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Management of Nuclear Materials and Radioactive Waste

PROGRAMME PROGRESS REPORT

January – June 1978

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 January – June 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 used to calculate pathways and dose rates to man has been refined.
Experiments have been carried out to determine leaching rates of vitrified high activity waste using water compositions related to specific geological disposal concepts.
Experiments have been set-up to study the interaction of actinides with ground water and geological media following their eventual leaching from vitrified waste.
An integral experiment on the monitoring of plutonium contaminated streams has been prepared in collaboration with the Dounreay reprocessing plant.

Project 2 : Chemical separation and nuclear transmutation of actinides

For the chemical separation of actinides from HAW, oxalate precipitation (OXAL process) and solvent extraction by HDEHP and TBP are being investigated. The tests of the flow-sheets on fully active HAW solutions are under way.

On the basis of different strategies considered for actinides transmutation in nuclear reactors, the work for the design of an actinide fuel element has been started.

Project 3 : Decontamination of reactor components

Studies are being carried out on the chemical decontamination, on the physico-chemical structure of the oxide layers and on the mechanisms of the decontamination process.
An evaluation is in progress on the possibility and interest to execute the decommissioning of the Ispra 1 reactor as a test case.

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 Research. 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 organized 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.

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2. PROJECTS

2.1. THE EVALUATION OF LONG TERM HAZARD OF RADIOACTIVE WASTE DISPOSAL

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 strengthen the links between the two programmes.

2.1.1 Waste Hazard Analysis

OBJECTIVES

The aim of this study is to get a comprehensive view of radioactive waste hazard, with particular emphasis on the quantitative value of the barrier system placed between waste and mankind. This aim will be pursued through the development and application of assessment methodologies, such as Fault Tree Analysis for the probabilistic assessment of the value of geological containment, and critical pathway analysis for the determination of environmental levels of radioactive pollution and corresponding dose rates to man.

For the first semester of 1978 the planned activities were:

A. Application of Fault Tree Analysis to specific disposal sites (Belgian clay formations).

B. Refinement of Model 1, used to calculate pathways and dose rates to man [1], through more careful assessment of waste inventories related to different fuel cycle options, and more detailed leaching models for the different conditioned waste types.

C. Development and application of new data handling techniques, aiming at ameliorating the probabilistic analysis of the geological retention barrier.

RESULTS

A. The Belgian clay formation of Boom was chosen during 1977 for the development of a site-specific application of the Fault Tree Analysis methodology, to quantify in probabilistic terms the value of this type of geological formation; the necessary geological information has been collected through a close collaboration with the C.E.N. of Mol.

   Three different Fault Trees have been constructed, having as their "Top Events" the releases of radioactivity towards groundwater, land surface and atmosphere, respectively. About thirty "Primary Events" have been identified; their probability levels are now being quoted.

B. Three different LWR fuel cycles have been considered, namely:

   - the once-through strategy,
   - uranium recycle only,
   - uranium and plutonium recycle.

   For each strategy, the quantities of wastes and their radiological

properties and chemical composition, as well as conditioning methods were examined; for each conditioned waste type a rough leaching model was defined, and the corresponding actinide release rates were calculated as a consequence of a flooding accident of the repository.

The disposal of unprocessed spent fuel elements exhibits the largest potential for releasing important flows of long-lived alpha-emitters towards the environment; however, retention processes in the deep subsoil between the geological formation and the biosphere could have, in most cases, the capability of assuring a very long delay before release of radionuclides into the environment. Therefore, a detailed evaluation of the parameters controlling the migration of radioelements in the subsoil has been undertaken.

C. A new data handling technique permitting the treatment of data in the form of probability histograms, has been developed and applied to Model 1, already described in [1, 2].
With this technique the parameters affected by large uncertainties can receive distributions of numerical figures, having probability levels associated with them; various possible results are then obtained in the form of a histogram, where different intervals of values are indicated with the corresponding probability levels.

An example of an output histogram is shown in Fig. 1, where different release rates of plutonium from bituminized waste are associated to different probabilities.

These techniques have been described in two papers, presented at international conferences [3, 4].

COLLABORATION WITH EXTERNAL ORGANIZATIONS

The collaboration established with C.E.N.-Mol for assessing the suitability of certain clay formations to accommodate waste produced by Belgian nuclear power plants, has already been described above.

A collaboration with laboratories involved in the indirect programme Radiation Protection is foreseen; preliminary contacts have been established with some of them, the objective being to provide the necessary data input for the radioisotope distribution models.

The second meeting of the Working Group Risk Analysis Linked to the Disposal of Radioactive Waste in Geological Formations (sub-group of fiche 7 of the Indirect Action Programme Management and Disposal

of Radioactive Waste) was held at Ispra on May 18-19, 1978. The aim of the group is essentially to facilitate the exchange of information among experts to point out research needs for a better risk evaluation, and to favour the establishment of common criteria for risk evaluation.

CONCLUSIONS

The methodologies developed at the JRC-Ispra for the probabilistic assessment of the safety of the disposal in a generic geological formation are well adaptable to real disposal sites.

The application of both probabilistic and deterministic models has shown that, although the conditioning ways and properties of the various waste types need to be better defined, the risk associated to a geological repository is rather small, both in terms of event probability and of resulting dose rates to man.

The importance of the soil retention barrier is great; its efficiency depends on many parameters which are very scarcely known: a great deal of investigation should be devoted to this topic.

PLANNED ACTIVITIES

The Fault Tree Analysis for a repository in clay formations will be completed by the end of 1978, so that a quantitative probabilistic assessment will be available.

The deterministic study described under item B. will be continued until completion of a Model 2. Wide use will probably be made of the data histogram treating techniques, for both kinds of studies. A careful examination of the efficiency of the soil retention barrier is also planned.

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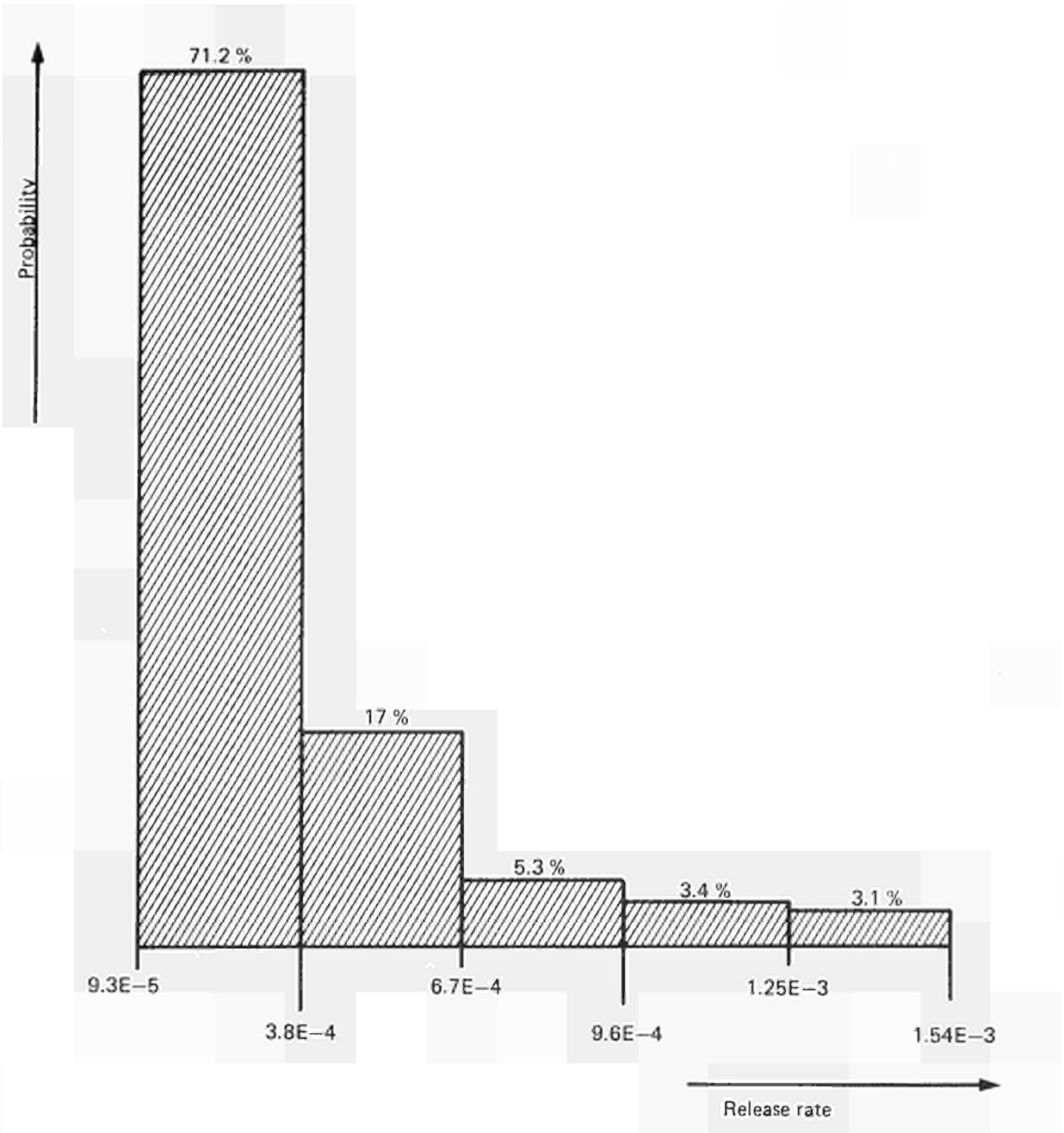

Fig. 1 – Probability histogram for different release rates (Ci/y) of plutonium from 1 ton of bituminized waste

2.1.2 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 Waste Hazard Evaluation studies.

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

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

RESULTS

Post-Irradiation Examination of Irradiated Glasses

The irradiation carried out in the HFR at Petten was intended to simulate, by the damage caused by fission fragments, the damage produced by the alpha-particles. A maximum of fission density of $4 \cdot 10^{17}$ fissions/cm^3, which corresponds to $2 \cdot 10^{22}$ displaced atoms/cm^3, was calculated for the irradiated samples.

The measurements of the stored energy, which were planned for the reporting period, will be completed only in July 1978, due to the delay in the delivery of the DTA (Differential Thermal Analysis) apparatus. A final report on the experiment will be prepared.

Glass Leaching Tests

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

Leaching tests of 1 year duration at 80°C and 50°C have been completed. The results of the analysis carried out by X-ray dispersive technique, using a scanning electron microscope, on the surface layers after 1 year leaching (Table 1), indicate a depletion of cations, much higher than was expected. The effect is more important after the leaching test at 80°C.

Additional surface analyses using a different method (spectrographic emission), are planned in order to confirm this effect.

Leaching tests of 4 months duration have been carried out at 80°C using water conditioned by percolation through a silica sand (silicon content 10 ppm).
No significant difference in weight loss and surface layer composition was observed in comparison with similar tests carried out with pure water.
This fact seems to confirm the hypothesis that silicon release is due essentially to a colloidal release.

Similar leaching tests with water percolated on clay are planned.

TABLE 1 - Variations of some constituents of the surface layer composition (weight %) after leaching tests of 1 year duration at 80 and 50°C

	Si	Na	Fe	Ni	Ce	U	Sm
Initial conc.	48	12	3.25	0.32	0.76	1.38	1.12
Final conc. test at 80°C	1.6	1.4	1.1	0.02	0.2	0.17	0.17
Final conc. test at 50°C	2.9	2.0	1.1	0.02	0.4	0.7	0.7

Stability Tests on Bitumen

Some preliminary tests on bitumen have been initiated 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.
From the preliminary tests it appears clearly that the model of the dissolving sphere utilized for the glasses is not applicable to bitumen. A model of constant leach rate coupled with a constant surface seems more likely. A series of tests have been initiated with the aim of measuring the amount of salt leached and the salt distribution inside the sample.
No significant result has been obtained up till now.

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

The energy stored by amorphous silica samples irradiated as indicated in Table 2 has been measured by thermal analysis. A large amount of

energy release (150 - 250 cal/g) is observed.
The annealing curve shows two maxima at about 470° and 610°C (see Fig. 1). The energy stored during irradiation may be attributed to atomic defects, SiO_2 defective units, both leading to long-range structural distortion, and/or to local distortion within the Si-O tetrahedra caused by leakage or modification of the Si-O bond, connected with ionizing collision.

The energy released after our irradiation is much larger than those obtained in previous measurements [1, 2], characterized by a higher irradiation temperature and much lower damage rates.
Therefore the energy stored during irradiation of amorphous silica seems to depend strongly on the temperature of irradiation, the dose rate and the type of particle.
At low temperature and at high dose rates a large amount of energy is stored.

A communication was presented at the International Topical Conference on "The Physics of SiO_2 and its Interfaces", at the T. J. Watson Research Center of York Town (N. Y., USA). The work performed at Ispra was of considerable interest to those scientists dealing with radiation damage in SiO_2.

