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Management of Nuclear Materials and Radioactive Waste

PROGRAMME PROGRESS REPORT

July - December 1977

ABSTRACT

This document is the progress report of the Programme Management of Nuclear Materials and Radioactive Waste of the Joint Research Centre for the period July - December 1977. 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 Boom clay formation (Belgium) has been chosen as the test case for the development of a site specific model of the geological barrier. The Fault-Tree Analysis is being used to quantity the probabilistic value of the barrier.
The results of leaching tests on glasses seem to confirm that the release of the elements is obtained not only by a solution process, but also by a colloidal phenomenon.
In the field of the actinides monitoring a guide is in preparation; chapter IV of this guide on Passive Neutron Assay has been completed.

Project 2 : Chemical Separation and Nuclear Transmutation of Actinides
In the field of actinides separation by solvent extraction the batch tests on simulated high activity waste solutions using HDEHP and TBP have been completed. The prepration of waste to be used for fully active experiments has been started by dissolution and processing high burn-up fuels.
Gamma- and neutron emission by actinides have been calculated in order to assess the problems generated by the presence of the actinides in the nuclear fuel cycle.

Project 3 : Decontamination of Reactor Components
This project initiated at the beginning of 1977. A review paper on the state of the art in the field of decontamination of LWR components is almost completed.
The activity has been oriented towards the decontamination by chemical methods and by thermal and redox cycling.

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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 1977 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:

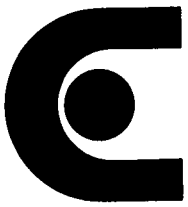
- Evaluation of Long-Term Hazard of Radioactive Waste Disposal, comprising essentially the identification and the evaluation of the long-term hazard of the permanent storage of radioactive waste in geological formations. This type of storage is considered at present to be the most appropriate to solve 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 also an Indirect Action in this field. In this Indirect Action, which is conducted by means of contracts with national laboratories, various aspects of waste conditioning technologies are studied and a large coordinated action for the study of waste disposal in various types of geological formations was established.

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



Projects

2. PROJECTS

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

The long-term hazard of radioactive waste disposal in geological formations, which is essentially due to the presence of actinides, is studied by the barrier approach which is 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 abiotic environment,
- 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. In particular at the beginning of 1977 a study has been started to verify the validity of the accelerated irradiation tests which are carried out in various laboratories to simulate long-term alpha-damage.

Experimental studies on actinides distribution in the environment relate to the chemico-physical interactions between leached out actinides and deep soil. 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 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 second semester of 1977 the planned activities were:

A. To start the application of probabilistic analysis of the risk to specific disposal sites (Belgian clay formations),

B. to initiate a critical revision of the model used to calculate pathways and dose rates to man,

C. to develop a preliminary assessment of the impact of radionuclide release into an aquatic ecosystem.

RESULTS

A. The Boom clay formation has been chosen as the test case for the development of a site specific model of the geological barrier. Fault Tree Analysis is being used to quantify the probabilistic value of the barrier.

   The necessary geological information has been gathered, most of it having been supplied by C.E.N.-Mol. Three different receptors have been identified, namely groundwater, land surface and atmosphere, corresponding to three different top events of the Fault Trees.

B. A revision of the actinide release and distribution model has been initiated, through a more careful examination of some points. The waste inventory has been re-evaluated by taking into account three different options for the LWR fuel cycle, namely:

   - the throw-away mode,
   - the uranium-only recycle,
   - the uranium and plutonium recycle.

   A more sophisticated terrestrial model has also been set up, while leaching models for various waste conditioning forms are now under investigation.

C. A study has been undertaken to develop a methodology for the assessment of the distribution of artificial radioactivity in a coastal marine ecosystem.

The various environmental compartments have been identified and the processes occurring within them have been outlined. The preliminary results have been presented in a technical note /1/.

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 Action 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 first 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 in Brussels on 21-22 November 1977.

The aim of the group is essentially to facilitate a detailed and timely exchange of information among experts and to favour the establishment of common criteria for risk evaluation. The JRC assures a permanent secretariat for the group.

CONCLUSIONS

The methodologies previously developed for a generic geological formation, have proved to be adaptable to real disposal sites without major difficulties. The preliminary assessment of the riskinvolved for future generations shows it to be very small, both in terms of event probability and of resulting dose rates to man, should the release occur; inhalation appears as the critical pathway.

PLANNED ACTIVITIES

The activities described under points A, B and C will be continued; more sophisticated data handling techniques will be set up, which will permit to treat information in the form of probability histograms, such that the effect of the uncertainty in data be apparent in the results.

