61 3.21	2.1 3.1 3.25
Subassembly	764	848		764	764	764	764	157	193	193	193	157
Pins/Subass.	64	62		49	64	63	64	264	204	236	264	264
Linear av. (W/cm)					231		231	165	203	204	178	178
Linear max. (W/cm)	410	440			601	440	601	412				
Thermal neutron flux core max	4.1				7.8		7.8		12			
$\times 10^{13}$ (n/cm^2 sec) core av.					3		3		5			
Fast neutron flux core max												
$\times 10^{14}$ (n/cm^2 sec) core av.	0.64				1		1		2.8			
$\bar{M}$ (MW/THM)	23.2	24.7			22.2		22.1		27.3	36.7	39	
Burn up of core fuel av.		28			19	19	19	33		20	24	
GWD(th)/T max	58	40		25	30		50	50	33.4	48	33	33

1) Nuclear Engineering Int.: April. Supplement 1977 p.30
2) Directory of Nuclear Reactors: Vol X IAEA (1976)
3) Atomwirtschaft: Nov. 1972 p.552

a) 100% full power
b) 110% over power
c) after running period

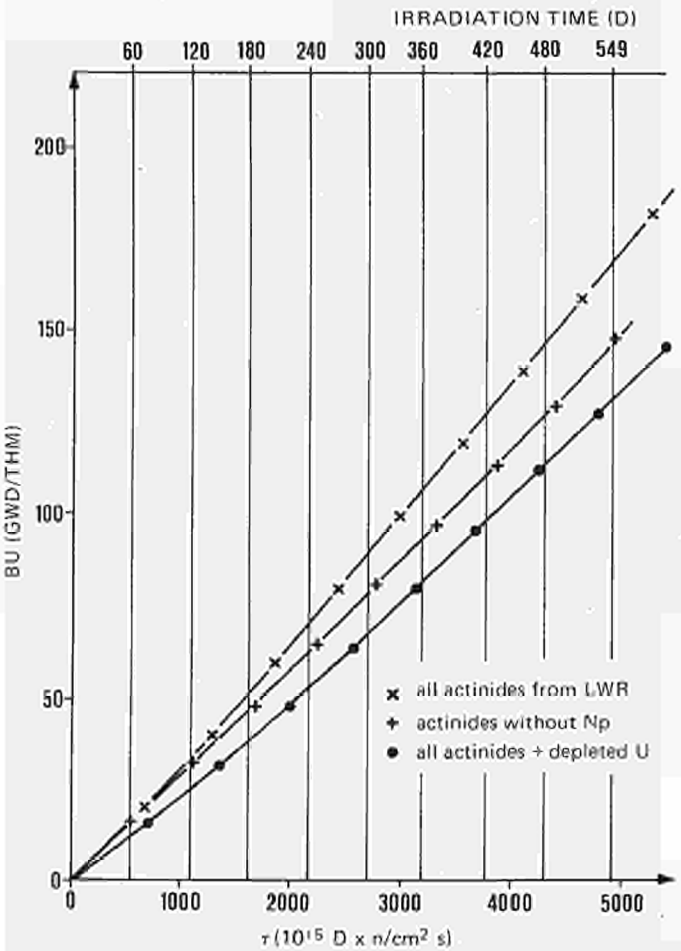


Fig. 1 – Burn-Up of Different Fins Inserted in the NA1 (ϕ = 18.92 · 10^{15} n/cm^2 -s) as Function of Fluence and Irradiation Time

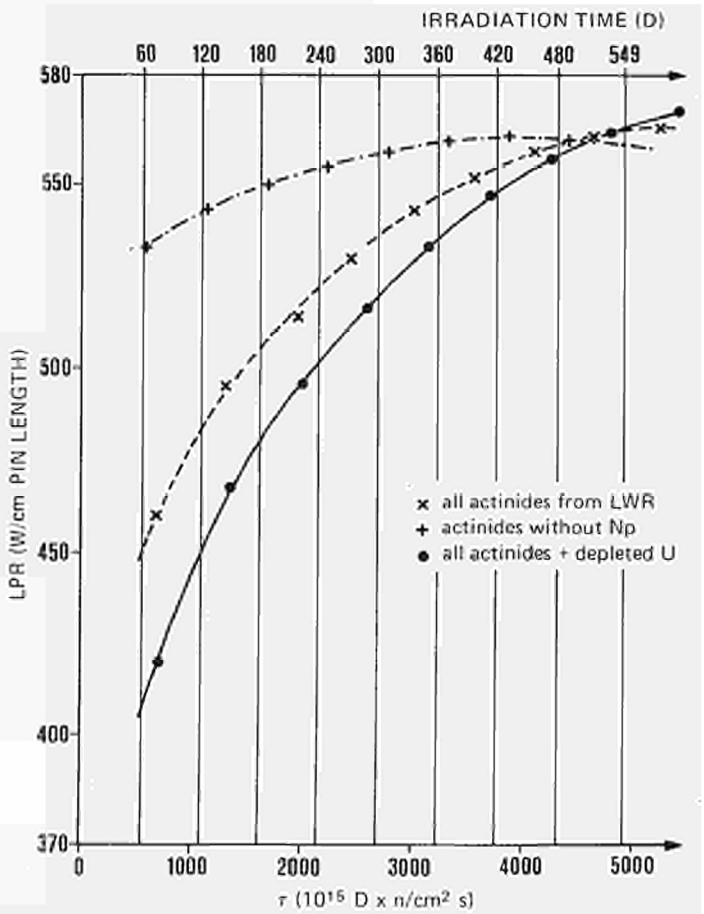


Fig. 2 – Linear Power Rating of Different Pins Inserted in the NA1 (ϕ = 8.92 · 10^{15} n/cm^2-s) as Function of Fluence and Irradiation Time

Reactor Physics Calculation for LWRs

In order to improve the calculation techniques for the build-up and transmutation of by-product actinides in LWRs, available at the JRC Ispra, test calculations were performed with the multi-group, space-dependent burn-up computer programs LASER and EREBUS[2]. The calculational results for fuel actinides are in general in a good agreement with experimental data for irradiated fuel from Yankee Rowe, Trino Vercellese, and Obrigheim reactors. An isotopic chain for by-product actinides is being implemented in EREBUS. Due to inherent limitations for the maximum number of isotopes and the sort of decay schemes some approximations have to be applied when constructing the by-product actinide isotopic chain of EREBUS.

Generation and Improvement of FBR Cross Section Libraries

The results for the linear power rating of minor actinide-containing fuel pins as function of the irradiation time depend on the capture and fission cross sections. The results given in one of the preceding subsections have been obtained using the one-group cross-sections of the ORIGEN code. A comparison between these data and the ones of other sources is reproduced in Table III. It shows the discrepancies between the libraries included in the burn-up codes ORIGEN and FISPIN (σ_f of Pu-238, Cm-242, Cm-243; σ_c of Pu-238; Np-237, Am-241, Am-242(1), Am-243, Cm-243).

Moreover, the influence of the reactor spectrum on the effective one-group cross sections may be realized by confronting once the results of JAERI for a soft blanket spectrum with those of a medium core spectrum and the other time the results of a UK set for a medium core spectrum of an oxide-fuelled FBR with those of the hard spectrum of a metal-fuelled assembly. Strong variations may be observed in σ_f of Np-237, Am-243 and Cm-244 as well as in σ_c of Np-237, Am-241.

It may be concluded that work has still to be performed in order to verify and to improve the data library of ORIGEN studying the effects of different reactor spectra on one- or few-group cross sections as well as of different differential or multi-group cross sections. This work aims at the generation of a new FBR library for minor actinides.