TABLE 2

Samples	Radiation	Energy (MeV)	Total dose (ion/cm^2)	DPA	Dose rate (ion/cm^2.s)
SiO_2	α	1.00	$1 \cdot 10^{18}$	$\sim$ 2.5	
SiO_2	Ni^{+6}	46.50	$1.6 \cdot 10^{15}$	0.16	$4.6 \cdot 10^{11}$
SiO_2	Ni^{+6}	46.50	$5 \cdot 10^{15}$	0.5	$6.9 \cdot 10^{11}$

COLLABORATION WITH EXTERNAL ORGANIZATIONS

The collaboration with AERE, Harwell has been pursued. Some preliminary contacts have been initiated with scientists from the Catholic University of Washington with regard to the characterization of special glasses. Moreover, some specific discussions have been held with the Naval Research Laboratory, Washington, on defective centers in SiO_2 and with the Sandia Laboratories N. M., on optical spectra in irradiated fused silica.

PLANNED ACTIVITIES

For the second semester of 1978 the following activities are planned:

Radiation Damage on Glasses

Measurement of the stored energy in the glasses irradiated in the HFR Petten. Redaction of the final report.

Leaching Tests

Leaching tests on glasses with water percolated on clay.
Leaching tests on bitumen.

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

Further measurements are planned, using high-energy electrons, alpha-particles and heavy ions in order to detect the relative contributions, at various dose rate, of ionizing collisions to the various physical properties of glasses.
Other quantities such as optical absorption will also be measured for a better understanding of the damage and annealing processes at high dose rates.

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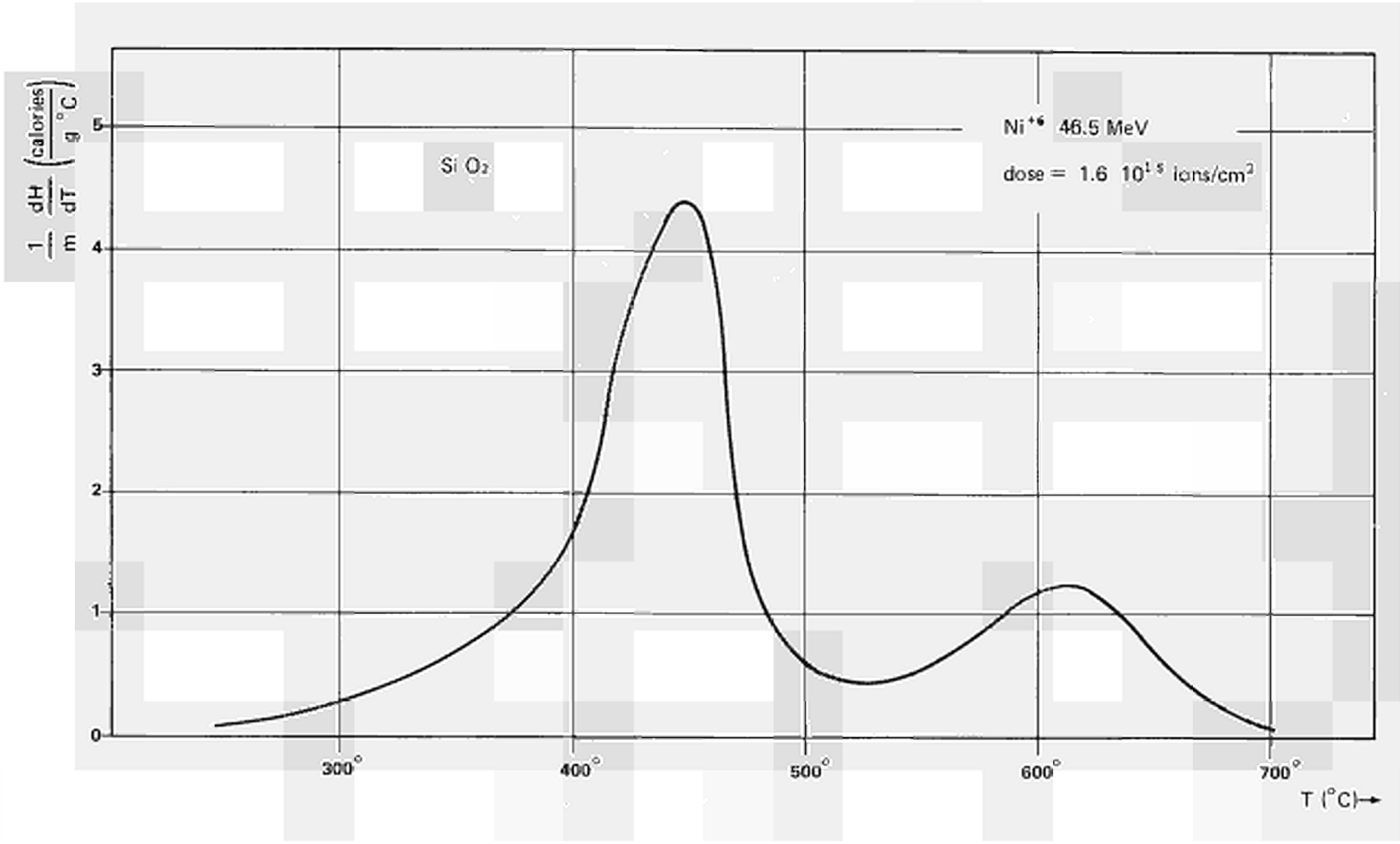

Fig. 1 — Rate of energy release after heavy ions irradiation at room temperature

2.1.3 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 first semester of 1978, the planned activities were:

a) The production of americium- and plutonium-spiked glasses and the development of methods for the determination of their physico-chemical forms in leached solutions.

b) Migration experiments with leached solutions through sandy soils and sediments under various simulated environmental conditions.

c) Identification of experimental studies needed for a better understanding of the interaction between actinides and the biosphere, to be carried out under the Radioprotection Programme of DG-XII.

RESULTS

a), b) Glasses containing Pu-238 and Am-241 have been produced with the techniques described in the previous Progress Reports. With these glasses the two lines of research defined in the objectives have been started.

Column experiments (height 200 mm, diam. 26 mm) with sand grain size between 200 and 400 µm, are at present being carried out. In the experimental system which was set up, water flows over crashed and sieved alpha-bearing glass and then through the column containing the soil. Water of a composition typical of the aquifer overlying the Boom clay formation of Mol, Belgium, has been used. This experiment will be continued for at least a period of 3 months. The break-through solution is being controlled and, at the end of the experiment, the column will be sliced and the alpha-activity measured in order to establish the actinide distribution profile.

Parallel to this work the leached solution is passed through an ultra-filtration system and then cationic-anionic resins. At the outflow of the last resin the solution is collected, co-precipitated with cerium ion and analyzed for alpha-activity.

This experimental system is at present running and will be (as above) continued for at least 3 months. The analysis of the distribution of activity in the different components of the chain will give an indication of the size and charge of the chemical species of the actinides liberated from the leached glass.

c) The JRC followed up the initial contacts made with various European laboratories (Progress Report July-December 1977) as well as with the Radiation Protection Programme of the Commission. The aim of this activity is to develop a harmonized series of experimental projects within the framework of the Radiation Protection Programme of the DG-XII, Brussels. Several of these proposals are to be considered by the ACPM Radiation Protection, scheduled for June 5-7, 1978. This approach was considered by the JRC and the Radiation Protection Programme to be the most effective solution to obtain a rapid start-up of those experimental activities which may contribute to the JRC risk assessment programme. It is envisaged that the JRC will actively contribute in this framework with the experimental work on the chemical speciation of actinides.

COLLABORATION WITH EXTERNAL ORGANIZATIONS

During the reporting period contact was made with the following national institutes:

- Fisheries Radiobiological Laboratory, Ministry of Agriculture, Fisheries and Food, Directorate of Fisheries Research, Lowestoft, United Kingdom,
- Biologische Anstalt Helgoland, Hamburg, Germany,
- Commissariat à l'Energie Atomique: CEN/Fontenay-aux-Roses and CEN/Cadarache.

CONCLUSIONS AND PLANNED ACTIVITIES

a), b) The experiments in progress will be continued and a detailed analysis of the results obtained from the first run will be carried out.

Column experiments with sand and clay mixtures under high lithostatic and hydrostatic pressures will be started in order to simulate, realistically as possible, the natural condition existing in the Belgian clay formations.

c) No special development is foreseen in the second semester of 1978.

2.1.4 Actinides Monitoring

OBJECTIVES

The study aims at the development of a methodology for plutonium waste monitoring.

In the planning for the reporting period we envisaged:

- to heighten the knowledge contained in chapter II, III, IV of our guide on Monitoring of Plutonium-Contaminated Solid Waste Streams [1],
- to write the chapter V of our guide on the Application of Active Neutron Assay [1],
- to test an on-line alpha-monitor for liquid waste.

RESULTS

1) Expertise

Chapters II, III and IV of our guide [1] have been submitted to a judgement by an external scientific institute. This expertise came to the conclusion that the adopted methodological approaches of chapters II and IV are fully valid, but some experiments are suggested for a better determination of the practical limits of some of the theoretical models.

Those experiments concern the removal cross section model for passive assay and the fast and thermal neutron induced fission effect in passive neutron assay.

As far as chapter III (gamma assay) is concerned, the conclusions will be available in the course of 1978.

2) Applicability of pulse-to-pulse correlations in the time domain (higher order coincidences)

Pulse-to-pulse correlations in the time domain yield information about the neutron emission process. For given spontaneous neutron emission processes, such information can be interpreted in terms of neutron multiplication effects.

A parametric study has been carried out by means of Monte Carlo simulations and statistical theory for determining the sensitivity of the pulse-to-pulse correlation technique relative to detector efficiency, ϵ, detection delay time, l, induced fission probability, P_{if}, and induced fission neutron multiplicity, $\bar{\nu}$.

The study shows the applicability of pulse-to-pulse correlation methods for rather high values of $\varepsilon > 20\%$, $P_{if} > 5\%$ (favoured by high $\bar{\nu}$-values).

3) Upgraded reference monitor for passive neutron assay

A first design of an upgraded reference monitor for passive neutron assay was elaborated. This design aims at the improvement of the neutron detection assembly as well as of the electronics. Components for this reference monitor have been ordered.

4) Integral experiment

An integral experiment on the monitoring of Pu-contaminated waste streams in a fuel reprocessing facility has been organized and prepared. This experiment aims at the demonstration of our monitoring concept as outline in the guide [1]. It is scheduled for a duration of two years, beginning July 1978.

5) Active neutron assay

Chapter V dealing with the application of the active neutron technique for plutonium waste monitoring, has been drafted partially.

6) Sample preparation

In order to minimize the measurement uncertainty caused by heterogeneity of wastes, a method of sample preparation by centrifuging has been set up. Preliminary experiments aiming at the definition of the influences of matrix materials, heavy particles size and densities have been carried out.

7) 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 have been started.

COLLABORATION WITH EXTERNAL ORGANIZATIONS

Contract with EUREX (CNEN, Saluggia, Italy) concerning the testing of on-line alpha-monitor (178-77 PIPGI, May 16, 1977).

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

Collaboration with the Plutonium Fabrication Plant (CNEN, Casaccia,

Italy) for the plutonium determination in solid waste.

Contacts with DNPDE (Dounreay, United Kingdom) in view of a collaboration on the monitoring of the contaminated solid waste streams.

CONCLUSIONS

The objectives for the reporting period have been reached as far as the neutron assay techniques are concerned. No progress was obtained for verification of the passive gamma reference monitor, because the completion of the apparatus is seriously delayed.
This apparatus will probably be available and operation only by the end of 1978. In view of this, new activities under the heading "sample preparation" and "X-ray pattern" have been started.

The testing of the on-line liquid alpha monitor in a fuel reprocessing plant has not yet been started due to delay in the operation of this plant.

The staffing of this programme activity was improved during the reporting period such that the difficulties concerning the establishment of the "Advisory Laboratory for Plutonium Waste Monitoring" (as announced in the Progress Report for July-December 1977) can be considered as overcome.

PLANNED ACTIVITIES

For the following semester are planned:

1) Experimental verification of the validity of the reference monitor for passive gamma assay,

2) As 1), but for passive neutron assay,

3) Development of a computerized system for pulse-to-pulse correlation techniques,

4) An integral experiment will be started in a fuel reprocessing facility,

5) Writing chapter V of our guide: Application of Active Neutron Assay,

6) Testing of an on-line liquid alpha monitor under operational conditions of a fuel reprocessing facility,

7) Development of sample preparation techniques,

8) Development of X-ray transmission techniques.

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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.

The report will also include an assessment on the separation of actinides from wastes of reprocessing plants other than HAW and an assessment on the different options for the disposal of separated actinides.

2.2.1 Chemical Separation of Actinides

OBJECTIVES

The activity performed during the reporting period was a continuation of the preceding experimental activity aimed at demonstrating, on a laboratory scale, the feasibility of the OXAL, HDEHP and TBP processes proposed for the HAW partitioning.
To this purpose during the reporting period it was planned to initiate HDEHP batch-extraction tests on fully active HAW solutions and countercurrent tests on simulated solutions.

The continuation of the OXAL process tests (oxalate precipitation and actinide/REs separation) on real HAW solutions, was also included in the planned activity along with the preparation of detailed flow-sheets and flow-diagrams needed for evaluating the engineering implications of each actinide separation process.

RESULTS

1. HDEHP Extraction Process

The possibility of reducing the acidity of HAW solutions without any previous actinide separation (direct HAW denitration), strongly depends on to what extent the coprecipitation of actinides during denitration can be prevented or minimized.