REFERENCES

/1/ MURRAY, C. N., AVOGADRO, A.,
"Preliminary Report on Modeling the Transfer of Radioactivity through a Marine Ecosystem". Technical Note

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, which is necessary for waste hazard evaluation.

The planned activities for the second half of 1977 were, as for the first semester, the following:

- Post-irradiation examination of glasses irradiated in the Petten Reactor,
- Continuation of the glass leaching tests,
- Extension of the stability tests to bituminized waste,
- Checking of the validity of damage simulation by means of accelerated tests. This is a new activity started in 1977, which requires an initial period of evaluation of possible irradiation means and of physical parameters to be measured.

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 relative fission densities have been evaluated by the analysis of two fission products, respectively Cs-137 and Ce-144. The results obtained are in agreement with the nuclear code calculations with exception of one sample. From the point of view of the fission density distribution the irradiation can be considered satisfactory.

The measuring of the leaching coefficients has been completed. The leaching coefficient was measured at 100°C by the Soxhlet test method. The tests lasted normally two weeks and the samples were weighed every week. No relation was observed between the fission density and the leaching coefficient. Besides the values of the irradiated samples are practically the same as those of the non-irradiated glasses.

The density measurements have shown an increase of density for fission density of the order of 10^{16} fissions/cm^3, followed by a rapid decrease.

A similar behaviour was observed by Peymal [1] on borosilicate glasses but the increase was effective for lower equivalent dose. Due to delay in the delivery of a DTA (Differential Thermal Analysis) apparatus of high sensitivity, the measurements of stored energy were not possible. They will be carried out at the beginning of 1978.

A preliminary report was presented at the First HFR Users Symposium, held in Petten, October 1977.

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.

A test at 80°C was terminated after 9700 h of leaching. A test at 50°C will be terminated in 3 months.

Preliminary results of surface analysis show that the composition of the surface layer changes largely during the leaching test. Particularly after a certain time some oscillations of composition appear. It seems confirmed that the release of the elements is obtained not only by a solution process but also by a colloidal release.

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.

Checking of the Validity of the Damage Simulation on Glasses by Means of Accelerated Tests

Preliminary irradiation tests have been performed at Ispra and Harwell. Amorphous silica samples, 100 µm thick, were irradiated by 1 MeV alpha particles at a flux of $3.5 \cdot 10^{13}$ ∝/cm^2.sec, until a total integrated flux of 10^{18} ∝/cm^2 was attained. During the irradiation, the sample temperature was kept below 25°C.

Other samples, having the same dimensions, were irradiated by Ni^{+6} heavy ions, using the Variable Energy Cyclotron available at AERE, Harwell. The irradiations were performed at an energy of 46.5 MeV, at two different fluxes and integrated doses of 0.5 and $1 \cdot 10^{12}$ ions/cm^2.sec

and 2 and $4 \cdot 10^{15}$ ions/cm^2. sec respectively. When a nominal value of 25 eV is assumed for the displacement energy of an atom in SiO_2, the mentioned doses are expected to produce 2.5 dpa, after alpha-particle irradiations and 0.25 and 0.5 dpa after heavy ion irradiations. The range of 46.5 MeV Ni^+ ions in silica can be evaluated to be of the order of 10 µm. This value was obtained by means of a computer simulation of the heavy ion irradiation experiment.

The energy stored by the irradiated samples has been measured by thermal analysis. DSC 910 and DSC 990 Dupont calorimeters have been used. The obtained results are shown in Fig. 2 and are compared with data reported by Roberts et al. [2], after neutron irradiation of $3.4 \cdot 10^{20}$ n/cm^2, equivalent to about 1 dpa. A quantitative analysis of the data is being performed for a comparison with analogous data originated by irradiation with irradiation sources inserted within the samples.

COLLABORATION WITH EXTERNAL ORGANIZATIONS

For the checking of the validity of damage simulation on glasses, collaborations have been established with the Stazione Sperimentale del Vetro, Murano (Italy) and with AERE, Harwell (UK).

CONCLUSIONS

The leaching tests carried out in the reporting period seem to confirm that in the evaluation of the waste hazard a mechanism of homogeneous dissolution of the glasses can be assumed.

Concerning the checking of the validity of the damage simulation, the first results, i.e. thermal analysis spectra of alpha- and heavy ions irradiated samples, are very encouraging because of the stored energy measured being high enough to offer the possibility of a comparison with the results obtained in other laboratories.