Differential and multi-group cross sections for by-product actinides have been collected from different sources. Neutron spectra of various FBRs have also been derived. By means of a computer program, the differential cross sections will be collapsed to one-group cross sections in order to estimate the effect of the various error sources and to generate a consistent data set which will be introduced into ORIGEN.

Shielding Thickness of Fabrication Facilities of Fuels Containing By-Product Actinides

The minor actinide elements (Np, Am, Cm, Cf) of concern in the transmutation concept, are characterized by a high neutron emission due to spontaneous fission and (α-n) reactions. In handling such hazardous materials the composition and wall thickness of the fabrication facilities should be dimensioned in order to limit the radiation exposure of personnel to a level of 2.5 m Rem/h.

On the other hand, at the moment when actinides are eventually recycled for transmutation, also the plants for the fabrication of fuels containing recycled plutonium will eventually require additional shielding and special fabrication technologies such as remote and automated operations.

In order to make possible an evaluation of the additional costs in fabrication deriving from the actinides recycling, calculations of the shielding thicknesses, necessary to reduce the neutron dose to 2.5 m Rem/h, have been performed for the following fuels[8]:

Table III – Comparison of One-Group Cross Section for FBRs Taken from Different References

		JAERI (3)		ORIGEN	UK (5)				(6)			(7)
		Blanket	Core	(4)	CFBR	DFR	Fispin	Hink	SAVR	TND	ENDF/B4	Beamon
Pu-238	σ_f	1.128	1.134	1.38			0.44			1.12		
	σ_c	1.111	0.4971	0.224			1.15			0.68		
Np-237	σ_f	0.1726	0.3268	0.36	0.34	0.67	0.338	0.31		0.32	0.32	
	σ_c	3.287	1.719	0.765	1.87	0.90	1.87	1.8			1.76	
Am-241	σ_f	0.4327	0.4283	0.463	0.45	0.57	0.404	0.39		0.45	0.4	
	σ_c	3.142	1.403	0.99	1.49	0.68	1.91	1.18		1.88	1.09	
Am-242 (1)	σ_f	5.230	3.743	1.83	3.33	2.53	3.33			3.73		
	σ_c	1.026	0.6455	0.403	0.10	0.07	0.10			0.38		
Am-243	σ_f	9.356-2	0.1970	0.237	0.19	0.39	0.188		0.19	0.22	0.18	
	σ_c	2.042	0.9074	0.555	1.66	0.70	1.70		0.42	0.9	0.81	
Cm-242	σ_f	1.534	1.929	0.42	1.26	1.53	1.26	1.94	0.58		0.52	
	σ_c	1.120	0.6794	0.380	0.50	0.20	0.50	0.25	0.55		0.5	
Cm-243	σ_f	3.748	2.655	0.32	3.14	2.23	3.14			0.39		
	σ_c	0.7264	0.4447	0.40	0.10	0.05	0.10					
Cm-244	σ_f	0.3731	0.5272	0.412	0.45	0.86	0.548					
	σ_c	1.194	0.5319	0.373	0.57	0.21	0.48					
Cm-245	σ_f			2.45	2.92	2.07	2.5					3.24
	σ_c			0.40			0.5					0.191
Cm-246	σ_f			0.3			0.4					0.246
	σ_c			0.302			0.5					0.297

- U/Pu fuels for LWRs (5% Pu) and FBRs (30% Pu)
- Actinides bearing fuels for homogeneous recycling in LWRs and FBRs and heterogeneous recycling in FBRs.

The results of the calculations, performed by means of the computer code ANISN, are summarized in Table IV.

Risk Assessment

The waste management code for the recycling of by-product actinides has been tested with preliminary actinide input and output matrices obtained from repeated operations of the ORIGEN code, for an 1 GWe LMFBR.

The core blanket average fuel composition of the reactor for the first cycle was:

U235 7.15×10^3 g/t_{HM}
U238 3.49×10^5 g/t_{HM}
Pu239 1.14×10^5 g/t_{HM}
Pu240 5.46×10^4 g/t_{HM}
Pu241 9.04×10^3 g/t_{HM}
Pu242 $3.67 \cdot 10^3$ g/t_{HM}

t_{HM} = tons of heavy core metal)

For all later cycles the U and Pu isotopes formed in the by-product actinides are added to the fuel.
At equilibrium actinide recycle conditions, the fuel composition is mainly extended by the following isotopes:

U234 5.37×10^1 g/t_{HM}
U238 3.08×10^2 g/t_{HM}
Pu238 1.22×10^3 g/t_{HM}

Other fuel isotopes have smaller contributions.

In a first numerical assessment[9] the hazard accumulation of the actinide waste was calculated for different loss fractions of actinides to the waste and different nuclear power generation periods (50, 400 and 800 years). Two fuel cycles were compared:

- a reference fuel cycle in which all by-product actinides are discharged to the waste
- a by-product actinide fuel cycle in which the self generated by-product actinides are recycled and only a small fraction of them is lost to the waste.

A good picture of the maximum potential of the by-product actinides recycling waste management alternative is given by the **actinide waste hazard ratio** r_I (t). In this ratio the hazards generated by the actinides in the waste with by-product actinide recycling are divided by the hazards of the actinide waste in case of the reference fuel cycle

r_I (t) = (waste hazard of actinides in the by-product actinide recycling case) : (waste hazard of actinides in the reference fuel cycle)

Figs. 3 and 4 give this ratio for a duration of 50 years nuclear power for inhalation of air and ingestion of water respectively. The actinide loss fractions of reference fuel cycle and by-product actinide cycle were identical in each ratio.

This ratio as function of time is quasi independent of the duration of the nuclear power generation but depends strongly on the loss fractions for fuel and by-product actinides.

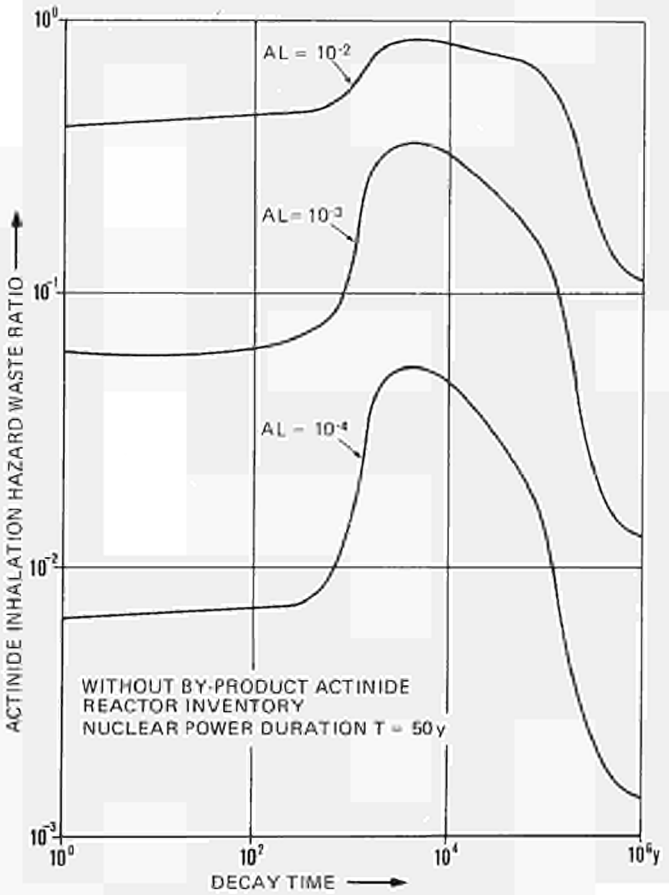


Fig. 3 — Actinide Inhalation Hazard Waste Ratio

Table IV — Shielding Thickness to Reduce the Total Neutron Dose Rate to 2.5 in Rem/h