An acidity reduction step is in fact required whenever one of the current actinide recovery methods, operating under low-acidity conditions, will be applied to HAW raffinates for partitioning purposes. Usually, as the pH will increase, an important fraction of actinides (mainly Pu) will coprecipitate with other hydrolysable metal-ions (Mo, Zr, Nb, Fe) present in the HAW solution. In this respect the behaviour of plutonium during the preceding denitration test on simulated HAW [1] appeared rather unexpected and led to a second series of experiments.
The precipitate formed during the denitration tests was suitably washed with formic acid and redissolved by hydrochloric acid. The fraction of residual plutonium still remaining in the precipitate was in the order of a few tenths of percent of the initial plutonium amount. The measured values may probably be decreased using nitric acid as washing solution.

According to the indications of these tests it appears that the plutonium content of precipitates generated during the HAW denitration by formic acid could be minimized when suitable process conditions are applied. Consequently, plutonium, still in solution after denitration, could be coextracted with americium and curium and its pre-

vious separation before denitration would become unnecessary.

The reaction mechanisms are presently being investigated.

Further denitration tests will be performed on simulated and real HAW solutions.

The HDEHP batch-extraction tests on fully active and acidic waste solutions have been initiated using as a solvent two HDEHP organic solutions having the following compositions:

a) 0.25 M HDEHP + 0.17 M TBP in dodecane,
b) 0.25 M HDEHP in mesitylene.

For all the extraction tests the operating conditions were fixed as follows:

- the HAW solution was always previously filtered,
- an organic to aqueous phase ratio of 1, a stirring time of 4 min and a temperature of 25°C were employed,
- the phase separation was attained by decantation.

The first set of experiments carried out using both the solvent mixtures, showed the formation of precipitates clearly visible at the interphase. As in the case of simulated HAW, such interphase precipitates were eliminated by addition of $NaNO_2$.

For all the back-extraction tests 0.8 M oxalic acid was employed as a stripping solution, the other operating conditions being equal to those of the extraction tests.

The preliminary results obtained from extraction and back-extraction tests were satisfactory. After three extraction stages no plutonium activity was detected in both organic and exhausted aqueous phases. After three successive back-extraction stages, the overall plutonium stripped from the organic was almost quantitative.

2. TBP Extraction Process

The results of concentration-denitration tests on simulated HAW, already obtained during the preceding period [2], showed that, if the formation of plutonium-bearing precipitates has to be minimized, the reduction of the HAW acidity must be carried out under well-controlled process conditions. The setting up of the suitably equipped glass reactor vessel has been continued.

The concentration-denitration step is performed under constant-

volume conditions. Inside the reactor vessel the level of the solution is kept constant by an automatically operated addition of both formic acid and HAW solutions. The acidity is measured by conductimetry.
A set of concentration-denitration tests carried out using simulated HAW solutions, has shown that a $HCOOH/HNO_3$ molar ratio of about 0.7 is needed to provide during and at the end of the process a nitric acid concentration not lower than 5 M/l, as required to minimize the fraction of adsorbed plutonium [2].

The setting up of the equipment will be completed in the next months. Active runs will start in October-November 1978.

3. OXAL Process

Experimental tests on simulated HAW solutions, aimed at optimizing the actinide/REs separation step, have been completed. As previously reported [2], a process based on column extraction chromatography (HDEHP supported on LEVEXTREL) has been tested. Two LEVEXTREL/HDEHP columns, containing different amounts of adsorber have been utilized to remove plutonium and americium from the HAW solution, at different acidity conditions respectively.

Typical results obtained by simulated HAW solutions are illustrated in Figs. 1 and 2. 99.7% of plutonium and zirconium initially present was fixed on the first column (~0.7 ml of adsorber) after adsorption and washing steps. Both the elements were quantitatively eluted at 70°C by 1 M oxalic acid (Fig. 1). After deacidification of the HAW solution (pH = 2) americium and REs were separated from FPs by adsorption on the second column (~9 ml of adsorber). About 99% of americium was then eluted by a complexing solution (0.05 M DTPA + 0.5 M glycolic acid); 3.7% and 27% of the initial Ce-144 and Eu-154 activity were detected in the americium eluate.

Based on these results the actinide fraction should not contain more than 5% of the REs, as was originally anticipated.
To attain a higher degree of actinide/REs separation, if needed, an additional purification step has to be foreseen.

Experiments are still under way, in order to improve the performance of the process.

4. Engineering Implication of Actinide Separation Processes

Tentative flow-sheets and flow-diagrams of the three processes are presently being prepared. A critical evaluation of the three flow-

sheets in order to estimate the engineering implications will be initiated in the second semester of this year.

CONCLUSIONS

The activities are basically proceeding according to schedule, although the tests on fully active HAW are proceeding more slowly than expected. A revision of the planning may eventually be required in the next months.

Special attention was paid to the setting up of both denitration and concentration-denitration processes which minimize the actinide content of precipitates formed. This effort is motivated by the need of preventing as much as possible, during the waste partitioning cycle the production of new alpha-bearing waste.

PLANNED ACTIVITIES

During the following six months is planned:

- to initiate denitration tests on real HAW,
- to continue HDEHP batch-extraction tests on real HAW,
- to continue the experimental work on OXAL process by testing on a fully active laboratory scale the actinide/REs separation step,
- to proceed to a first evaluation of the engineering implications of the three flow-sheets.

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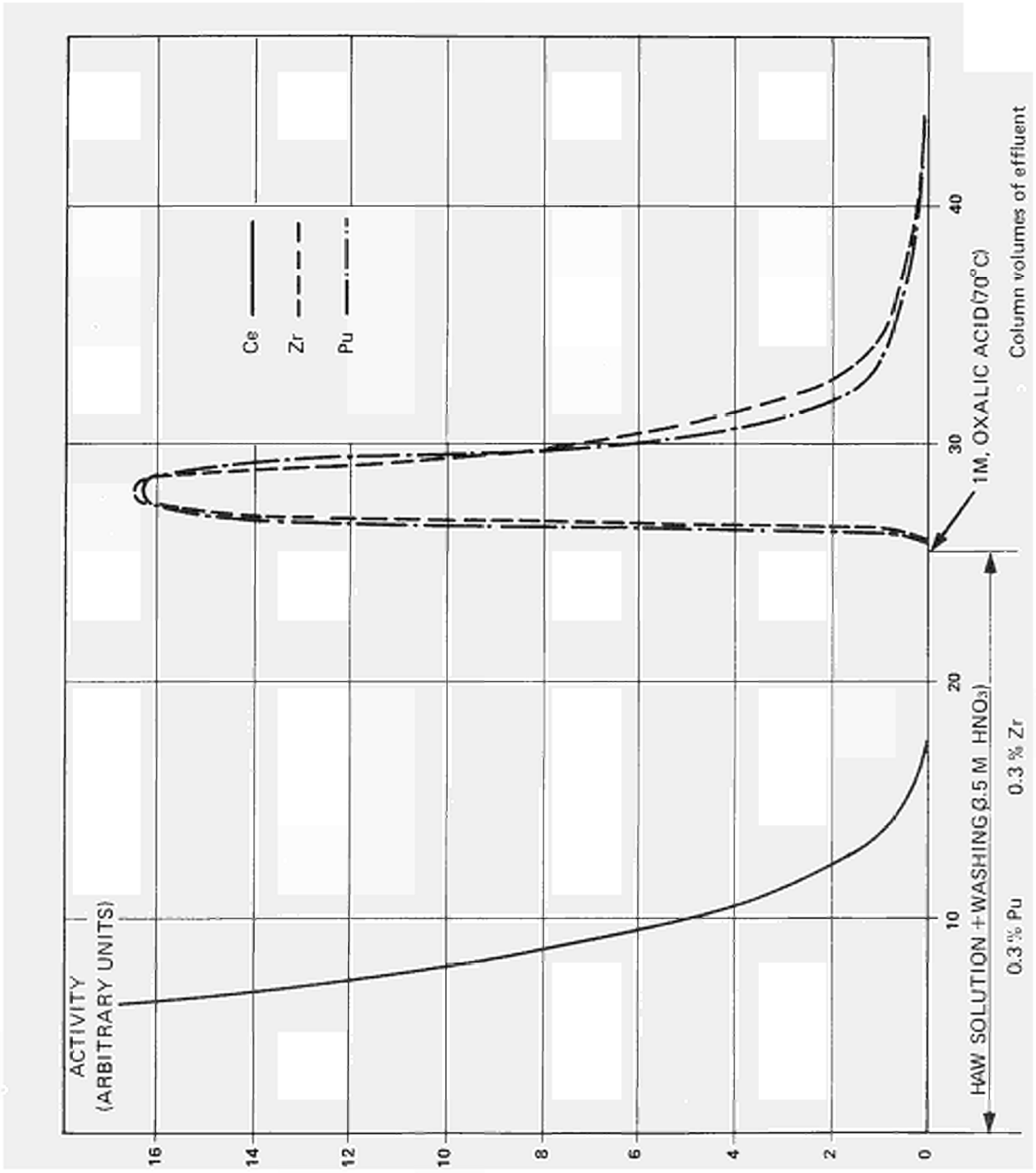

Fig. 1 – Actinide/R.Es separation by column extraction cromatography. Separation of Pu-Zr from Am-R.Es through the 1st LEVEXTREL/HDEHP column

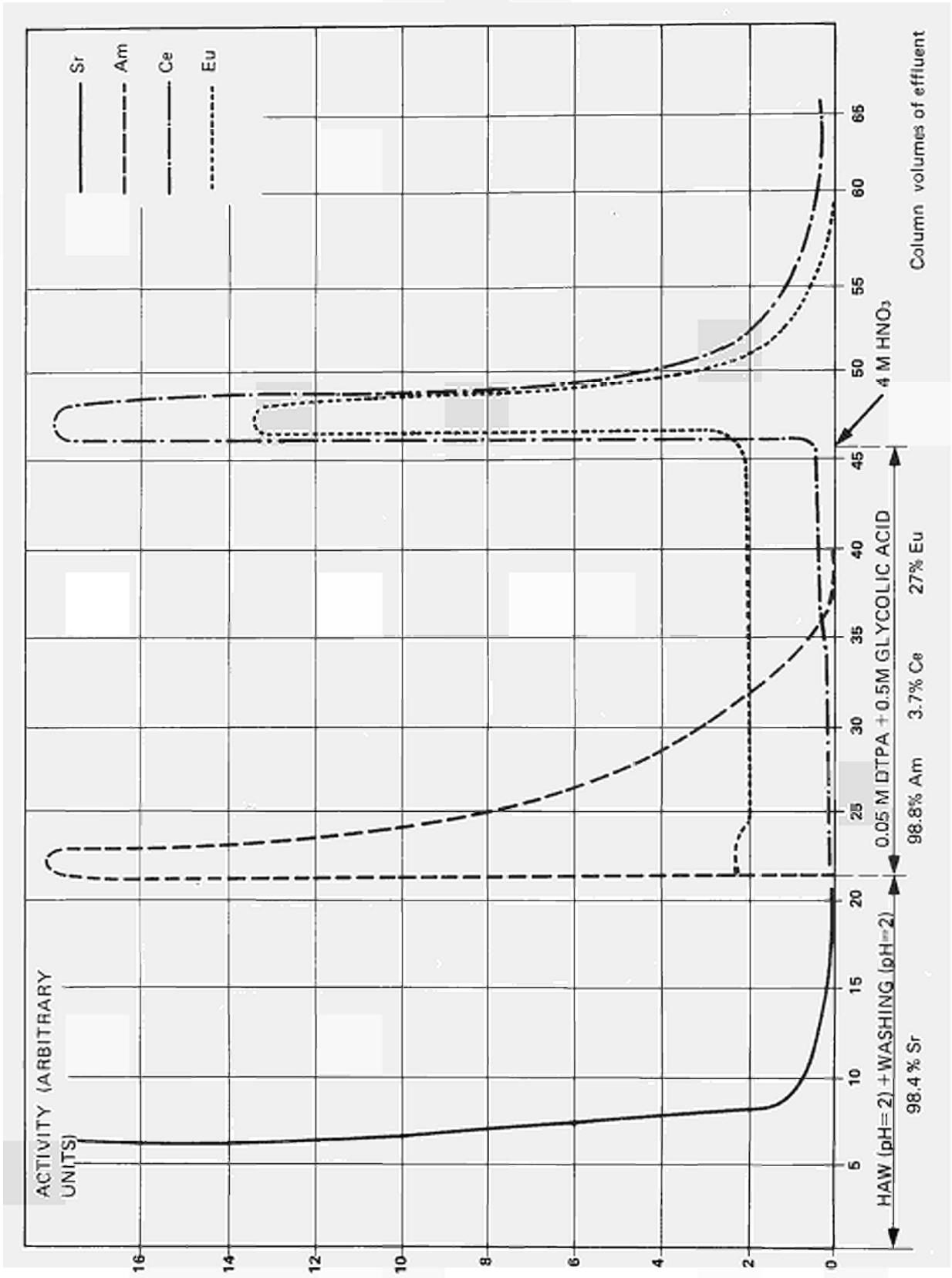

Fig. 2 – Actinide/R.Es separation by column extraction cromatography. Separation of Am from R.Es through the 2nd LEVEXTREL/HDEHP column

2.2.2 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 transmutation of actinides other than fuel in fission power reactors and to propose the best suited recycle strategy.

In the present reporting period, preparatory work regarding the fuel element design, improvement of the risk source assessment code and establishment of collaboration with external institutes were accentuated.