PLANNED ACTIVITIES

In the first semester of 1978 the following activities are planned:

- Radiation Effects on Glasses

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

- Leaching Tests on Glasses

  Completing the tests at 50°C and the analysis of the surfaces. Begin-

ning of tests in water conditioned by percolation through a column of sand and clay.

- Leaching Tests on Bitumen

Beginning of systematic tests on leaching of bitumen loaded with salts of different solubilities.

- Checking of the Validity of the Damage Simulation on Glasses by Means of Accelerated Tests

Further irradiations are planned in the near future, at various dose rates and integrated doses, including electrons accelerated at 1 MeV or more, in order to allow the observation of the effects of various types of particles on the atomic mechanisms of damage in amorphous silica. Samples of more complex compositions, i.e. borosilicates, will also be examined.

Theoretical studies of formation, recombination and agglomeration of defects in these materials will also be pursued, in collaboration with the Theoretical Physics Division of AERE, Harwell.

REFERENCES

/1/ LE CLERC, P., "Action des rayonnements sur les verres", B.I.S.T., 7, Nov. 1965

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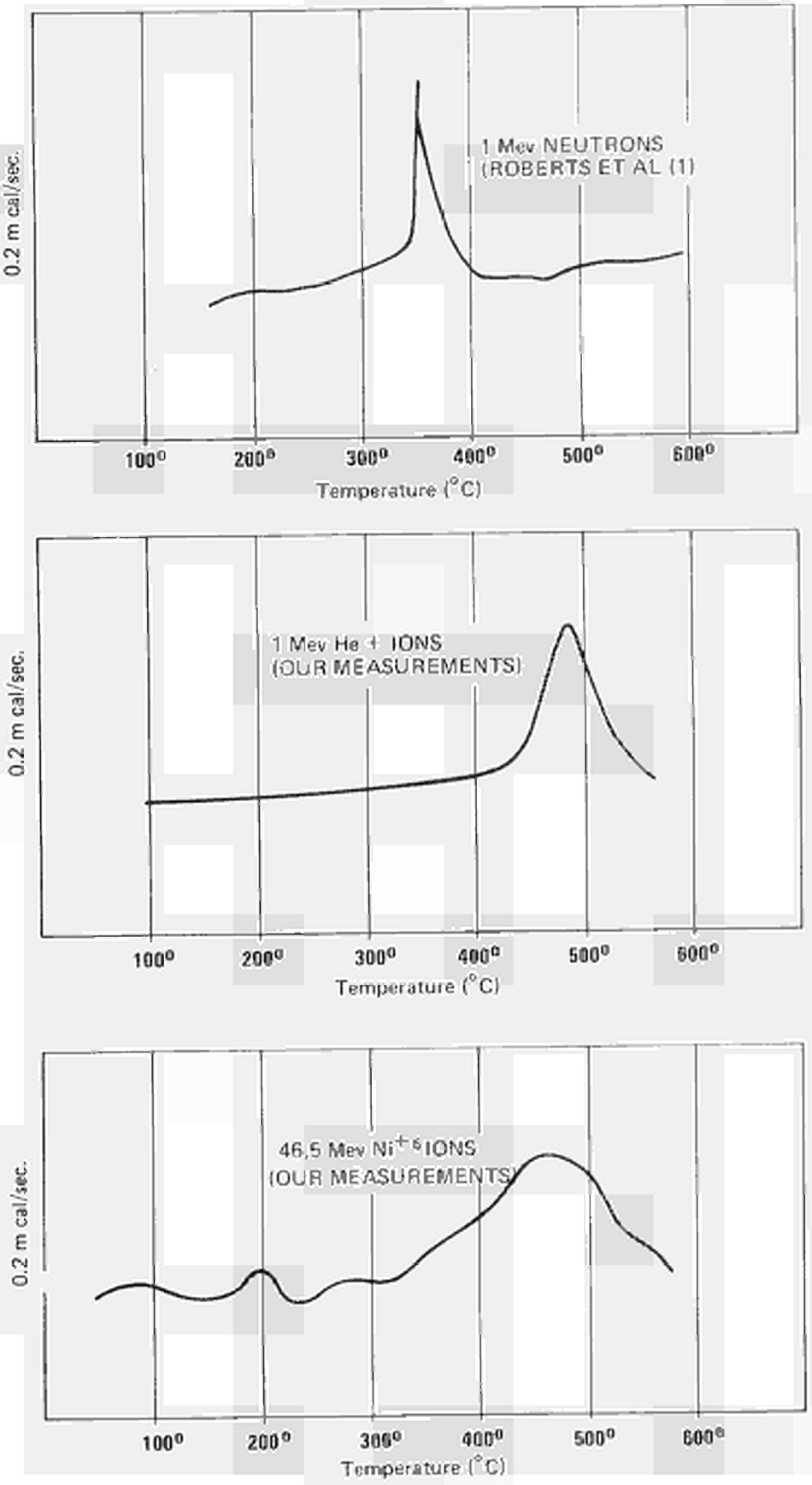