			Neutron g^{-1} sec^{-1}	Neutron sec^{-1}	Shielding thickness (cm) H_2O	Polyethylene	Concrete
Plutonium fuel fabrication Amount per day and per line	200 kg for LWR containing 5% Pu	from a LWR discharge	1.12 + 3	1.12 + 7	15	13	36
		from a LWR equilibrium cycle	1.62 + 3	1.62 + 7	17	14	40
	50 kg fro FBR containing 30% Pu	from a LWR discharge	1.12 + 3	1.68 + 7	17	14	40
		from a FBR equilibrium cycle	5.17 + 2	7.75 + 6	13	11	33
		from a LWR equilibrium cycle	1.62 + 3	2.43 + 7	19	15	44
Actinides homogeneous recycling Amount per day and per line	200 kg for LWR containing 0.132% actinides (264 g)	from a LWR equilibrium cycle	1.7 + 9	4.49 + 11	74	61	136
	50 kg for FBR containing 0.65% actinides (328.5 g)	from a LWR discharge	7.84 + 5	2.8 + 8	31	25	67
		from a FBR discharge	6.45 + 5	2.3 + 8	30	24	65
		from a FBR equilibrium cycle	3.49 + 6	1.2 + 9	38	32	81
	50 kg for FBR containing 6.61% actinides (3.305 g)	from a LWR discharge	7.84 + 5	2.5 + 9	43	35	87
		from a FBR discharge	6.46 + 5	2.1 + 9	42	34	85
		from a FBR equilibrium cycle	3.49 + 6	1.1 + 10	51	42	101
Actinides heterogen - recycling Amount per day and per line	328,5 g actinides (Np-Am-Cm) corresponding to one sub-assembly per year (5.41 + 4 g) for FBR - Material from	LWR discharge	7.84 + 5	2.57 + 8	30	25	65
		FBR discharge	6.46 + 5	2.12 + 8	29	24	64
		FBR equilibrium cycle	3.49 + 6	1.15 + 9	38	31	78
	3.305 g actinides corresponding to sub-assembly per year (5.41 + 5 g) for FBR - Material from a	LWR discharge	7.84 + 5	2.59 + 9	43	35	87
		FBR discharge	6.46 + 5	2.13 + 9	42	34	85
		FBR equilibrium cycle	3.49 + 6	1.15 + 10	51	42	101

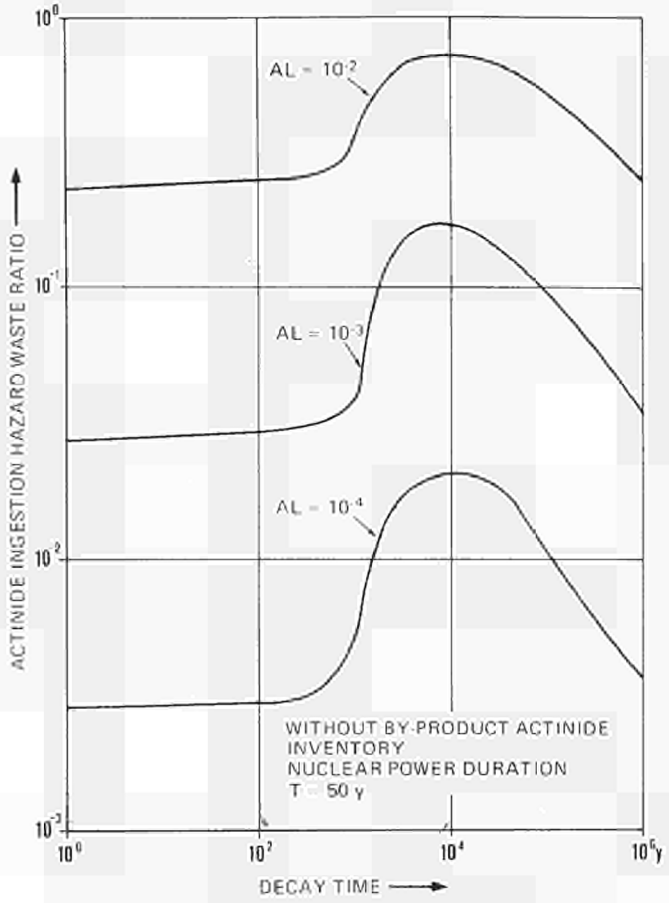


Fig. 4 – Actinide Ingestion Hazard Waste Ratio

It appears that only in the case that the actinide losses to the waste are as small as AL = 10^{-4} the actinide recycling concept leads to a ratio which is dominantly below 1%.
In case the loss factor would be in the order of AL = 10^{-2} there would not be an advantage to recycle actinides from the hazard point of view.
The pronounced increase of the ratio between 10^3 and 10^5 years is provocated by the sharp decrease of the by-product actinide waste in this interval of the decay period.
In case the by-product actinides would be recycled in special target elements with a two times larger residence time of special target elements in the reactor the ratio would be reduced by a factor of 2 approximately.

The long term hazard is at times greater than $5x10^5$ years dominated by the decay of the Np-237 and its daughter products. The recycling of by-product actinides leads to a strong reduction of the Np-237 content. For this reason the actinide waste hazard ratio reaches very low values at a decay time larger than $5x10^5$ years.

In case that after the end of the nuclear power generation period only the by-product actinides of the last reactor charge are accounted to the waste and all the fuel actinides are still considered as fuel another ratio is obtained which gives a more pessimistic view on the actinide recycling concept. This hazard ratio is defined:

r(t) = (waste hazard of actinides in the by-product actinide recycling case) + (hazard of last by-product actinide reactor inventory) : (waste hazard of actinides in the reference fuel cycle)

It decreases with the duration of the nuclear power generation period because the relative contribution of the last reactor by-product actinide inventory is proportional to the inverse of this duration. For an infinite nuclear power generation period this ratio becomes identical to the ratio not considering the last by-product actinide inventory of the reactor.
The ratio obtained for the inhalation and ingestion hazards (Figs. 5 and 6) are below 10% throughout the considered decay period only for a duration of nuclear energy greater than 400 years and loss factors to the waste as low as AL = 10^{-4}. A nuclear power generation period of only 50 years would be too small even for a loss factor of 10^{-4} to pass the 10% limit in 10^6 years.

Figs. 3 to 6 are based on a comparison of the two fuel cycles in which the global loss fractions of fuel and by-product actinides were the same for the two fuel cycles. In case the reference fuel cycle has higher loss fractions to the waste due to its simpler fuel reprocessing and fabrication plants as compared to anticipated plants for the by-product actinide recycling scheme then the ratios will naturally decrease[9].

The results of Figs. 3 and 6 are only preliminary data. More detailed studies are going on for the elaboration of another fast reactor cross section set to be used in ORIGEN for the calculation of better fuel and by-product actinide input and output matrices.
However, the incertainty of the cross sections and of the theories used to calculate the isotope evolution might affect these ratios considerably. Another source of uncertainty derives from the loss fractions of actinides to the waste (AL).
During the numerical tests of the waste management code it appeared that the number of isotopes considered has to be increased from 37 to about 100 for decay periods longer than 10^5 years.
Such a modification of the program is at present in progress. It will have a subroutine which constructs its own decay schemes after having defined for each isotope the decay modes and their respective decay fractions.