RESULTS

Selection of Recycle Strategies

Before work on fuel element design could be started, some fundamental decisions on the recycle strategy and the transmutation devices had to be taken. The criteria to be observed when selecting possible recycling schemes were:

1. High transmutation rate per in-core time period in order to minimize waste arisings during the various processing steps and the inventory of highly hazardous material.
   Consequently, reactors with high neutron fluxes, long in-core residence times of fuel elements and high attainable burn-ups should be chosen. Moreover, minor actinides should be irradiated in special regions of the reactor.

2. Small reactivity penalties due to minor actinides and fission product impurities introduced into the reactor.
   Consequently, fast reactors are to be preferred.

3. Availability of the technology at about the year 2000. Consequently, only reactors which are now in phase of design or already operative are taken into consideration. However, developing potential of the various design parameters which could improve the transmutation strategy should be included in the assessment.

4. Small perturbations of the normal reactor operation concerning flux and power distribution.
   This means that in case of a heterogeneous recycle scheme, the minor actinide-containing fuel elements should be spread over chosen core zones.

5. Possibly similar radiation levels of normal fuel elements and minor

actinide-containing fuel elements. Therefore, only reactors with Pu-recycling or recycling of U-233 are being considered. Moreover the application of diluting materials is indicated.

6. The change in fabrication, transport and reprocessing technology due to the presence of the minor actinides should be kept small and the implication on the fuel cycle costs should be minimized. Consequently, minor actinides should be contained in a minimum number of fuel elements which, however, before reprocessing, could be mixed with normal fuel elements. Recycle through a uranium-fed LWR will be excluded.

As a conclusion of these requirements, which are partly competing with each other, the following recycle strategies will be investigated in more detail:

- recycle through the core of a fast breeder reactor (quasi-heterogeneous),
- recycle through a light-water reactor with plutonium recycle (homogeneous),
- recycle through a pebble-bed reactor with spherical fuel elements (heterogeneous).

Review of Performance Data for Fuel Element Design of the Fast Breeder

In ref. [1], the performance data of various fast breeder fuel elements have been collected. They are reproduced in Table 1. The high maximum permissible linear power rating, maximum neutron flux and maximum burn-up led to the decision to choose the NA 1 design as reference fast breeder for transmutation.

Design Work for Minor Actinide-Bearing Fast Breeder Fuel Pins

Design work for the fuel element is based on the assumption that all geometric parameters which could influence the thermo-hydraulics of the coolant channel, remain constants. Thus, the outer diameter of the pin and the maximum permissible linear power rating shall not be varied.

Supposed that the target pin with 85% of theoretical density of the oxides, is going to be inserted into the reactor zone with the highest flux of

$$\phi = 8.9 \cdot 10^{15} \text{ n/cm}^2\text{.s}$$

the linear power ratings shown in Fig. 1 were obtained for the minor actinide compositions of Table 2.

TABLE 1 : CHARACTERISTICS OF FBRs

		FRANCE		U.K.			F.R.G.				USSR		JAPAN	U.S.A.			
		PHENIX	SUPER PHENIX	DFR	PFR	CFR	SNR 300	SNR 2	NA1	GC FBR	BN 350	BN 600	MONJU	EFFBR	CR BR	AIFO	GEFO
Reactor power (MW)	thermal	563	2910	72	600	2900	736		2500	2780	1000	1480	714	200	950	2400	2417
	electric.	250	1200	15	270	1320	312	1200	1000	1030	350	600	300	66	360	1002	1011
Temperatures (°C)	core inlet	400	395	200	400	400	377		430	273	300	380	390	290	387		
	core outlet	560	535	350	562	562	546		580	555	500	550	540	430	540		
	midd wall	650	620		660	620									636		
	clad (max)	700	700		700	700	670			700	680	700	700		640		
	fuel (max)						2400		(2800) 2400								
Dimensions (mm)	core height	850	1000		910	1000	950		955		1060	750	900		910		
	core ϕ	1390	3660		1470	2900	1780		2860		1580	2050	1780		1880		
	clad o.d.	6.6	8.65	6.0	5.84	5.84	6.0	7.6	6.7	8.2	6.1	6.9	6.5		5.84		
	clad i.d.			5.32					6.0								
	rad. gap																
Subassemblies		103	358		78	342	205		210				196		198		
Pins/subass.		217	271		325	325	169		331		169	127	169		217		
Fuel material		UO_2/* PuO_2	UO_2/ PuO_2	U	UO_2/ PuO_2	UO_2/ PuO_2	UO_2/ PuO_2		UO_2/ PuO_2		UO_2	UO_2/* PuO_2	UO_2/ PuO_2	U	UO_2/ PuO_2		
Pu in zone 1 (%)		18															
Pu in zone 2 (%)		25															
Pu in zone 3 (%)																	
Cladding material		ss	ss		ss	ss	ss		Incoloy 800		ss	ss	Zircon.	ss			
Power rating																	
Linear mean (W/cm)				320				452									
Linear max. (W/cm)		430	450		450	450	460		566		470	530	457	250	475		
$\overline{M}$ (MW/T H.M.)						160			143.9							116	155.6
Neutron flux $x10^{-15}$ ($n/cm^2.s$)	core (max)	7.2		2.5	8.5	11	6.0		8.92				4.0	4.7			
	core (mean)								6.35								
Fluence (max) at clad (n/cm^2)		$3 \cdot 10^{23}$	$4.6 \cdot 10^{23}$												$3 \cdot 10^{23}$		
Burn-up of core fuel (GWD(th)/T)	mean	50	70	53	70	70	55		79		50	90	80			67.7	104.5
	max						87		125	100					150		

TABLE 2 - Ni [gram-atoms/cm pin length]

	Np-237	Am-241	Am-242M	Am-243	Cm-242	Cm-243	Cm-244	Cm-245	Cm-246	Cm-247	Cm-248	Cf-249
FBR 1st cycle	3.23-3	4.06-3	7.23-5	2.47-3	1.22-4	7.83-6	1.78-4	5.68-6	1.74-7			
FBR equilibrium	2.26-3	2.28-3	1.56-4	2.57-3	1.11-4	6.59-5	1.33-3	1.74-4	1.03-4	1.16-5	5.56-5	4.52-7
U-LWR	7.27-3	6.23-4	1.16-5	1.53-3	7.31-5	1.09-6	4.69-4	3.07-5	3.59-6			
FBR 1st cycle without Np	-	5.99-3	1.07-4	3.64-3	1.80-4	1.16-5	2.63-4	8.39-6	2.57-7			
FBR equilibrium without Np	-	3.37-2	2.31-4	3.80-3	1.64-4	9.74-5	1.97-3	2.57-4	1.52-4	1.72-5	8.23-6	6.68-7
U-LWR without Np	-	2.29-3	4.27-5	5.63-3	2.69-4	4.00-6	1.73-3	1.13-4	1.32-5			

Note that these isotopic concentrations refer to the irradiation time point zero. During irradiation the composition is changed such that reactivity and specific power of the target elements will vary.

The presence of the fertile Np-237 leads to a considerable increase of the specific power during an in-core time period. In addition, the initial linear power rating results already as considerably high compared to 566 W/cm permitted.

Proposals for the reduction of the power rating are discussed in ref. [2]. The geometrical and performance data for the pin are summarized in Table 3.

TABLE 3

Pellet diameter (mm)	6
Cladding outer diam. (mm)	6.7
Cladding thickness (μ)	350
Power rating (W/cm)	566
Burn-up (MWd/t)	125,000
Max. wall cladding temp. (°C)	620 in - 590 ext
Pellet density (%TD)	85
Max. neutron flux ($n/cm^2.s$)	$8.92 \cdot 10^{15}$
Cladding material	Incoloy 800

In case the linear power rating is reduced by material dilution, using alternatively as diluting material UO_2, MgO, or ZrO_2, the compositions in grams per cm of pin length of Table 4 must be chosen in order not to exceed the maximum permissible linear power rating.

TABLE 4

Diluent Material	Total oxides g/cm pin length	Fuel Composition g of oxide/cm pin length					
		U	Mg	Zr	Np	Am	Cm
UO_2	(2.675)	(0.749)			1.391	0.428	0.107
MgO	2.177		0.174		1.459	0.435	0.109
ZrO_2	2.349			0.352	1.434	0.446	0.117
	1.934				1.393	0.425	0.116
	2.10					1.659	0.441

In addition to dilution, the linear specific power rating can also be reduced by different geometric configurations of the pellets. This would represent the advantage of not introducing any material other than minor actinides to the reactor and the fuel cycle facilities. The annular pellet configuration, the thicker cladding sheath version and the pellet with largely distributed porosity, are shown in Figs. 2 to 4. A combi-

nation of thicker cladding and increased porosity may be an optimum solution.

A selective valuation of these proposals is envisaged in collaboration with the Karlsruhe Establishment of the JRC by investigating:

- temperature profiles and levels,
- oxygen redistribution,
- thermal and fission gas stress to the cladding as well as corrosion and embrittlement,
- pellet behaviour under power conditions,
- assessment of fabrication problems,
- construction of phase diagrams for actinides and diluents.

Risk Assessment

A Waste Management Code for the recycling of actinides in a nuclear reactor has been elaborated and is at present in its numerical test phase. The code permits to calculate the actinide arisings for the following cases after n cycles of a nuclear power reactor operation:

1) Actinide Input and Output for the cases called Actinide Recycling (AR) and No Actinide Recycling (NAR)

2) Actinide Waste Arisings for the Actinide Target Elements (TE), the Normal Fuel Elements (NE) for the recycling case and for the fuel elements with no actinide recycling.

The code uses as input data the actinide vectors calculated for the n-th cycle with more sophisticated burn-up codes and calculates the isotopic concentrations built up in the different components of the fuel cycle via vectors specifying the isotopic mass flow to waste repositories and or to the surroundings.

Calculated for each component of the fuel cycle are the accumulated isotopic vectors (units: g/tHM and g/GWye), the neutron output from spontaneous fission or (α-n) reactions with oxide (units: neutrons/tHM and neutrons/GWye), the actinide decay heat (units: W/tHM and W/GWye), and the inhalation and ingestion hazards (units: m^3 air/tHM and m^3 air/GWye and m^3 H_2O/tHM and m^3 H_2O/GWye respectively).

All quantities are calculated during the Nuclear Power Period as function of the number of completed actinide recycle periods and during the Decay Period as function of time. This code will be used to assess the risk sources of the two alternative fuel cycles under investigation.

COLLABORATION WITH EXTERNAL ORGANIZATIONS

- Contract with the Austrian Academy of Science, Vienna

The work regarding this contract has been successfully accomplished by providing a final report [3] and neutron cross section data for Am-243. The aim of this contract was to investigate whether sufficient nuclear input data are existing to compute reaction cross sections of americium and curium isotopes for fast neutrons by means of theoretical models for the nucleus, to develop a suitable computer program for calculating $\sigma_c(E)$, $\sigma_f(E)$, and $\sigma_{n,2n}(E)$; and to supply these cross sections to Am-243.

The final report contains the following items: description of a modified version of the computer program STAPRE for particle-induced activation cross sections in which the fission process has been included. Description of the computer program CAPTRE needed to calculate the cross sections for lower neutron energies. Indication of literature [4] regarding statistical model parameters which are needed as input parameters for calculating neutron cross sections for various minor actinide isotopes. Listing of transmission coefficients for U-238 derived by means of the IUPITOR-K/V code which can, in a first-order approximation, be applied also to calculate neutron cross sections for other isotopes.

By comparing calculated results for $\sigma_f(E)$ of U-238 with ENDF/B 4 data (see Fig. 5), the correct working of the computer program has been proved. Fig. 6 shows the results for $\sigma_f(E)$ of Am-243. A similar situation as in the case of the fission cross section of Am-241 was found, where experiments of W. Hage et al. as well as model calculations by F. M. Mann, HEDL, demonstrated that ENDF/B 4 data are much too large for energy ranges below 100 keV. Fig. 7 gives the calculated results for the capture cross section of Am-243, compared to the ENDL-76 data set of Saclay. It may be seen that the theoretical values are partly, by a factor of 5, higher than the Saclay values.

It is intended to utilize the new data for deriving effective one-group cross sections for different fast breeder neutron spectra and to apply them in ORIGEN as well as more refined FBR physics calculations.

- Contract with KFA Jülich

A one-year study contract with the "Institut für Reaktorentwicklung" of the KFA Jülich has been concluded to investigate the pebble-bed reactor as device which produces and transmutes actinides other than fuel. This reactor type uses spherical fuel elements of about 5 to 10 g of heavy metals in the form of coated particles, included in porous

graphite and two further pyrolytic sheaths.
The potential advantages of this reactor as transmutation device consist in the very high attainable burn-ups, the possibility to recycle minor actinides in separate spheres which may be distributed homogeneously over the whole core (i.e. to realize a heterogeneous recycle scheme without perturbing the macroscopic power distribution), and a possible re-feed of the spheres after discharge from the reactor without reprocessing.

In addition to the reactor physics calculations by methods developed at Jülich, information on the fabrication and reprocessing of spherical fuel elements will be provided. The results of this study will be provided in the middle of 1979.

- Contract with CNEN Casaccia

A one-year study contract is being concluded with the "Dipartimento Ricerca Tecnologica di Base Avanzata" of the CNEN Casaccia with the scope of obtaining refined reactor physics calculations for two FBRs with different hard neutron spectra. By this the preliminary calculational results for transmutation rate, specific power rating and reactivity effects of minor actinide-containing fuel elements performed at Ispra [5], shall be verified.