Fig. 1 — Stored energy of irradiated silica (samples weights ≃ 4 mg)

2.1.3 Interaction of Actinides with Environment

OBJECTIVES

The objective of this study is the quantitative description of the behaviour of the actinides in terrestrial and aquatic environments following loss from a geological repository containing alpha-bearing wastes.

For the second half of 1977 the planned activities were:

a) Production of a borosilicate glass spiked with Pu-238 and start-up of the study of the chemical characterization of the leaching solution.

b) Start-up of the experimental activity on plutonium migration through sub-soil columns.

c) Continuation of the study of Pu-behaviour in an aquatic system in order to determine the distribution and size range of formed particles as a function of pH and redox potential.

RESULTS

Glass Production

Due to the late delivery of Pu-238 the initial production of Pu-238 spiked glass had to be postponed until November 1977. The standard production procedures for the borosilicate glass are now well established and consist of three successive fusions of glass matrix powder (60% SiO_2, 18.8% B_2O_3, 6.2% Al_2O_3, 15% Na_2O) containing 20% of simulated waste oxides. The initial two fusions are carried out in nickel crucibles under a normal atmosphere, the last fusion is made in a graphite crucible in an inert atmosphere of argon at 1200°C. This is undertaken in order to reduce the molybdate phase formed in the previous melts.

Examination of slices of glass under a metallographic microscope showed that the matrix contained evenly distributed metallic inclusions probably composed of nickel and molybdenum.

Laboratory Simulation

a) Migration Studies

Pu-237 has been used to carry out initial studies on the migration of this element through aquatic sediments (lake, simulated estuarine and sea sediments) both at ambient and elevated temperatures (20°C and 80°C).

Two aliquots of each of the three sediments were contaminated in the laboratory with Pu-237 (+ 4 form chloride solution) by neutralizing a spiked solution to pH 6 - 8; sorption of the plutonium by the sediments was continued for 2 days. After this time the contaminated sediment was transferred to the top of columns containing identical but non-contaminated sediment. Lake, estuarine and seawater were passed through the corresponding columns over a period of 100 days, the pH of the water being controlled at either pH 8.0 or pH 6.0. The flow rate through the columns was adjusted to about 5 ml/h. Measurement of the break-through activity was carried out every second day. At the end of the 100 days the columns were frozen to - 20°C and then sliced into approximately 1 cm lengths. The distribution of activity was then measured by gamma-counting; the results for the first four columns are shown in Fig. 1.

The data indicate that the method employed is adequate to determine the evolution of the migration of plutonium in the sediment columns. The identification of the migrating species will form the basis of a subsequent part of the experimental programme over the next year; this will also include the determination of the chemical and mineralogical characteristics of various soils and sediments used in the present programme.

b) Aquatic Distribution and Transfer of Actinides

Preliminary studies were performed on the size distribution and evolution of suspended particulate material containing plutonium with time and pH variations. The results obtained indicate that in a freshwater environment particles $\geq$ 1.2 µm are rapidly formed (or particles already in the water absorbing very quickly the added plutonium) and these settled out of the water column in a few days. The transfer of plutonium from the water or the suspended particulate matter to biological organisms seemed to increase over the first two weeks and then to decrease slightly, coming to an intermediate steady level for the remaining 2.5 months of the experiment. During this latter period the water activity was below 2.5 pCi/ml.

COLLABORATION WITH EXTERNAL ORGANIZATIONS

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

- Deutsches Hydrographisches Institut, Hamburg, Germany,
- Biologische Anstalt Helgoland, Hamburg, Germany,
- CNEN, Laboratorio per lo Studio della Contaminazione Radioattiva del Mare, Fiascherino, Italia,
- CEN, Mol, Dept. of Radiobiology, Belgium.