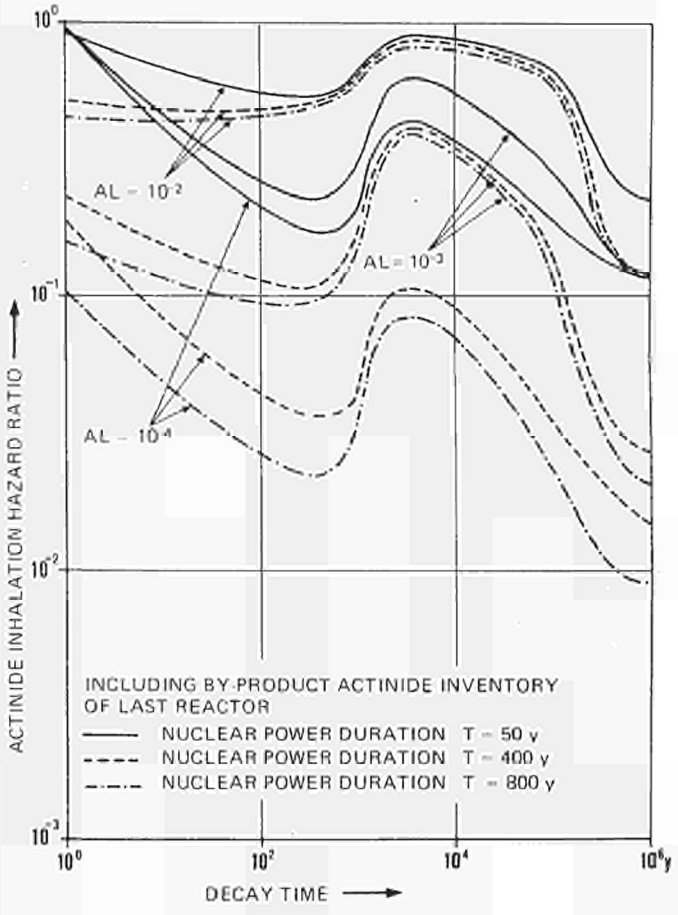


Fig. 5 – Actinide Inhalation Hazard Waste Ratio

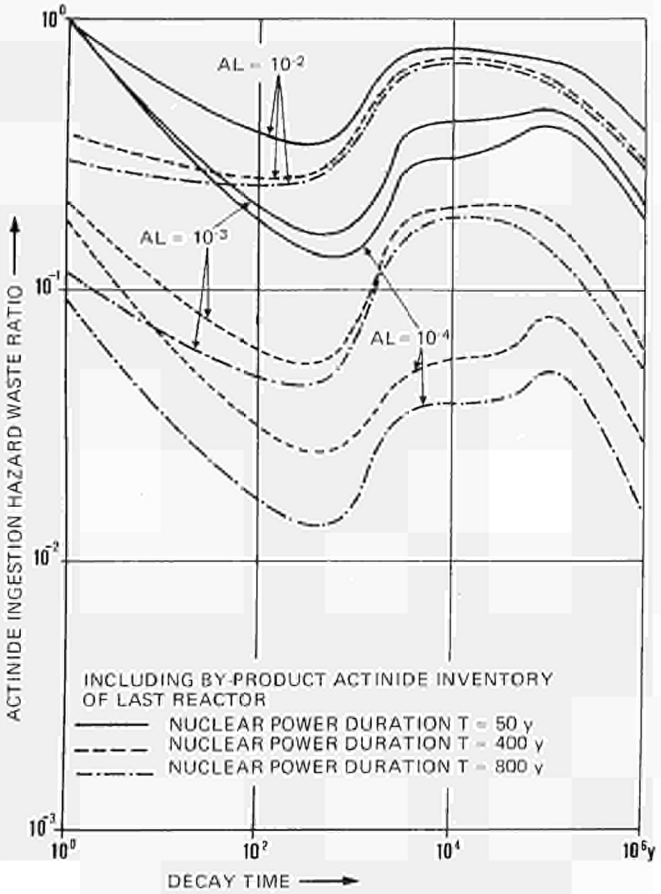


Fig. 6 — Actinide Ingestion Hazard Waste Ratio

COLLABORATION WITH EXTERNAL ORGANIZATIONS

The contracts with KFA Jülich and CNEN Casaccia regarding the reactor physics calculations for thorium-fuelled reactors and fast breeder reactors proceed normally. At present the first progress reports are being prepared.

CONCLUSIONS

The work concerning the updating of LWR and FBR libraries has been slightly delayed due to difficulties with the application of the new operation system installed in the computer centre at Ispra.
Caused by a reduction of manpower, it was decided to give preference to the design work concerning the by-product actinide pins and the elaboration of arguments for a suitable recycle strategy. The work on cost evaluation was therefore interrupted.

PLANNED ACTIVITIES

In the next semester the following results are expected:

- first results on the assessment of new fabrication routes;
- radial temperature profiles for FBR target pins containing actinides;
- influence of actinides on fuel/cladding chemical interactions;
- fuel element design for LWRs which contain by-product actinides;
- comparison of experimentally determined isotopic concentrations in irradiated fuel pins with calculational results for FBRs and LWRs and improvement of nuclear libraries for by-product actinides
- extension and refinement of the risk evaluation calculations,
- definitive choice of computer programs for the LWR physics calculations.

REFERENCES

1 Programme Progress Report «Management of Nuclear Materials and Radioactive Waste» JRC-Ispra Establishment, January-June 1978, No. 3528

2 S. GUARDINI: «Preliminary Burnup Calculations for Actinide Transmutation in LWRs»; Working Paper No.17 (Sept.1978)

3 K. TASAKA et al.: «Build-up and Decay of Fuel Actinides in the Fuel Cycle of Nuclear Reactors»; JAERI-M 6541

4 M.J. BELL: «ORIGEN - The ORNL Isotope Generation and Depletion Code»; ORNL-4628 (May 1973)

5 M.G. SOWERBY, Harwell: Private Communication

6 H. KUESTER, M. LALOVIC: «Influence of Various Nuclear Data Sets on the Prediction of Actinides in Fast Reactor Fuel after Discharge» NEA CRP meeting Petten (June 1977)

7 S.L. BEAMON, E.A. AITKEN: «Feasibility Studies of Actinide Recycle in LMFBRs as a Waste Management Alternative»; NR-CONF-001, Seattle (1976)

8 E. ZAMORANI, J. CAMETTI, C. PONTI: «Actinides Transmutation in FBRs: Impact on Shielding Thickness of Fabrication Facilities and on the Recycling in the Year 2000», Working paper No.16 (July 1978)

9 L. ANSELMI, K. CARUSO, W. HAGE, E. SCHMIDT: «On the Hazard Accumulation of Actinides Waste in a Pu-Fuelled LMFBR Power Economy with and without By-Product Actinide Recycling» (to be published)

Actinide Cross Section Measurements

OBJECTIVES

The Am-241 fission cross section has been measured during 1976 and 1977 applying two methods : the neutron and the fission product detection methods. The cross section data of the neutron detection method were analysed during 1977 for all the measurements executed with monoenergetic neutron bursts generated in thin targets using the $^{7}Li(p,n)$ reaction. All experiments with white neutron spectrum bursts using thick Li targets were analysed on the basis of the existing target specifications during late 1978. The specification work of the targets used in both techniques is being continued in other laboratories. Another activity concerning the (n,γ) and (n,f) cross section measurement of Am-243 has been started.

RESULTS

Fig. 1 gives the Am-241 fission cross section as function of energy, obtained with the neutron detection method.
The data are based on the Am-241 sample specification available on June 1978. In the energy range of 10 keV $\geqslant$ E $\geqslant$ 120 keV all cross sections were measured using continuous neutron spectrum bursts of 0.8 ns duration and a flight path of about 6 cm. Kinetically collimated neutrons were generated in thick Li(p,n) targets adjusting the proton energy just above the reaction threshold. The neutron time of flight spectra obtained from the fissile samples were analyzed in 4 different ways in order to evaluate the influence of the systematic errors due to different interpretation procedures.