Also this study will be terminated in the middle of 1979.

CONCLUSIONS

It is obvious that the recycle strategies proposed for minor actinides are of preliminary nature which have to be adjusted after having results for cost and risk analysis and more detailed investigations in the fields of reactor physics, fuel element fabrication and reprocessing.

Due to administrative difficulties the formalization of the contracts for evaluating the pebble-bed reactor and the fast breeder has been delayed for half a year. However, it seems still possible to get, by means of the results still to be provided, a feedback effect on the choice of the recycle strategy.

PLANNED ACTIVITIES

During the next reporting period the following activities are envisaged:

- Generation of updated libraries for LWRs and FBRs in order to be able to reproduce experimentally determined isotope distributions in irradiated fuel samples;

- Proposal of the minor actinide-containing fuel pins for LWRs;
- Parametric fuel cycle cost evaluation;
- Preliminary results from the risk source computer program.

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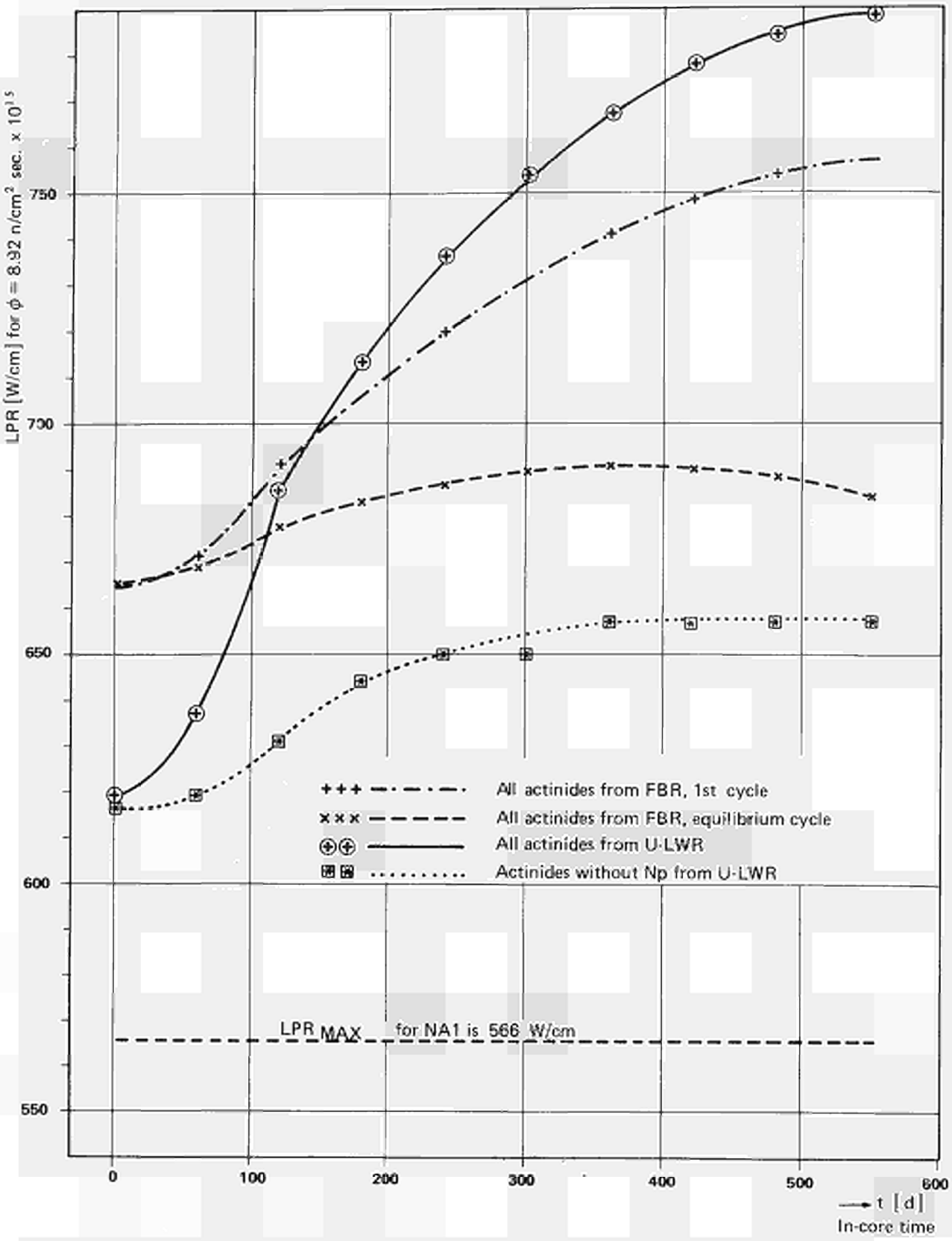


Fig. 1 — Linear power ratings for minor actinide - containing fuel pins as function of irradiation time