CONCLUSIONS AND PLANNED ACTIVITIES

a) Owing to the delayed delivery of plutonium-238, the planned studies on physico-chemical characterization of solutions leached from glass will be started at the beginning of 1978.
These will be studied using a variety of separation techniques, initially consisting of ultrafiltration, solvent extraction and selective co-precipitation.

b) Following the preliminary experiments carried out in 1977, a systematic investigation on migration of plutonium and americium leached from glass, with soil and sediment columns will be started.

c) The definition of an experimental programme on the interaction of actinides with the biosphere is under detailed examination. The JRC contribution in the framework of a possible international cooperation (Radioprotection Programme - Indirect Action and specific national institutes) will be directed toward the physico-chemical parameters (speciation) affecting the bio-availability of actinides in benthic organisms (water-sediments interface).

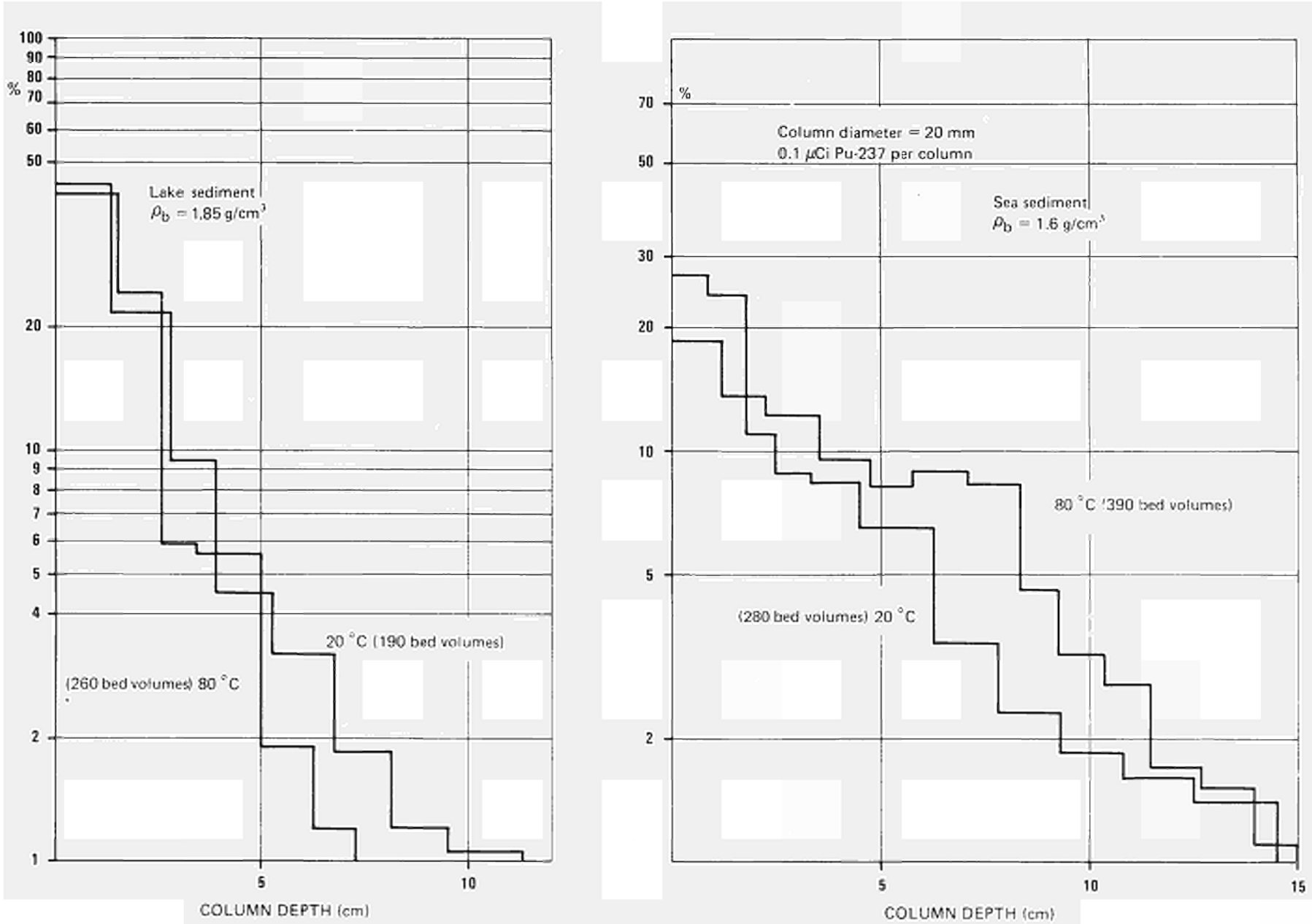

Fig. 1 — Migration behaviour of Plutonium into lake and sea sediment

2.1.4 Actinides Monitoring

OBJECTIVES

The study aims at the development of a methodology for plutonium waste monitoring in compliance with pertinent regulations. This goal is going to be pursued in the framework of an Advisory Laboratory. In the planning for the reporting period, we scheduled theoretical and experimental work concerning the development of reference instruments and methods based on passive gamma and passive neutron techniques.