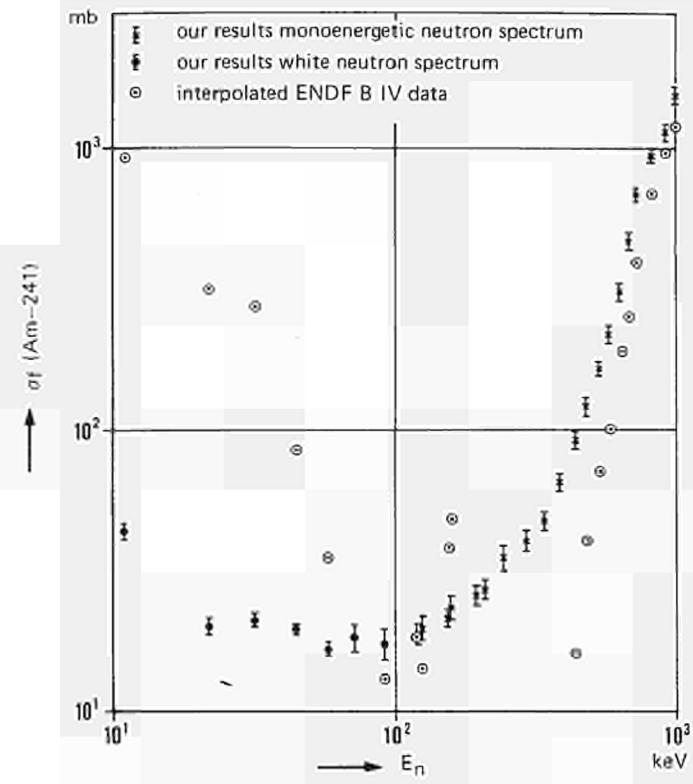


Fig. 1 – Fission Cross Section of Am-241

It was found that these errors are negligible compared with the statistical error except at the lowest neutron energy at 11.3 keV. At this energy the unfolding procedure of the time of flight spectra, in order to correct the shift and spread of the prompt fission neutron time distribution along the path between sample and neutron detector, has a marked influence on the cross section result.

In case the time of flight spectrum would not have been unfolded the cross section would be 32.8 mb instead of 44.2 mb obtained with the standard unfolding procedure without any curve smoothing prior or after unfolding. Our results agree well especially with those of SHPAK[1] below 120 keV, within the error limits. However, our data are not yet finalized.

More detailed investigations are carried out at present concerning the exact composition of the Am-241 target.

The data above 120 keV were already reported in previous progress reports and in Ref.[2].

So far only the following errors are investigated for the cross section measurements:

- Statistical errors of all TOF spectrum manipulations
- Systematic errors of the TOF interpretation procedures
- Flight path errors
- Error of the detected prompt fission neutron spectrum
- Error of the second neutron group appearing in the Li(p,n)-reaction above 2.373 MeV proton energy[3].

Limited work was carried out on the fission neutron detection method. The isotopic composition of the U-235 sample has been remeasured at the Geel Establishment of the JRC (CBNM) and the previous results were confirmed with smaller error limits. An independent measurement of the isotopic composition using α-spectrometry with thin foils yielded the same results as those of the CBNM.

The uncertainty existing in the isotopic composition of the Am-241 fission targets has not yet been resolved by the CBNM.

The preparation of the new measurements on Am-243 concerning (n,γ) and (n,f) cross-sections is in progress. Main parts of the instrumentation received at the beginning of October, are now in operation.

Two experimental methods are in development, one for the scintillation counting of fission neutrons in an intense gamma ray field and another for the counting of fission fragments in a spark gaseous chamber, insensitive to alpha activities.

COLLABORATION WITH EXTERNAL ORGANIZATIONS

The Am-241 measurements were performed in collaboration with the staff of the Institute for Applied Nuclear Physics at the KfK Karlsruhe.

In collaboration with the staff of this Institute a paper was presented at the International Conference on Neutron Physics and Nuclear Data for Reactor and other Applied Purposes, Harwell, September 25-29, 1978[4].

The Am-243 fission cross section measurements are prepared in close collaboration with the above mentioned Institute.

CONCLUSIONS

The activity on the Am-241 fission cross section is interrupted until new data are available on the isotopic compositions of the different targets. The staff employed in this activity will reinforce the assessment studies on nuclear transmutation of actinides and the actinide monitoring.

PLANNED ACTIVITIES

There are no major activities planned at present for the analysis of the Am-241 fission cross section. The preparation of the two measurement techniques for the Am-243 fission cross section and n-γ cross section will be continued.

REFERENCES

1 D.L. SHPAK, Yu.B. OSTAPENKO, G.N. SMIRENKIN: «Subbarrier Neutron Fission of Am-241» Zhet. Pis. Rad. 10 n. 6 (1969) pp 276-279

2 W. HAGE, H. HETTINGER, F. KAEPPELER, S. KUMPF, K. WISSHAK: «Measurement of the Fission Cross Section of Am-241» Proceedings First Technical Meeting on the Nuclear Transmutation of Actinides - JRC-Ispra, March 1977, EUR-5897e,f (1977)

3 H. LISKIEN, A. PAULSEN: «Neutron Production Cross Sections and Energies for the Reactions ^{7}Li (p,n) ^{7}Be and ^{7}Li (p,n) ^{7}Be*» - Atomic Data and Nuclear Data Tables 15 n.1 (1975)

4 K. WISSHAK, F. KAEPPELER, W. HAGE: «Measurement of the Subthreshold Fission Cross Section of Pu-240 and Am-241» - International Conference on Neutron Physics and Nuclear Data for Reactor and other Applied Purposes, Harwell, 25-29, September 1978.

2.3 Decontamination of Reactor Components

OBJECTIVES

The evaluation of the report prepared by the firm Laborelec under contract with the JRC[1], and our own bibliographic research (report in publication) permit the following conclusions:

1 Radiation fields in nuclear power plants are an increasing problem.
2 The most suitable and most applied method for primary circuit parts made from austenitic alloys (austenitic stainless steel, incoloy or inconel, making up approx. 70% of the internal surface of nuclear reactors) is the so-called hard chemical decontamination (APAC and similar processes).
3 For some particular reactors (CANDU) a sufficient radiation field decrease was obtained with soft decontamination (i.e. redox cycling process or Candecon process).
4 Gaps of knowledge exist regarding the mechanisms of contaminated oxide film formation and the mechanisms of decontamination of austenitic alloys.

On the basis of these conclusions and of the recommendations of the Advisory Committee for Programme Management to orient the activity towards the waste problems related to decontamination processes and decommissioning operations, the following activities have been planned:

- Systematic study of chemical decontamination,
- Study of the physico-chemical structure of oxide layers,
- Study of the mechanisms of decontamination processes,
- Evaluation of the decommissioning of the Ispra-I reactor.

RESULTS

Systematic Study of Chemical Decontamination

Chemical decontamination is a well-known technique which has been largely applied mainly on pieces of moderate dimensions.

Scope of the present work is to verify which is the minimum concentration of salt giving an acceptable decontamination level, in order to minimize the amount of final sludges generated by the chemical decontamination.

The work will be performed using known chemical systems. In particular, firstly, a three stage system is being analysed, namely:

- pre-treatment stage : Citrox (Turco Decon 4521)
- oxidizing stage : AP (Turco Decon 4502)
- final stage : Citrox (Turco Decon 4521)

If other proved systems become available, they will be tested as well.

As already mentioned in the previous Programme Progress Report[2] three types of samples have been examined:

- Samples obtained oxidizing surfaces of AISI 304 L at 600°C in overheated steam. These samples can be considered representative of a surface with a long time of exposure in the reactor.
- Samples obtained by decomposition of a solution of ferrous tartrate on a surface of AISI 304 L. These samples can be considered representative of a surface with a short time of exposure in the reactor.
- Contaminated pieces from power stations, used essentially to verify the results obtained on non-active samples.