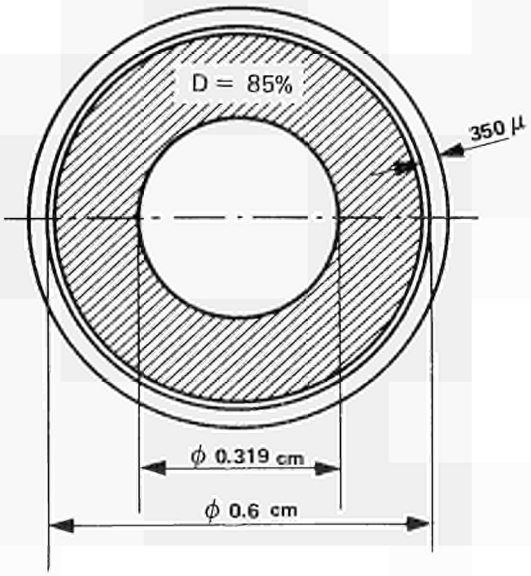


Fig. 2 – Annular pellet configuration

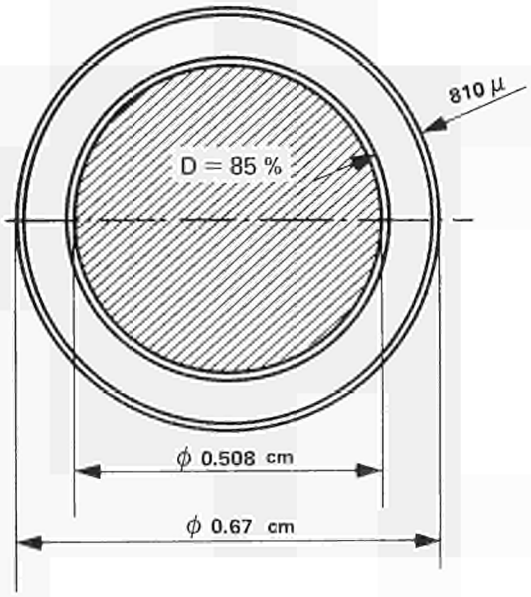


Fig. 3 – Pellet configuration with thicker cladding

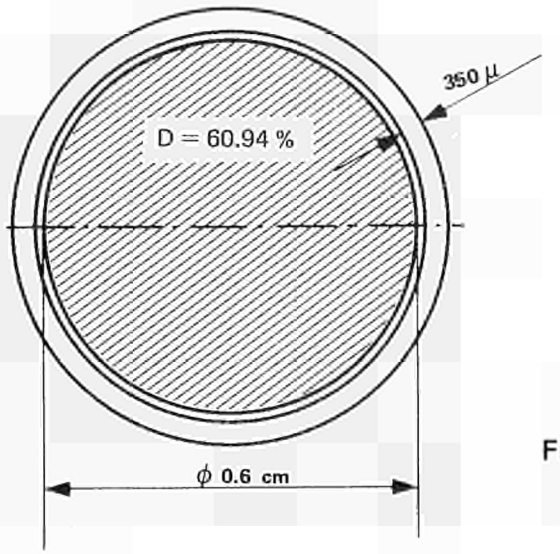


Fig. 4 – Pellet configuration with largely distributed porosity

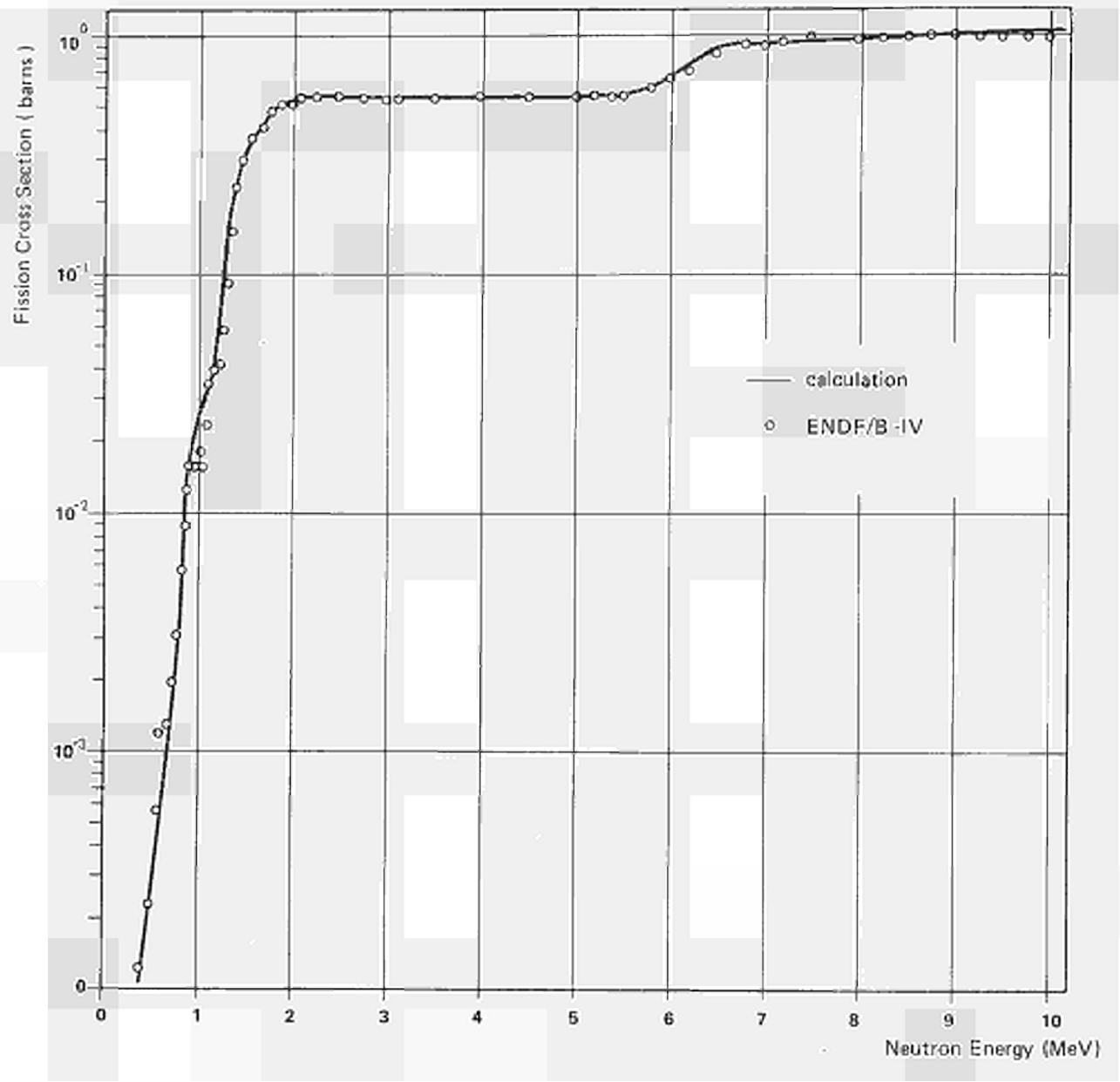


Fig. 5 – Fission cross section of U^{238}

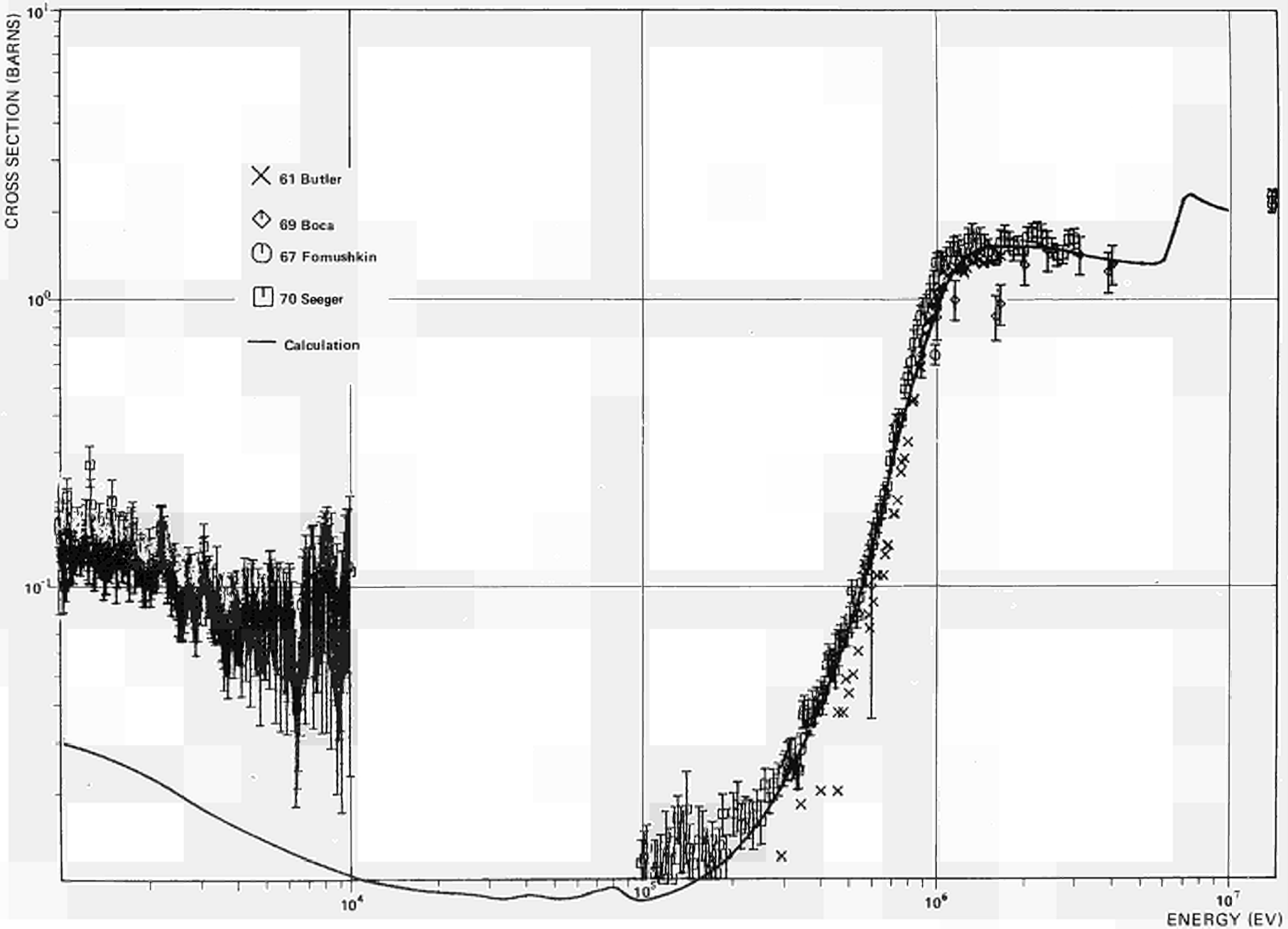


Fig. 6 — ^{243}Am (n, f) Cross Section

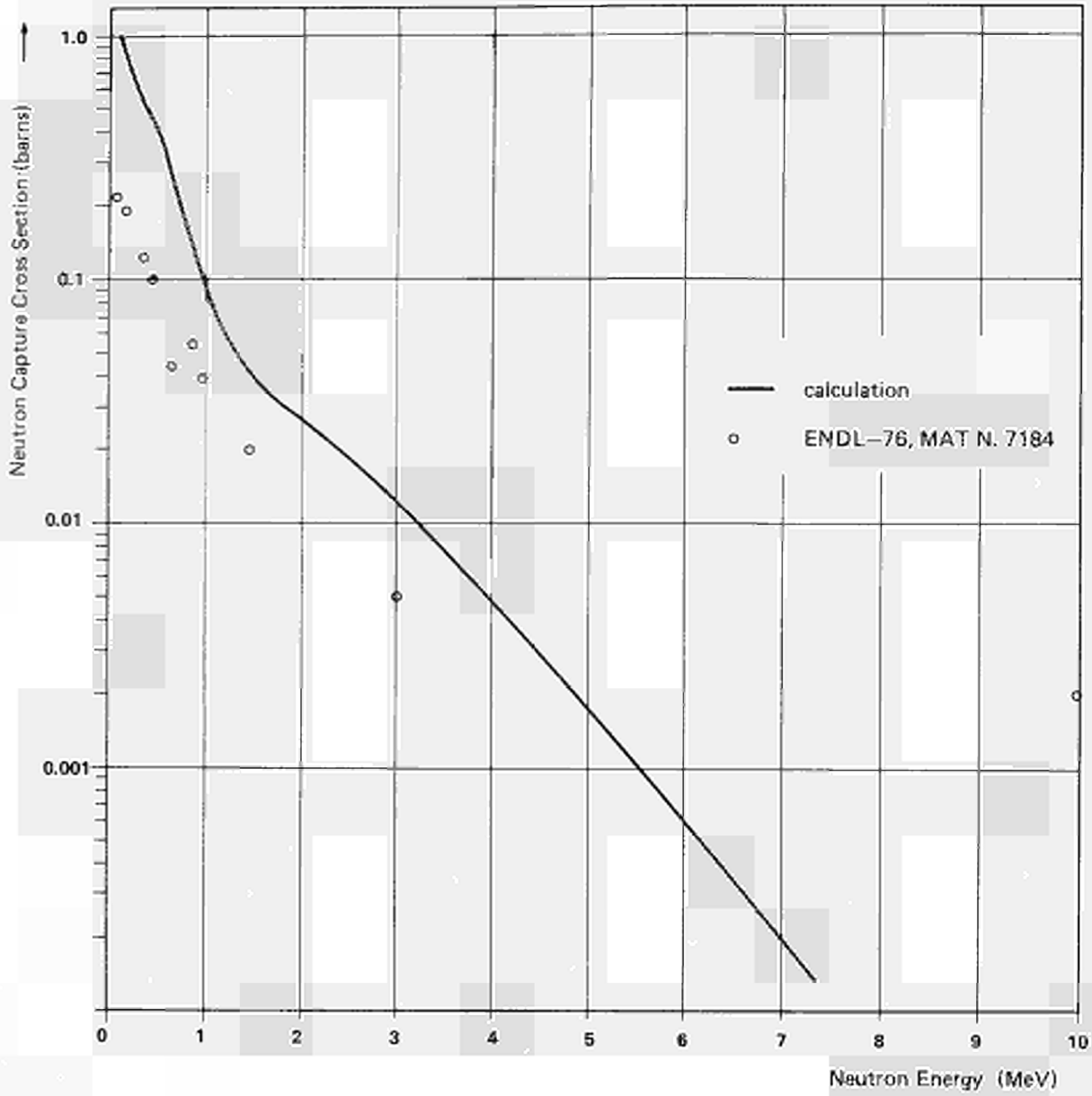


Fig. 7 – Neutron capture cross section of Am^{243}

2.2.3 Actinide Cross Section Measurements

OBJECTIVES

During December 1977 the Am-241 fission cross section has been measured applying the fission product detection method with a quasi 4π gas scintillation chamber. The experiments have been carried out in the energy range of 230 keV $< E_n <$ 1170 keV. As neutron source the 3 MeV Van de Graaff accelerator of the Institute for Applied Nuclear Physics of the KfK, Karlsruhe has been used.

The first part of the year was foreseen for the analysis of the experimental data.

RESULTS

During these experiments the double fission chamber had in its test position a mixed target consisting of (0.942 ± 0.22) mg U-235 and of (0.917 ± 0.12) mg Am-241. In the reference position a U-235 target of (4.76 ± 0.10) mg had been used. The test and reference targets irradiated by a monoenergetic pulsed neutron beam were at a distance of 48.5 cm and 54.5 cm respectively from the neutron generating target (Li(p, n)Be).

The neutron pulse duration was smaller than 0.8 ns and the neutron pulse frequency 5 MHz. The resulting fission product energy spectra were recorded for two sub-chambers, one belonging to the test, the other to the reference target as three-dimensional time-of-flight energy spectra. For the time axis 252 and for the energy 16 channels were available. The neutron energy of the Van de Graaff has been monitored with a Li-glass detector. The experiments were performed at 4 different neutron energies 230 keV, 772 keV, 976 keV and 1170 keV.

The energy-dependent time-of-flight spectra were transformed into reaction rate ratios. The Am-241 to U-235 fission cross section ratio was derived from the expression

$$\frac{\sigma_A(E_1)}{\sigma_U(E_1)} = \frac{R_1(E_1)R_2(E_2)}{R_2(E_1)R_1(E_2)}\left[\frac{\sigma_A(E_2)}{\sigma_U(E_2)} + \frac{\varepsilon_U N_U}{\varepsilon_A N_A}\right] - \frac{\varepsilon_U N_U}{\varepsilon_A N_A}$$

$$\left(\frac{\sigma_A(E_2)}{\sigma_U(E_2)}\right)_{E_2=230\ \mathrm{keV}} \ll 1$$

$R_i(E_j)$ = reaction rate of target i measured at neutron energy E_j

i = 1 test target, 2 reference target

j = 2 energy below the fission threshold of Am-241

$\sigma(E_\gamma)$ = fission cross section

A, U = index for isotope Am-241 and U-235 respectively

N_U, N_A = number of Am-241 and of U-235 atoms in the test target

$\varepsilon_U, \varepsilon_A$ = detection efficiency for fission products of Am-241 and of U-235 fission counts.

Using the specification of the test target, the above formula can be simplified to:

$$\frac{\sigma_A(E_1)}{\sigma_U(E_1)} = \frac{R_1(E_1)R_2(E_1)}{R_2(E_1)R_2(E_1)} 1.07312 - 1.05349 \qquad \text{for } E_2 = 230 \text{ keV.}$$

Two different measurement series carried out at the same set of neutron energies, agreed with each other within the derived error limits. However, the obtained fission cross section values were a factor of about 1.5 greater than the results obtained with the neutron detection method. This discrepancy is yet unexplained and several investigations are in progress.

COLLABORATION WITH EXTERNAL ORGANIZATIONS

The measurements were performed in collaboration with the staff of the Institute for Applied Nuclear Physics of the KfK, Karlsruhe, and analyzed by the JRC staff.

CONCLUSIONS

An explanation for the discrepancies between the two measurements has not yet been found and further investigations have to continue in order to clarify the obtained results.

PLANNED ACTIVITIES

It is hoped that a mass spectrometric analysis of the test target can provide the necessary information to clarify the obtained discrepancies. Until those data are available no further analysis of the experiment will be done.

The work on the calculation of the error limits of the cross sections measured with white neutron spectra is being continued.

A feasibility study is in progress for a fission cross section measurement technique with multiple neutron coincidences.
This technique should be applied for the measurement of the Am-243 fission cross section.

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 preparation) 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) sufficient radiation field decreases were 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 mechanism 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-1 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.
One of its drawbacks is that it produces a large amount of radioactive waste. The type of waste does not present special problems of conditioning, as its composition is quite similar to the normal waste of power plants (filters and ion exchange resins). The main problem lies in the amount of waste produced.

As the most common process uses highly concentrated chemicals, concentration methods are useless and the volume of the final sludges is quite important.

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 analyzed, namely:

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

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

The tests have been performed on three types of samples.
A first type of sample has been obtained oxidizing samples of AISI 304 L at 600°C in overheated steam during 30 days. A compact oxidized layer is obtained consisting of a homogeneous external part and an irregular internal part which enters into the oxidized surface.
These samples can be considered representative of a surface with a long time exposure in the reactor.

A second type of samples has been obtained exposing samples of AISI 304 L in a solution of ferrous tartrate at 300°C during 15 days. At this temperature the tartrate decomposes with simultaneous particle deposition and oxidation. The external part of the oxide layer is porous and presents a poor adherence; the internal part consists of a slightly oxidized layer.
These samples can be considered representative of a surface with a short-time exposure in the reactor.

A third type of sample has been prepared using pieces obtained from the Gundremmingen and Obrigheim power stations. They present a contamination due principally to Co-60. They will be used mainly to verify the results obtained with the other two types of samples.

A series of systematic tests has been planned operating at two temperatures (80° and 95°C) and with three different concentrations (see Table 1).

The decontamination tests have been performed in a thermostatized beaker, while gently stirring, during 4 hours for each phase. The same solution was used for the first and third phases.

TABLE 1 - Concentrations (wt%) used in the decontamination tests

	1st phase (Citrox)	2nd phase (AP)	3rd phase (Citrox)
High	20	30	20
Medium	8	12	8
Low	2.5	4	2.5

The effect of each treatment was determined by measuring the weight variation for the simulated samples and the Co-60 activity variation for the contaminated samples.

At present the tests on the simulated samples at 95°C have been completed. The results are reported in Tables 2 to 4.

Operating with the high concentration the attack was very strong. After the 1st phase the weight loss was higher than the weight of Fe_3O_4 formed during the sample treatment. After the 3rd phase the samples showed a corroded irregular surface.

Operating with the medium concentration the results are very satisfactory for the samples simulating a long-time exposure but the attack seems too strong for the samples simulating a short-time exposure.

Operating with the low concentration the results are satisfactory for the samples simulating a long-time exposure. However, the effect of the oxidizing phase seems to be negligible.

It has to be noted that at 95°C the influence of the 1st phase is predominant. Some tests at 80°C have shown, on the contrary, a lower influence of the 1st phase.

If the tests on the contaminated samples confirm these results, experiments will be carried out using only the 1st phase at higher temperature.