RESULTS

During the reporting period the major activity was concerned with:

1. Experimental verification of the interpretational model for spontaneous fission neutron measurements (passive neutron assay).
2. Interpretation of spontaneous fission neutron measurements on plutonium-contaminated solid and liquid wastes.
3. Writing Chapter IV of our Guide [1] "Passive Neutron Assay".

The Interpretational Model

The interpretational model [1] for passive neutron assay has been tested experimentally in order to verify the theoretical prediction of some important neutron interaction effects on the rate of auto-correlated neutron detection signals.

The fast neutron removal effect by hydrogenous matrix materials was measured for a set of 10 wooden cylinders ranging in equivalent sphere radius [1] between 4 and 20 cm. Two types of wood having densities of about $\rho = 0.39$ g/cm^3 and 0.60 g/cm^3 were studied. The comparison with theoretical predictions shows agreement within experimental errors.

Thermal neutron induced fission effect was verified by interpreting spontaneous fission neutron measurements with aqueous plutonium-solutions of 1.5 l volume, ranging in plutonium density between 0.66 g/l to 66.0 g/l. It was found that the form of the experimental curve was described fairly well by the theory but the theoretical scale factor was about 30% too high.

The Interpretation of Passive Neutron Measurements

The interpretation of passive neutron measurements on plutonium-contaminated wastes from a plutonium handling laboratory (CNEN, Casaccia)

were continued, taking adjusted parameters of thermal neutron induced fission effects as outlined above. In accordance with waste monitoring objectives as outlined in Chapter II of ref. [1], the error limits span the range of maximum possible deviation from the quoted result. The possible bias of these results and their variances will be determined by destructive analysis of some selected waste items. This destructive analysis will be made by the Plutonium Laboratory of CNEN, Casaccia.

Chapter IV of our Guide "Passive Neutron Assay" has been written. It is going to be published as a EUR-report.

COLLABORATION WITH EXTERNAL ORGANIZATIONS

Contract with EUREX Saluggia, Italy, concerning the testing of on-line alpha-monitor (178-77 PIPGI, May 16th, 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 D.N.R.E. (Dounreay, United Kingdom) in view of a collaboration on the monitoring of the contaminated solid waste streams.

CONCLUSIONS

The development of the interpretational model for passive neutron measurements on plutonium-bearing materials in general and particularly on plutonium-contaminated solids, has been continued and partially tested experimentally. The experimental tests on fast neutron removal effects demonstrate the validity of our theoretical approach. The theoretical predictions of the thermal neutron-induced fission effect were higher by about 30% with respect to the experiments. For clarification of this discrepancy we need more accurate experimental data. Such experiments were scheduled for the reporting period, but were delayed due to lack of qualified personnel. These experiments will probably start in early May 1978.

The experimental tests on a liquid alpha-monitor in the EUREX plant (CNEN, Saluggia) have not been initiated either. These are scheduled now for the beginning of 1978 depending on the possibilities of EUREX.

Some academic studies on plutonium monitoring on the basis of our Guide [1] were started in a University (two Ph.D. - and one master-theses). Those studies will provide the enlargement of the scientific basis in this field.

In order to achieve the final goal, being the establishment of an Advisory Laboratory for the plutonium waste monitoring, the staffing of this activity has to be improved, otherwise we must eventually restrict the work to the development of the methodology only.

PLANNED ACTIVITIES

For the following semester are planned:

1. Experimental verification of the validity of the reference monitor for passive gamma-assay as conceptually defined in Chapter III of our Guide.
2. As 1., but for passive neutron-assay.
3. Testing of an on-line liquid alpha-monitor under operational conditions of a fuel reprocessing facility.
4. Writing Chapter V of our Guide: "Application of Active Neutron Assay."

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BIRKHOFF, G., BONDAR, L., NOTEA, A., SEGAL, Y., Chapter II: "Principles and Theory of Radiometric Assay", EUR 5636e (1976);

BIRKHOFF, G., NOTEA, A., Chapter III: "Application of Passive Gamma-Assay", EUR 5637e (1976);

BIRKHOFF, G., BONDAR, L., Chapter IV: "Application of Passive Neutron-Assay"; Chapter V: "Application of Active Neutron Assay", in preparation.