The examination at the scanning electron microscope (SEM) of the samples of the first type, at different stages of attack, have shown that the first treatment with Citrox dissolves mainly the external layer of the oxide. In the case of contaminated samples the first treatment removes about one half of the activity.

The second treatment (oxidation) is effective mainly in opening the oxide inclusions which enter into the base metal. Its influence on the removal of activity from the contaminated samples is practically negligible. The aim of this treatment is to open the oxide inclusions and perhaps to change their chemical characteristics in order to make possible an effective attack during the second treatment with Citrox.

The results of the experiments indicate that for the samples of the second type, only superficially oxidized, the first treatment is sufficient. Thus the tests on these samples have been discontinued.

The work is now concentrated on the effect of the second treatment with AP on heavily oxidized samples.

Structure and Chemical Composition of Oxide Films Formed in High Temperature Water on Austenitic Alloys

In order to study the structure and chemical composition of the oxide films (see previous Programme Progress Report[2]) the surface of specimens prepared at the JRC, were analysed by Auger electron spectroscopy (AES) and secondary ion mass spectrometry (SIMS).
The analyses were performed under research contract by Dornier System.
Detailed investigations were carried out on two specimens:

- reference specimen made of non-oxidized stainless steel tube
- surface oxidized in oxygen-free, but hydrogenized water during 1,800 hours.

The following conclusions can be drawn on the use of AES and SIMS:

1 The concentration depth profiles obtained from AES and SIMS combined with surface removal by ion bombardment, give very detailed information about the elemental distribution in the oxide films.
2 High surface roughness reduces the precision of the concentration measurements, but not in a significant way.
3 In spite of the very low concentration of cobalt (0.06-0.2%) its depth distribution in the oxide film can be measured with SIMS.
4 SIMS has to be preferred to AES for the determination of chromium in presence of large quantities of oxygen and for concentrations lower than 1%.

5 AES and SIMS can be utilized for the analysis of oxide films in the thickness range between tens of Å (thin passivation films) and a few microns, which is the lower limit for the application of the scanning electron microscope (SEM) microprobe.
This is very important because oxide films on austenitic alloys fall largely in that thickness range.

In addition to the AES-SIMS investigations, scanning electron microscope (SEM) analyses were carried out on surfaces and surface sections prepared from tube segments cut from the small high-temperature water test loop used for the oxidation of the surfaces.
The following specimens were analysed:

- non oxidized reference specimen for surface roughness investigations
- specimens oxidized at 300°C in oxygenized water for 165, 750 and 1200 hours
- specimen oxidized at 300°C in oxygen free, but hydrogenized water for 1,800 hours

The optical and SEM micrographs and SEM microprobe concentration analyses obtained on the reference specimen and on the specimen oxidized for 1,800 hours, were used for the interpretation and confrontation with the AES and SIMS measurements mentioned above.

The SEM micrographs have shown that the oxide films formed on austenitic steel at 300°C in hydrogenized water are much thicker than the films formed under the same conditions in pure oxygenized water.

A report on the results of these studies is in preparation.

The depth distribution of cobalt in the oxide film can be measured with SIMS; however, the surface resolution of SIMS is low (analysed area ~ 0.1 cm^2). AES has a high surface resolution (analysed area ~ 10^{-6} cm^2) but it is not sensible enough for the detection of concentrations below 1%. In addition instruments are not available for the investigation with SIMS and AES on contaminated samples.
Thus for the investigation on the surface distribution of Co-60 in oxidized austenitic steel we are considering the possible use of autoradiography.

Mechanisms of Decontamination Processes

For the study of the mechanisms of decontamination special samples are in preparation (1,800 hours of oxidation in the small high temperature water test loop and 5,000 hours of oxidation in a thermal exchange test tube).

Evaluation of the Decommissioning of the Ispra-I Reactor

The Ispra-I reactor is a 5 MW research reactor which had been in operation more than 10 years. It is a reactor cooled and moderated by heavy water with a graphite reflector. The scope of the present activity is to evaluate the problems which will be encountered in the decommissioning of this reactor and to define which are the problems in common with the decommissioning of power plants. If areas of general interest are found, a research programme based on the decommissioning of Ispra-I as a test case will be proposed.

A general mapping of the activity levels in the various parts of the reactor is in progress. Only the activities of the concrete of the biological shielding and of C-14 in the graphite reflector are still to be measured.
A general scheme of the dismantling of the reactor core has been prepared. For the most active parts (thermal shielding and reactor internals) the most practical solution seems dismantling under water. This would require a preliminary operation to modify the biological shielding in order to make possible the formation of an internal pool.
A report is in preparation.

COLLABORATION WITH EXTERNAL ORGANIZATIONS

Contract with Dornier System for the characterization of oxide layers by means of SIMS and AES techniques.

CONCLUSIONS

The systematic study of the chemical decontamination has been continued on inactive and contaminated samples. SEM analyses and radioactivity measurements made possible to draw conclusions on the effect of various stages of the chemical treatment.

The analyses carried out by SIMS and AES have demonstrated the high potential of these techniques for the study of the structure and chemical composition of the oxide films.
These analyses have been complemented with SEM investigations.

Concerning the decommissioning of the Ispra-I reactor, the problems related to the dismantling operations have been evaluated.

PLANNED ACTIVITIES

Systematic Study on Chemical Decontamination

The effect of the second treatment with AP on heavily oxidized samples will be investigated as far as concentration and temperature is concerned.

Study of the Physico-Chemical Structure of Oxide Layers

Due to the very interesting results obtained with SIMS, AES and SEM, the study of the structure of the oxide layers with these techniques will be continued.

Evaluation of the Decommissioning of the Ispra-I Reactor

If the evaluation indicates that the decommissioning of the Ispra-I reactor can produce information of interest for power plant decommissioning, a programme proposal for the plan 1980-1983 will be prepared. Consultation with experts of Member Countries will be performed

REFERENCES

1 R. ROOFTHOOFT: «La décontamination dans les centrales nucléaires refroidies à léau légère - Résultats dúne enquête dans les centrales Européennes», EUR 5898f (1977).

2 Management of Nuclear Materials and Radioactive Waste - Programme Progress Report of the Joint Research Centre, January-June 1978, No. 3528

Conclusions

3. CONCLUSIONS

In the second semester of 1978 the programme has been executed in a satisfactory agreement with the planning.

The following main comments on the obtained results and planned activities apply to the various projects.

Evaluation of Long-Term Hazard of Radioactive Waste Disposal

In the field of the waste hazard analysis the work for the application of the methodology developed at Ispra, to quantify the probabilistic value of the geological barrier in the Boom (Belgium) clay formation, has been almost completed.

The model used to calculate pathways and dose rates to man has been applied to an hypothetical waste repository, to compare on the basis of their long-term risks the one-through strategy and the uranium/plutonium recycle strategy in LWRs.

In the field of the radiation damage studies on glasses, the post-irradiation analyses on the glass samples irradiated in the HFR reactor have been completed. The results obtained have shown the validity of the damage simulation by fission fragments. This method, developed at Ispra, has the advantage to make possible the simulation of extremely long damage period without increasing considerably the experimental effort. A new irradiation experiment is in preparation corresponding to a damage period for vetrified HAW of about 10^6 years.

Laboratory experiments and theoretical investigations are also carried out in order to provide basic information for the interpretation of the radiation damage phenomena in vetrified waste.

For the study of the interaction of actinides with geological media, experiments on plutonium migration in sand column have been completed. Experiments on the migration of plutonium and americium in columns of the soil which overlies typical clay formations, are in progress.