TABLE 2 - Decontamination tests with high concentration solutions
(weight in grams)

		Weight of O_2 absorbed	Weight of Fe_3O_4 on the sample	Weight variation 1st phase	Weight variation 2nd phase	Weight variation 3rd phase	Weight variation Total
Long-time exposure samples	Mean values	0.01560	0.05643	-0.06961	+0.00109	-0.09532	-0.16384
Short-time exposure samples	Mean values	0.00012	0.00186	-0.00630	-0.00024	-0.03689	-0.04343

TABLE 3 - Decontamination tests with medium concentration solutions
(weight in grams)

Long-time exposure samples	Mean values	0.01572	0.05686	-0.04124	+0.00148	-0.01438	-0.05414
Short-time exposure samples	Mean values	0.00012	0.00187	-0.00380	+0.00037	-0.00063	-0.00406

TABLE 4 - Decontamination tests with low concentration solutions
(weight in grams)

Long-time exposure samples	Mean values	0.01020	0.03690	-0.02074	+0.00007	-0.00277	-0.02344
Short-time exposure samples	Mean values	0.00012	0.00174	-0.00145	+0.00004	-0.00013	-0.00154

Physico-Chemical Structure of Oxide Layers and Mechanisms of the Decontamination Processes

In order to improve hard decontamination processes and to make soft decontamination sufficiently effective, a better knowledge is required in the following fields:

a) Structure and chemical composition of oxide films formed in high temperature water on austenitic alloys,

b) local distribution of active isotopes (in most cases active Co-isotopes) in the oxide films,

c) mechanisms of action of the decontamination processes.

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

Various authors suggest that the morphology of these oxide films is similar to those formed on carbon steel, i.e. oxide films consist of two layers. The inner oxide layer, formed by oxygen migration into the metal and metal ion migration outwards, is a porous oxide film with the dimensions and structure of the original metal.
The outer oxide layer is formed by precipitation of oxide crystals, owing to supersaturation of the aqueous phase produced by the metal ions which migrate outwards from the metal through the oxide. In the case of carbon steel both oxide layers consist of magnetite.

In the case of austenitic alloys some authors suggest that spinel type oxides such as $NiO.Fe_2O_3$ or $FeO.Cr_2O_3$ are formed. The very small quantities of oxides formed on austenitic alloys in high temperature water did not permit, in the past, to demonstrate neither the two-layer structure nor the real chemical composition of the oxides. However, this should be possible with modern tools such as scanning electron microscopy (SEM) and surface analysis by Auger electron spectroscopy and secondary ion mass spectrometry (AES and SIMS).

The research has been oriented along the following lines:

- preparation of suitably oxidized stainless steel specimens in a small high-temperature water test loop (set up during the previous research programme),
- morphological analysis of the specimens by means of SEM (to be carried out in the JRC laboratories),
- analysis of chemical composition profiles of the oxide on the specimens by means of AES-SIMS (to be carried out under research contract).

Later on these studies shall be completed by SEM investigations of contaminated surface specimens in the Medium Activity Laboratory of the JRC. Contaminated reactor parts from nuclear power plants of the Community, are already available for the preparation of suitable specimens.

b) Local Distribution of Cobalt in Oxide Films on Austenitic Alloys

Some authors suggest that radioactice cobalt ions generated in the reactor water are incorporated in the inner oxide layer by migration into the pores and precipitation on the pore walls and in the outer oxide layer by co-precipitation together with the respective spinel type oxides.

Chemical composition depth profiles made with SIMS could be sufficiently sensitive to reveal the distribution of cobalt within the oxide layers.

The results obtained by the contractor with SIMS will be evaluated also in this respect.

c) Mechanisms of Action of the Decontamination Processes

- Hard Decontamination

For primary circuit parts in austenitic alloys the decontamination procedures consist always of strong oxidizing and alkaline treatment as the first step and of a complexing and acid treatment as the second step. The hypothesis was put forward that with the first step the chromium spinels, which are very resistant to the chemical attack, are destroyed by oxidation of chromium to chromium-VI and solubilization as chromate. The remaining porous iron oxide structure can be dissolved more easily afterwards by the complexing acid treatment.

The application of the above mentioned investigation techniques (SEM, SIMS, AES) to oxidized specimens which had undergone the first step treatment, should give an answer on the validity of the mentioned hypothesis.

- Soft Decontamination

Very little knowledge exists on the effect of thermal or redox cycling on the oxidized austenitic alloy surfaces. The small test loop which is available from previous work, is particularly useful for the investigation of corrosion product release during cycle operations. If the amounts of corrosion products released during cycling are confronted with the changes in quantity and composition of the ad-

herent oxide, as they can be obtained by the mentioned surface investigations (SEM, SIMS, AES), useful information about the mechanism and the efficiency of these cycling procedures should be obtained.

The main results obtained in the different fields are the following:

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

A research contract with Dornier System, operating a specialized laboratory, has been concluded in order to obtain the confirmation of the applicability of AES and SIMS techniques for our purposes.

Certain problems concerning specimen preparation (transformation of tube sections into flat surfaces without surface contamination by oil or grease) were resolved in the JRC-laboratories, and four specimens were submitted to investigation at Dornier System.
The specimens were obtained with the following experimental procedures:

- 1) reference specimen made of non-oxidized stainless steel tube
- 2) surface oxidized in oxygenized water at 300°C for 84 hours,
- 3) as no. 2, but oxidized during 500 hours,
- 4) surface oxidized in oxygen-free, but hydrogenized water during 1,800 hours.

Some preliminary results obtained by Dornier on specimen no. 4 permit to draw the following conclusions:

- AES and SIMS techniques can be successfully applied for the analysis of our specimens. For the quantitative evaluation, corrections for surface roughness effects will be necessary. The correction will be determined by comparison of the results of rough and smooth non-oxidized metal surfaces, and by microscopic studies of sectioned specimens before and after application of the SIMS ion bombardment.
  This work has to be carried out at the JRC-Ispra.
- The AES depth profile of elementary distribution does not reveal a two-layer morphology as expected from literature information.
- The AES depth profile shows slight unexpected gradients of the ratios between the elements iron, chromium, nickel and oxygen.
- The SIMS analysis of the surface near the oxide-water interface shows higher oxygen contents than expected assuming that the layer

is formed by spinel type oxides ($MeO.Me_2O_3$).

The final conclusions of the investigation made by Dornier is expected in July 1978.

In addition to the AES-SIMS investigations, SEM investigations of the surfaces and surface sections will be necessary. Especially the preparation of metallographic section specimens for SEM has required some optimization work. The following standard procedure was developed during the last period:

- cutting of sections from the loop tubings with contamination by oil and grease;
- covering of oxidized specimen surfaces with a stainless steel layer of about 3 micron thickness, by means of a sputtering (evaporation by ion bombardment) technique;
- embedding of the specimen in Wood metal and application of standard metallographic polishing techniques.

The procedure is now definitely established and the first series of specimens, corresponding to those investigated by means of the AES and SIMS techniques, is in preparation.

c) Mechanisms of Action of the Decontamination Procedures

- Hard Decontamination

  The studies require suitable specimens. Some specimens will be obtained from the small release test loop during July, when 1,800 hours of oxidation at 300°C in water with about 0.5 ppm dissolved oxygen, will be reached.

  Another specimen, submitted to more than 5,000 hours of oxidation in a thermal exchange test loop of the JRC, will be available in September. Both specimens are suitable for surface investigations before and after treatment with decontamination solutions.

- Decontamination by Thermal and Redox Cycling

  In the reporting period the loop was run to prepare a suitable oxide layer on the test section, for cycling experiments. These runs will be terminated before the end of July.

  Three thermal cycles resulted from starting and shut-down of the loop for maintenance work. The release of solid and dissolved corrosion products during the runs were checked by chemical analysis. The evaluation of the results will be completed as soon as the micrographs of the surface sections allow a comparison between the quantities of adherent and released oxide.

All tests were carried out with feed water with an electric conductivity of 0.15 micro-ohms and a content of dissolved oxygen of about 0.4 ppm. The flow rate in the test section was always 8 m/s. Every test is subdivided into the following phases:

1) initial cold run of about 48 hours at 60°C,
2) heat-up from 60° to 300°C within 4 hours,
3) warm run during up to 300 hours at 300°C,
4) cool-down from 300° to 60°C within 4 hours,
5) final cold run of about 48 hours at 60°C.

Filter and ion exchanger samples were always taken at the same time interval during the three tests.

Test no. 1 was made with a new stainless steel (AISI 304) test section, suitably etched before the start of the test. During the tests nos. 1, 2 and 3 the total oxidation time at 300°C was 119 hours, 223 hours and 666 hours, respectively.

The following general conclusions can be drawn from the results:

1. The main quantities of metal were always released during the heat-up phase and the very first hours at 300°C.
2. The release peak during the cool-down phase always brings about minor quantities.
3. The contributions of the individual metals to the overall release rates change from the 1st to the 3rd run: the contribution decreases from 38.8% to 4.8% for chromium, increases from 34.4% to 77.6% for iron, and decreases from 25.8% to 17.5% for nickel.

Evaluation of the Decommissioning of the Ispra-I Reactor

The Ispra-I reactor is a 5 MW research reactor which had been in operation for 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.

Following the IAEA definition of the three successive stages of decommissioning, it can be said that the Ispra I reactor, today, is in its first stage.

The second stage criteria are mainly imposed by economical considerations (minimization of surveillance and maintenance), and are normally different for different reactors.

The third stage (dismantling) seems more appropriate for an operation aiming at obtaining results of general interest.

A general mapping of the activity levels in the various parts of the reactor is being performed. The external circuits have been examined. Most of them show a contamination of the order of 10^{-3} $\mu Ci/cm^2$, mainly due to Co-60. In some parts of the reactor activities have been measured of up to 30 $\mu Ci/cm^2$.

During the following period samples will be extracted and measured of graphite from the reflector and of concrete from the biological shielding. The activity of the thermal shielding will also be evaluated.

A first screening of the possible subjects of research has been made. The following topics seem of particular interest:

- methods of total decontamination of the external circuits,
- methods of conditioning of activated concrete,
- variation of C-14 pick-up by vegetation in a zone where activated graphite is burned,
- general evaluation of the total amount of waste generated, following different strategies.

It is clear that a solution for the disposal of the generated wastes has to be found before any dismantling operation is started.

COLLABORATION WITH EXTERNAL ORGANIZATIONS

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

CONCLUSIONS

The first results of the systematic tests on chemical decontamination indicate that the concentration of chemicals can be reduced if the treatment temperature is increased.

The first results on the composition of crud and surface deposits show a substantial difference when the circuit is operated in oxidizing or reducing conditions. There is hope to obtain good results by a redox cycling or at least to reduce in this way the resistance of the deposited

layer.

The mapping of the residual activity in the Ispra-I reactor is continuing. The values of the contamination on the external circuit are encouraging.

PLANNED ACTIVITIES

Systematic Study on Chemical Decontamination

Verification of the results with radioactive samples. Completion and verification of the low temperature tests. Possible extension to higher temperatures.

Effect of Thermal and Redox Cycling on Oxide Film Morphology

Tests of redox cycling and analysis of the crud detached and of the resulting surfaces. Evaluation of the implications to the waste management.

Preliminary Evaluation of the Decommissioning of the Ispra-I Reactor

Completion of the mapping of the residual activity. In-depth analysis of the possible subjects of research. Evaluation of the type of disposal requested.

REFERENCES

/1/ ROOFTHOOFT, R., "La décontamination dans les centrales nucléaires refroidies à l'eau légère - Résultats d'une enquête dans les centrales Européennes", EUR 5898f (1977).

Conclusions

3. CONCLUSIONS

In the first 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 methodology developed at Ispra is being utilized to quantify the probabilistic value of the geological barrier in the Boom (Belgium) clay formation.

During the reporting period the model used to calculate pathways and dose rates to man has been refined through more careful assessment of waste inventories related to different fuel cycle options and more detailed leaching models for the different types of conditioned wastes.

Progress has been made in the studies on the stability of conditioned waste and the interaction of actinides with environment, which are directed to provide input data for the waste hazard models.
Experiments have been started to determine leaching rates of glasses using water compositions related to specific geological repositories.
For the study of the interaction of actinides with geological media and groundwaters following their eventual leaching from vitrified waste, glasses spiked with plutonium and americium have been produced and methods for the determination of the physico-chemical forms of the actinides in the leached solutions, are under study.
Migration experiments with leached solutions through sandy soils and sediments under various simulated environmental conditions have been started.

In order to provide input data concerning the interaction of actinides with the biosphere contacts have been taken with various European laboratories operating in the framework of the indirect programme Radiation Protection. In fact, the development of a harmonized series of experimental projects in the framework of the Radiation Protection Programme could be the most effective solution to obtain the data needed for the risk assessment programme of the JRC.

In the field of actinides monitoring the work for the preparation of a guide on the monitoring of plutonium contaminated waste streams is progressing: at present the chapter dealing with the active neutron assay is in preparation.
Collaborations have been established with scientific institutes and nuclear

plant operators. In particular an integral experiment is worth mentioning, to be carried out in the Dounreay reprocessing facility, on the monitoring of plutonium contaminated waste streams. This experiment aims at the demonstration of the monitoring concept developed at Ispra. It is scheduled for a duration of two years, beginning July 1978.
During the first semester of 1978 the staff allocated to this activity was reinforced.

The planning of future activities for the project Evaluation of the Long-Term Hazard of Radioactive Waste Disposal is shown in Table 1.

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, experiments have been carried out on simulated HAW solutions in order to clarify some particular points. During the reporting period it was also possible to pass to the second phase of the programme which includes the test of the three flow-sheets on fully active HAW solutions.