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, 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 JRC, during the reporting period, has edited the Proceedings of the First Technical Meeting on the Nuclear Transmutation of Actinides, held at the JRC-Ispra, March 16-18, 1977 (sponsored by the Nuclear Energy Agency of the OECD).

The activity of the JRC in this field includes experiments on the radiochemical methods required for actinides separation and assessment studies on the possibility of actinides transmutation in nuclear reactors. The assessment studies will consider, in addition to the reactor physics aspects, the implications on the fuel cycle of this advanced strategy (technological developments required, 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.

2.2.1 Chemical Separation of Actinides

OBJECTIVES

The objective of this activity is to develop on a laboratory scale a complete process flow-sheet for actinide removal from HAW. To test the feasibility of the proposed HAW partitioning flow-sheets (OXAL, HDEHP and TBP processes) tracer and fully active laboratory scale experiments have to be carried out under batch and countercurrent operating conditions.

During the reporting period it was therefore planned to complete the batch-extraction tests on HDEHP and TBP as well as on OXAL processes using a simulated HAW solution and to start experiments using a fully active HAW solution to be prepared in the hot cell of the LMA laboratory.

RESULTS

1. HDEHP Extraction Process

Most process steps of the HDEHP flow-sheet have already been tested under simulated conditions and results have been reported in the preceding Programme Progress Report [1] and in the external publication [2].

During the reporting period the following investigations have been carried out on simulated HAW solutions:

- Selective Back-extraction of Trivalent Actinides

  One of the operating conditions of the HDEHP process [1] still under investigation at tracer level, was the type of carboxylic acid to be employed along with DTPA for selective stripping of trivalent actinides from loaded HDEHP.
  The obtained results have shown that no significant difference exists between back-extraction kinetics measured using lactic or glycolic acid.

- De-acidification of HAW by Extraction of Nitric Acid

  To reduce the acidity of HAW an alternative option to the denitration by formic acid, could be the countercurrent extraction of HNO_3 from HAW by TBP [3, 4].

  The possibility of obtaining a simultaneous removal of plutonium and HNO_3 by a single extraction step using standard equipments (mixer-settler) was also investigated.

Preliminary tests showed the possibility of reducing the acidity of a simulated HAW solution from 4.5 to 0.1 M/l of nitric acid by a countercurrent extraction using 50% TBP in dodecane. The formation of precipitates and of interfacial cruds has been observed. Experiments are being done in order to find remedies.

2. TBP Extraction Process

The behaviour of residual plutonium during the preparation of concentrated and denitrated HAW as foreseen by the TBP flow-sheet [5] has been experimentally investigated on simulated HAW solutions.

- Plutonium Behaviour During HAW Concentration

The concentrated acidic HAW solution was obtained by successive concentration-denitration steps taking care that the acidity is maintained during the whole process within defined limits. The obtained experimental results are reported in Table 1.
They show that the adsorption of plutonium on the precipitates can be minimized by keeping, during the concentration-denitration, an acidity level not lower than 5 M/l of nitric acid.

Table 1 - Behaviour of Pu, Am and some other metal-ions during the concentration-denitration of 5,000 l/ton HAW

Acidity conditions during the concentration-denitration	% Element (3) remaining in the precipitate after leaching (4)						
	Pu-239	Am-241	Ce-144	Ru-106	Zr-95	Nb-95	Fe-59
8M > HNO_3 ≥ 5M (1)	~0.1	~0.015	~0.16	~0.25	7	23.5	∽0.5
8M > HNO_3 ≥ 1.7M (2)	4	~ 0.1	~0.5	~0.4	20	25	n.m.*

(1) concentration factor ≃ 12
(2) " " ≃ 14
(3) referred to the amounts present in the original HAW solution
(4) by cold and hot 4M HNO_3
* not measured

A concentration-denitration reactor vessel, equipped with a conductimetry cell, is being set up for a better control of the acidity during the process.

- Plutonium Behaviour During Denitration of Concentrated Acidic HAW

During the concentration and interim storage of concentrated HAW solutions an acidity above 5M HNO_3 would minimize the formation of plutonium-bearing precipitates.

However their production cannot be avoided during the subsequent denitration step, when the HAW acidity is significantly reduced. Experimental denitration tests by formic acid showed that precipitates containing 2-10% of residual plutonium were produced. A possibility to prevent them would be offered by a selective extraction of plutonium from concentrated acidic HAW provided that the residual plutonium is originally present in extractable form. The results obtained from batch-extraction tests performed with HDEHP were satisfactory [5].