In the field of actinides monitoring, the collaborative programme for an Integral Experiment on the plutonium waste measurement system of the Dounreay Nuclear Power Development Establishment (DNPDE) has been started.

The planning of future activities for the project Evaluation of the Long-Term Hazard of Radioactive Waste Disposal is shown in Table I.

Chemical Separation and Nuclear Transmutation of Actinides

The JRC activities in this field are planned in such a way to have a maximum of information merging in the second half of 1979. It is, in fact, intended to prepare by the end of 1979 a major report dealing with a critical evaluation of the feasibility of the chemical separation and nuclear transmutation of actinides.

In the field of the chemical separation of actinides, considerable progresses have been made in the study of the experimental conditions which make possible to keep plutonium in an extractable form during the operations of concentration and denitration of the HAW solutions.
The positive results obtained indicate the possibility of a simultaneous separation of Pu, Am and Cm in a single extraction step of the HDEHP and TBP processes. Engineering evaluations of the three flow-sheets, proposed for the HAW partitioning, have been initiated.

In the field of the assessment studies on nuclear trasmutation progresses have been made in reactor physics calculations, fuel element design, evaluation of shielding problems and of risk.

The planning of future activities for the project Chemical Separation and Nuclear Transmutation of Actinides is shown in Table II.

Decontamination of Reactor Components

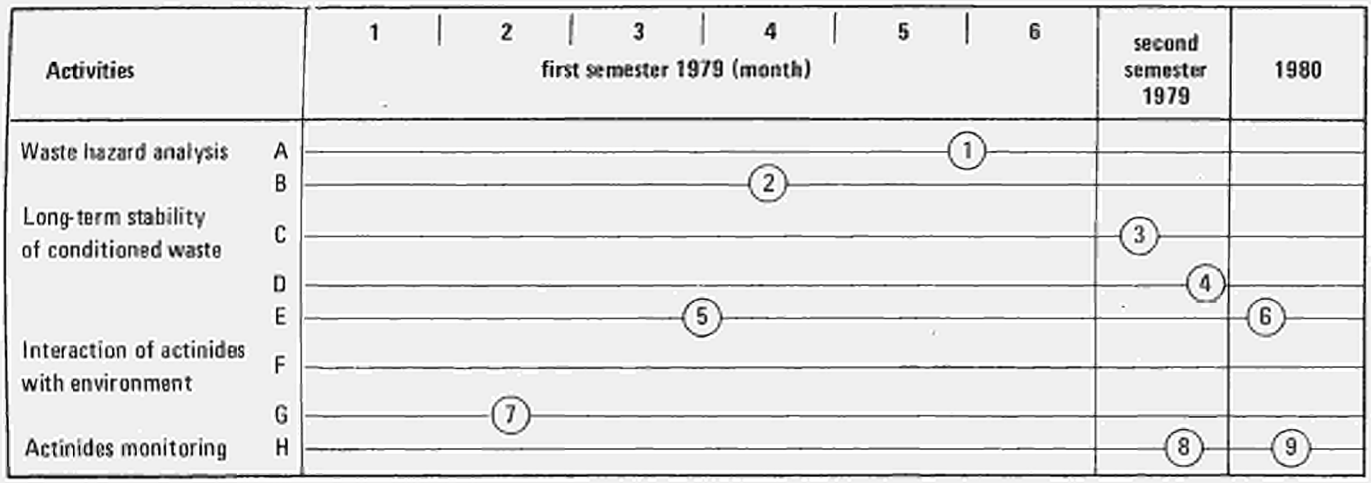
A large number of analyses using scanning electron microscopy (SEM), Auger electron spectroscopy (AES) and secondary ion mass spectrometry (SIMS) have been performed in order to investigate the physico-chemical structure of oxide layers and the mechanisms of chemical decontamination. This study is expected to produce data useful for the optimization of the decontamination processes in nuclear power plants.

The planning of future activities is shown in Table III.

For further information, please contact the Programme Manager

PLANNED ACTIVITIES AND IMPORTANT MILESTONES (JANUARY - JUNE 1979)

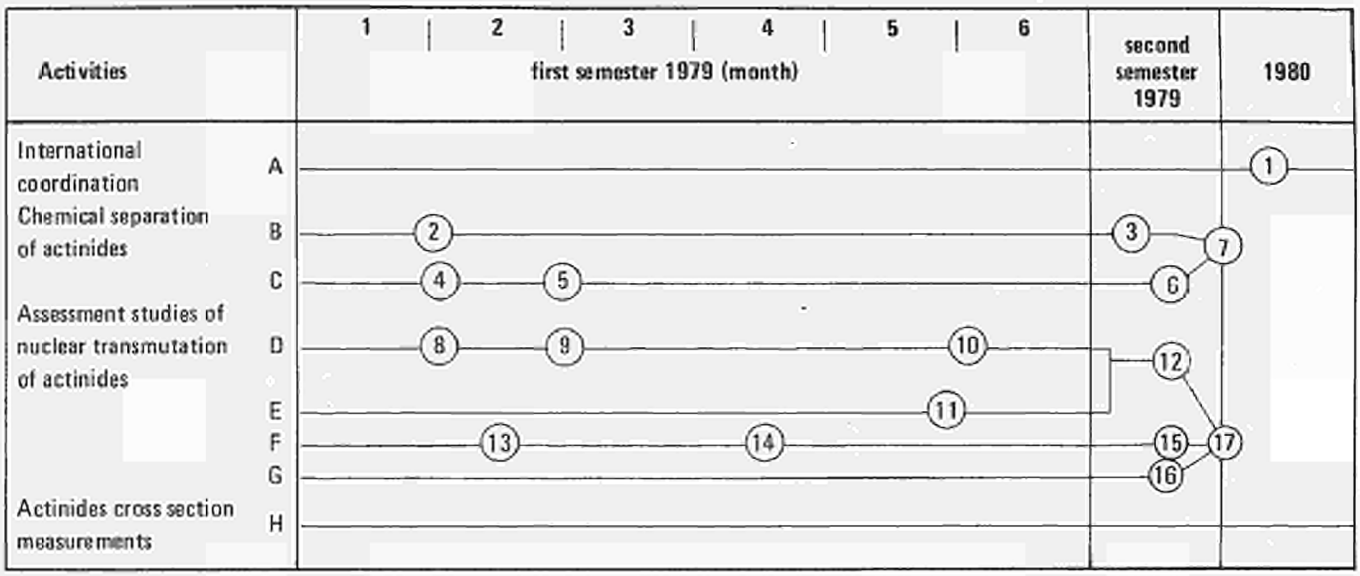
Table I – PROJECT 1: Evaluation of the Long-Term Hazard of Radioactive Waste Disposal



A	Modelling of failure of geological disposal	1	Final report on the application of FTA to the Belgium clay formation
B	Actinides distribution in environment following failure of geological barrier	2	Application of the deterministic section of the model to the Belgium clay formation
C	Leaching test on vetrified waste	3	Completion of the experiments with water in conditions of geological disposal
D	Leaching test on bituminized waste	4	Completion of the experiments oncontiminized waste
E	Experiments of radiation damage in glasses	5	New irradiation of glasses in the HFR reactor
		6	Completion of the experiments on the validity of accelerated tests
F	Interaction with abiotic environment	7	Conclusion of initial column experiments with leached actinides
G	Interaction with biosphere:		Collection of data and promotion of activities in a strict link with the indirect programme Radiation Protection
H	Plutonium waste monitoring	8	Revision of the Guide
		9	Completion of the integral experiment in a reprocessing plant

PLANNED ACTIVITIES AND IMPORTANT MILESTONES (JANUARY - JUNE 1979)

Table II — Planned Actinides and Important Milestones for the Project 2: Chemical Separation and Nuclear Transmutation of Actinides