Due to the delay in the recruitment of a chemical engineer, the critical evaluation of the three flow-sheets in order to estimate the engineering implications will be initiated in the second semester of 1978.

The staff allocated to the assessment studies on nuclear transmutation of actinides has been reinforced by a reduction of the effort on the actinides cross section measurements.

Due to the complexity of the study which includes reactor physics calculations, fuel element design, evaluations of cost and risk and of implications on the nuclear fuel cycle, we plan to obtain contributions from external organizations by means of contracts on specific topics.

The planning of future activities for the project Chemical Separation and Nuclear Transmutation of Actinides is shown in Table 2.

Decontamination of Reactor Components

After a period of time devoted to a literature review and to preliminary experiments, the activity has been started on some lines of interest for

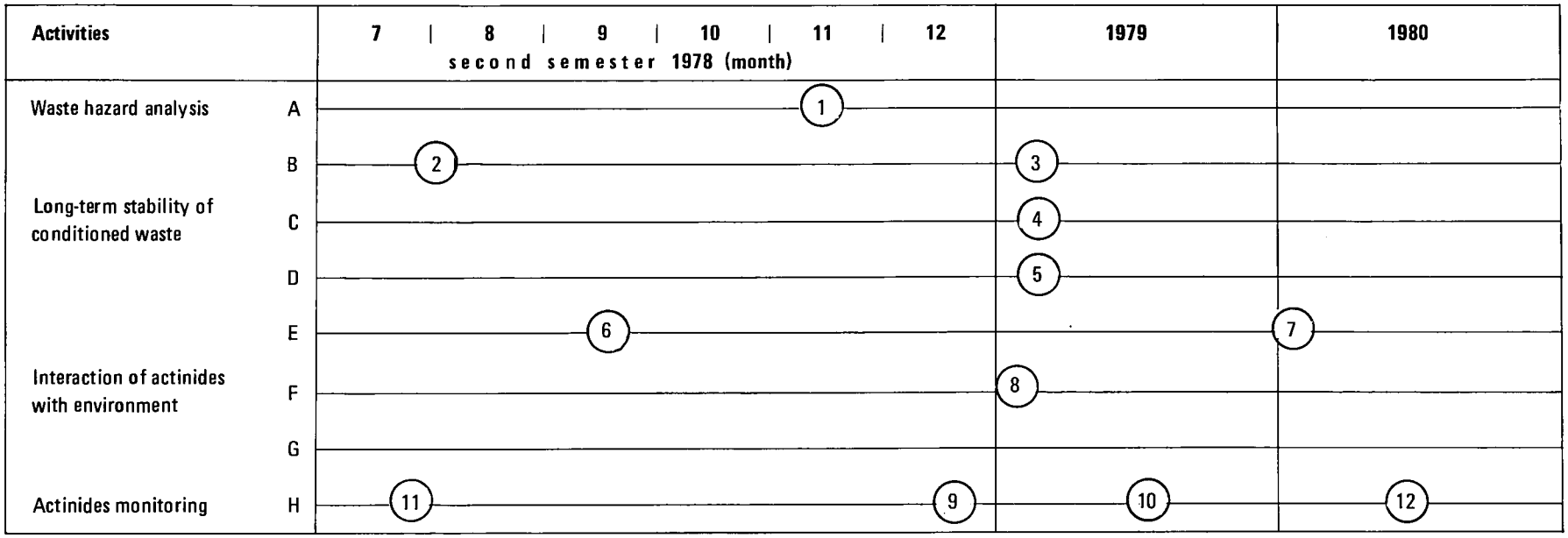
decontamination processes in power plant operations.
The studies on the physico-chemical structure of oxide layers and on the chemical decontamination take advantage of the use of modern tools of investigation, such as scanning electron microscopy (SEM), Auger electron spectroscopy (AES) and secondary ion mass spectrometry (SIMS).

During the reporting period an evaluation of the possibility and interest to execute the decommissioning of the Ispra-1 reactor as a test case, has been started.

The planning of future activities is shown in Table 3.

For further information, please contact the Programme Manager.

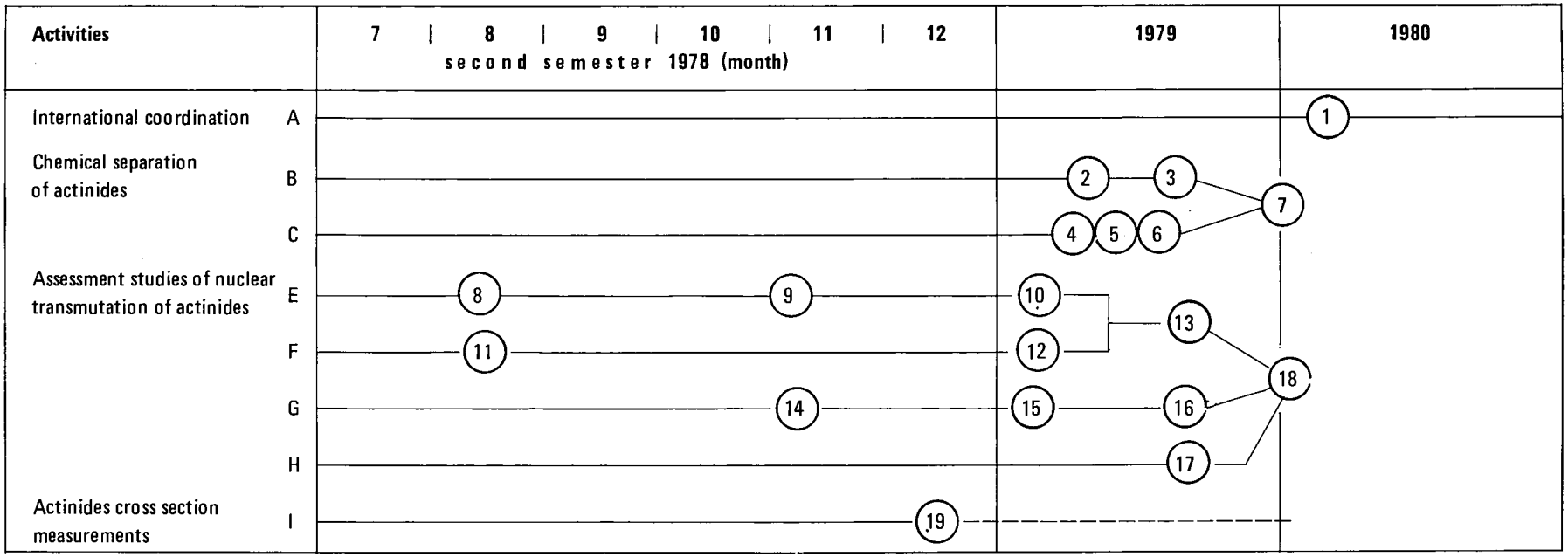
TABLE 1 : PLANNED ACTIVITIES AND IMPORTANT MILESTONES FOR THE PROJECT 1 : EVALUATION OF THE LONG-TERM HAZARD OF RADIOACTIVE WASTE DISPOSAL



A Modelling of failure of geological disposal
B Actinides distribution in environment following failure of geological barrier
C Leaching tests on vetrified waste
D Leaching tests on bituminized waste
E Experiments of radiation damage in glasses
F Interaction with abiotic environment
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

1 Completion of the model for clay formation (site specific) and start-up of the preparation of models for other geological formations
2 Model revision (more detailed waste inventory, more detailed environment model)
3 Model application to different fuel cycle strategies
4 Completion of the experiments with water in conditions of geological disposal
5 Completion of the experiments on bituminized waste and possible start-up of experiments on other matrices
6 Decision point: completion of the experiments on the radiation damage simulation by fission fragments or start-up of a new irradiation test
7 Completion of the experiments on the validity of accelerated tests
8 Conclusion of initial column (soil and sediments) experiments with leached actinides and development of further experimental programme
9 Completion of the chapter V of the guide "Application of active neutron assay"
10 Revision of the Guide
11 & 12 Start-up and completion of the integral experiment in a reprocessing plant

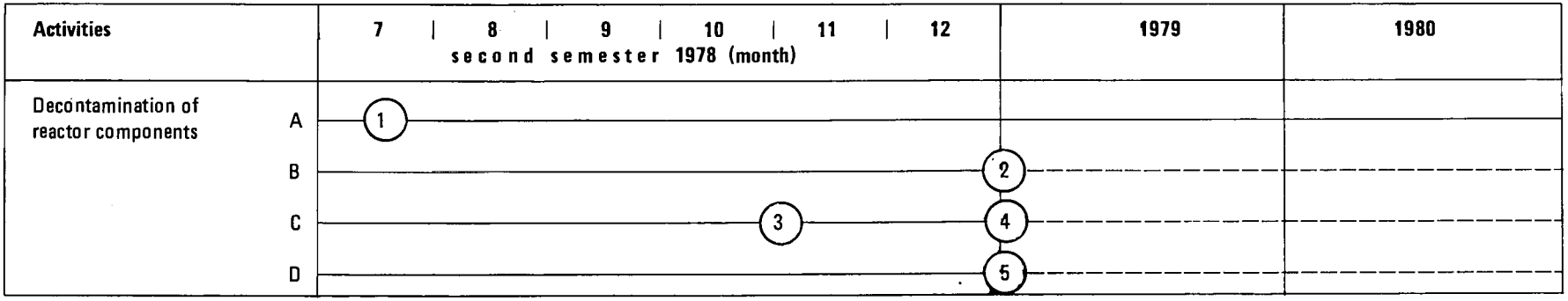
TABLE 2 : PLANNED ACTIVITIES AND IMPORTANT MILESTONES FOR THE PROJECT 2 : CHEMICAL SEPARATION AND NUCLEAR TRANSMUTATION OF ACTINIDES



A International coordination in the framework of the activity of the OECD Nuclear Energy Agency
B OXAL process
C Solvent extraction
E Reactor physics calculations
F Collection of chemical and physical data for fuel element design
G Cost and risk analysis
H Study of the implications of actinide recycling on the fuel cycle
I Differential cross section measurements

1 Organization of an international meeting
2 Completion of the experiments on OXAL at fully active scale
3 Completion of preliminary engineering evaluations
4 Completion of the fully active scale batch experiments with HDEHP and TBP
5 Start-up of the countercurrent experiments with HDEHP
6 Preliminary engineering evaluations
7 Report on feasibility of actinides separation. Tentative cost evaluation. Proposal for further actions
8 Establishment of reactor physics calculation methods for FBR and THTR
9 Generation of nuclear data set
10 Completion of reactor physics calculations
11 Proposal of preliminary fuel element designs containing actinides other than fuel
12 Definitive proposal of fuel element
13 Elaboration of an overall recycle strategy
14 Establishment of cost calculation procedure
15 Establishment of the methodology of risk assessment
16 Results of cost and risk assessment
17 Changes in nuclear plants and transportation
18 Preparation of a report on the feasibility of the actinides transmutation
19 Completion of the measurements on Am-241

TABLE 3 : PLANNED ACTIVITIES AND IMPORTANT MILESTONES FOR THE PROJECT 3 : DECONTAMINATION OF REACTOR COMPONENTS



A Systematic study on chemical decontamination
B Study on the physico-chemical structure of oxide layers
C Partial decontamination using thermal redox cycling
D Preliminary evaluation of the decommissioning of the Ispra 1 reactor

1 Comparative tests on contaminated samples
2 Decision point
3 End of the experiments on the thermal cycling
4 Decision point
5 Redaction of an operational proposal - Decision point

4. JRC PUBLICATIONS

Published or Presented

1) BERTOZZI, G., CARETTA, A., SCHNEIDER, H., "On the Risk of Radionuclide Leaching by Groundwater and Biosphere Transport with Input Data Uncertainties Described by Probability Distributions", paper presented at the Deutsches Atomforum, Hanover, April 4-7, 1978

2) BERTOZZI, G., CARETTA, A., SCHNEIDER, H., "Probabilistic Risk Analysis of Radioactivity Release and Transport from Geologic Disposal of Radioactive Wastes", paper presented at the Meeting on "Probabilistic Analysis of Nuclear Reactor Safety", organized by ANS, Los Angeles, May 8-10, 1978

3) LANZA, F., PARNISARI, E., "Evaluation of Long-Term Leaching of Borosilicate Glasses", EUR 5947e (1977)

4) ZAMORANI, E., "Surface Dose Rate by Actinides Mixed in Uranium-Plutonium Fuel Elements", EUR 5917 e (1977)

5) GIRARDI, F., "Removal of Long-Lived Alpha-Emitters from Radioactive Waste", paper presented at the Canadian Nuclear Association 18th Annual International Conference, Ottawa, June 11-14, 1978

6) ANTONINI, M., MANARA, A., LENSI, P., "Ion Irradiation and Stored Energy in Vitreous SiO_2", paper presented at the International Topical Conference on the Physics of SiO_2 and its Interfaces, York Town, March, 1978

7) ROOFTHOOFT, R., "La décontamination dans les centrales nucléaires refroidies à l'eau légère - Résultats d'une enquête dans les centrales Européennes", EUR 5898f (1977).

Submitted for Publication or Presentation

1) BIRKHOFF, G., BONDAR, L., "Monitoring of Plutonium Contaminated Solid Waste Streams - A Guide for Design and Analysis of Monitoring Systems"; Chapter IV: "Application of Passive Neutron Assay", to be published as EUR report.

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