A	International coordination in the framework of the OECD Nuclear Energy Agency	1	Organization of an international meeting
B	OXAL process	2	Completion of fully active experiments on separation of actinides from rare earths
		3	End of laboratory experiments on OXAL process and completion of preliminary engineering evaluations
C	Solvent extraction	4	Completion of fully active experiments on HDEHP latch extraction and HAW denitration
		5	Initiation of countercurrent experiments on simulated HAW extraction by HDEHP or, alternatively, fully active experiments on TBP latch extraction
		6	Completion of aset of laboratory experiments on solvent extraction and of engineering evaluation studies
		7	Report on the feasibility of actinide separation
D	Reactor physics calculations	8	Establishment reactor physics calculation methods for FBR and THTR
		9	Generation of nuclear data set for FBR and THTR
		10	Completion of reactor physics calculation for FBR, THTR and LWR
E	Fuel element design	11	Proposal of fuel elements for FBR and LWR
		12	Elaboration of an overall recycle strategy
F	Cost and risk analysis	13	Establishment of the methodology of risk assessment
		14	Establishment of cost calculation procedure
		15	Results of cost and risk assessment
G	Study of the implications of actinide recycling on the fuel cycle	16	Modification in nuclear plant and transportation
		17	Report on the feasibility of nuclear transmutation
H	Measurement of the cross sections of Am 241 and Am 243		

PLANNED ACTIVITIES AND IMPORTANT MILESTONES (JANUARY - JUNE 1979)

Table III — Planned Activities and Important Milestones for the PROJECT 3: Decontamination of Reactor Components

Activities	1	2	3	4	5	6	second semester 1979	1980
	first semester 1979 (month)							
Decontamination of reactor components A							(1) (2)	
B							(3)	
C				(4)				
D		(5)						

A Systematic study on chemical decontamination

B Study on the physico-chemical structure of oxide layers

C Partial decontamination using thermal-redox cycling

D Evaluation of the decommissioning of the Ispra 1 reactor

(1) Completion of the study on the APAC system

(2) Explorative tests on total decontamination

(3) Decision point

(4) Decision point

(5) Redaction of an operational-proposal Decision point

4. JRC PUBLICATIONS

Published or Presented

1. G. BIRKHOFF, L. BONDAR: «Monitoring of Plutonium Contaminated Solid Waste Streams - A Guide for Design and Analysis of Monitoring Systems. Chapter IV: Application of Passive Neutron Assay» - EUR 6027e, 1978.
2. F. GIRARDI: «Removal of Long Lived Alpha-Emitters from Radioactive Waste» - Paper presented at the 18th Annual Intern. Conference of the Canadian Nuclear Association, Ottawa (Canada), June 1978.
3. K. WISSHAK, F. KAEPPELER, W. HAGE: «Measurement of the Sub-Threshold Fission Cross Section of Pu-240 and Am-241» - Paper presented at the Intern. Conference on Neutron Physics and Nuclear Data for Reactor and other Applied Purposes, Harwell (U.K.), September 1978.

Submitted for Publication or Presentation

1. G. BIGNOLI, G. BERTOZZI: «Modeling of Artificial Radioactivity Migration in Environment. : a Survey» - To be published as EUR Report.
2. H. SCHNEIDER, A. CARETTA: «Functional Relation of Risk to Input Data Uncertainties» - to be published as EUR Report.
3. A. SALTELLI, A. AVOGADRO, G. BERTOZZI: «An Assessment of Plutonium Chemical Forms in Groundwater and their Migration through Porous Media» - To be presented at the Workshop on the Migration of Long-Lived Radionuclides in the Geosphere, CEC-OECD, Brussels, January 1979.
4. G. BERTOZZI, M. D'ALESSANDRO, F. GIRARDI: «A Risk Analysis Methodology for Deep Underground Radioactive Waste Repositories» - To be presented at the Colloque Intern. sur l'Evacuation des Déchets Radioactifs dans le Sol, IAEA/NEA, Otaniemi (Finland), July 1979.
5. A. AVOGADRO, F. LANZA, C. N. MURRAY: «Leaching of Borosilicate Glasses and Transport of Transuranic Nuclides through Deep Aquifers» - To be presented at the Colloque Intern. sur l'Evacuation des Déchets Radioactifs dans le Sol, IAEA/NEA, Otaniemi (Finland), July 1979.
6. F. LANZA, E. PARNISARI: «Evaluation of Long-Term Leaching of Borosilicate Glass in Pure Water» - To be presented at Intern. Symp. on Ceramics in Nuclear Waste Management, Cincinnati (USA), April 30-May 2, 1979.
7. M. ANTONINI, F. LANZA, A, MANARA: «Simulation of Radiation Damage in Glasses» - To be presented at Intern. Symp. on Ceramics in Nuclear Waste Management, Cincinnati (USA), April 30-May 2, 1979.
8. L. CECILLE, F. GIRARDI, F. MANNONE, F. MOUSTY: «Separation of Actinides from Purex HAW Raffinates: Development of Experimental Studies at JRC Ispra Establishment» - To be presented at the Actinide Separation Symp. - Pacific Chemical Conf., Honolulu, April 1979.
9. O. GAUTSCH, F. LANZA, F. VAN RUTTEN, P. WEISGERBER: «Decontamination of Water Cooled Reactors Primary Circuits and Components: A Bibliographic Review.» - To be published as EUR Report.
10. G. BIRKHOFF: «Monitoring of Plutonium Contaminated Solid Waste Streams - A Guide for Design and Analysis of Monitoring Systems. Chapter V: Application of Active Neutron Assay» - To be published as EUR Report.

LIST OF AUTHORS

Programme Manager: M. Bresesti
Assistant to the Programme Manager: J. Ley
Scientific Coordinator: F. Girardi

CONTRIBUTORS

Project 1:

A. Avogadro, G. Bertozzi, G. Bignoli, G. Birkhoff, L. Bondar, A. Caretta, M. D'Alessandro, F. Girardi, S. Kumpf, F. Lanza, P. Lensi, A. Manara, N. Murray, J. Schneider, T. Visani.

Project 2:

L. Anselmi, L. Cecille, H. Dworschak, M. Forte, F. Girardi, W. Hage, B. Hunt, F. Mannone, F. Mousty, E. Schmidt, E. Zamorani
L. Koch (JRC - Karlsruhe), F. Kaeppeler, K. Wisshak (Institute for Applied Nuclear Physics, KFK, Karlsruhe)

Project 3:

F. Lanza, P. Weisgerber.

ERRATA CORRIGE

p.8 1st column, line 21: delete important

p.8 2nd column, line 5: delete superscript 5

p.9 Fig. 1: for Reased read Released

p.10 Fig. 2: read: Metallographic Photograph of the Surface of a Leached Bitumen Sample - 40% $NaNo_3$ - Showing the Homogeneity of the Distribution

p.11 1st column, line 6: 3) To study the dependence of stored energy on the type of bombarding particle and impurities content

p.11 1st column, line 7: delete Applying ..... equation

p.11 Fig. 2: read: 1.7 not 17.

p.13 1st column: for PuO_3 read PuO_2

p.14 Fig. 2: b) Read: Towards ....

p. 17 Line 9: read UO_2

p.18 Fig. 1: for Dow E50 read Dowex 50

p.18 Table I: (2) read not detected

p.20 Fig. 1: for Fins read Pins and 8.92 not 18.92

p.22 Table IV: read 2.5 m Rem/h

p.32 Milestone 7 shouldappear on line F interaction of actinides with environment

p.32 Point 4: read Completion of the experiments on bituminized waste

p.33 Point 4: read batch extraction ; Point 5: read batch extraction.

p.34 C: read thermal-redox

CDNA07229ENC