3. OXAL Process

Experimental tests on simulated HAW solutions have been continued in order to optimize the oxalate precipitation and the actinide/REs separation steps. During the reporting period special attention was paid to the second step. Instead of using the solvent extraction technique, a process based on column extraction chromatography was selected, using as stationary phase HDEHP supported on LEVEXTREL, a product manufactured by BAYER.

- Description of the Process

The actinide and REs oxalates precipitated as previously described [1, 6] are dissolved by hot nitric acid (final acidity ≃ 3 M/l). Pu, Np, (Zr, Mo, U, Fe) are fixed on the chromatographic column, whereas other components (Am, Cm, REs + Sr, Ba and FPs traces) remain in the solution flowing through the column.
The fixed elements are eluted at 60-80°C by means of a saturated oxalic acid solution.
The solution flowed through the first column, is de-acidified up to a pH value of 2; Am, Cm and REs are fixed on a second LEVEXTREL column whereas Sr, Ba and FPs traces flow through the column. Am and Cm can then be selectively eluted by a DTPA solution buffered at pH 3 and REs by a 3-4M HNO_3 solution.

Results obtained from experimental tests on simulated HAW showed that the separation of actinides by the OXAL process is quite satisfactory. Separation yields of about 99.5% for plutonium and neptunium and 99.8% for americium and curium can be in fact attained.
The Am- and Cm-fraction retain 5% of the rare earths initially present. An additional purification cycle is necessary if a lower REs content is wanted.

4. Experiments on Fully Active HAW Solutions

The preparation of fully active HAW solutions by dissolution of UO_2 fuels, irradiated at a burnup of about 25,000 MWD/t, and by simulation of the PUREX process using solvent extraction, has been started in November 1977 in a suitably equipped hot cell of the

LMA laboratory. So tests on real HAW solutions for the OXAL and HDEHP processes will be started in January 1978.

The construction of two new chemical lead cells was finished. These cells are being equipped for countercurrent experiments. An analytical laboratory is also being equipped for the analysis of HAW samples. Emission spectroscopy using a copper spark source, will be used.

5. Engineering Implications of Actinides Separation

An investigation of the problems related to the development of full-scale radiochemical separation plants, based on the laboratory flow-sheets, was initiated. As a first step detailed flow-sheets and flow-diagrams are being laid-out.

CONCLUSIONS

The completion of batch experiments on HDEHP and TBP, as well as on the OXAL process using a simulated HAW solution, is the most important objective fulfilled during the reporting period.

Sufficiently high separation yields for Pu, Np, Am and Cm have been measured (see Table 2), according to the different process flow-sheets. Concerning the OXAL process preliminary experiments carried out on real waste from reprocessing plants, confirmed the results of the experiments on simulated HAW solutions. These processes have to be tested now on fully active HAW solutions.

Due to some delay in the hot cell preparation, the start-up of this phase of the work will be possible in January 1978.

PLANNED ACTIVITIES

During the following semester it is planned:

- to initiate HDEHP and TBP batch-extraction tests on real HAW,
- to continue OXAL process tests (oxalate precipitation and actinide/REs separation) on real HAW,
- to initiate countercurrent tests on simulated and traced HAW,
- to lay-out detailed flow-sheets and flow-diagrams of the different processes in order to evaluate the engineering implications of actinides separation.

Table 2 - Percent separation yields measured by HDEHP and TBP extraction and oxalate precipitation tests, performed in batch on simulated HAW solutions

Process Flow-sheet	Actinide Separation Method	Elem.	Process (1) Conditions	Separation Yields (%)
TBP Extraction Process (option 1)	Solvent Extraction	Pu	0.25M HDEHP ~ 4M HNO_3	>99.8 (2)
			30% TBP ~ 4M HNO_3	75 (2)
		Am (Cm)	30% TBP 1.3M $NaNO_3$ 0.6M $Al(NO_3)_3$ 0.17M HNO_3	>99.9 (3)
HDEHP Extraction Process (option 1)		Pu	0.25M HDEHP ~4M HNO_3	>99.8 (2)
		Np		80 (2)
		Am (Cm)	0.25M HDEHP pH ~ 2	96.2 (2)
Modified OXAL Process	Denitration + oxalate precipitation	Pu	Simultaneous addition of formic and oxalic acid	99.5
		Np		99.2
		Am (Cm)		99.8

(1) for simulated HAW solutions at different concentration levels: ~ 500 l/ton U for TBP and ~ 5,000 l/ton U for HDEHP and OXAL processes,
(2) after 1 extraction stage,
(3) after 4 extraction stages.

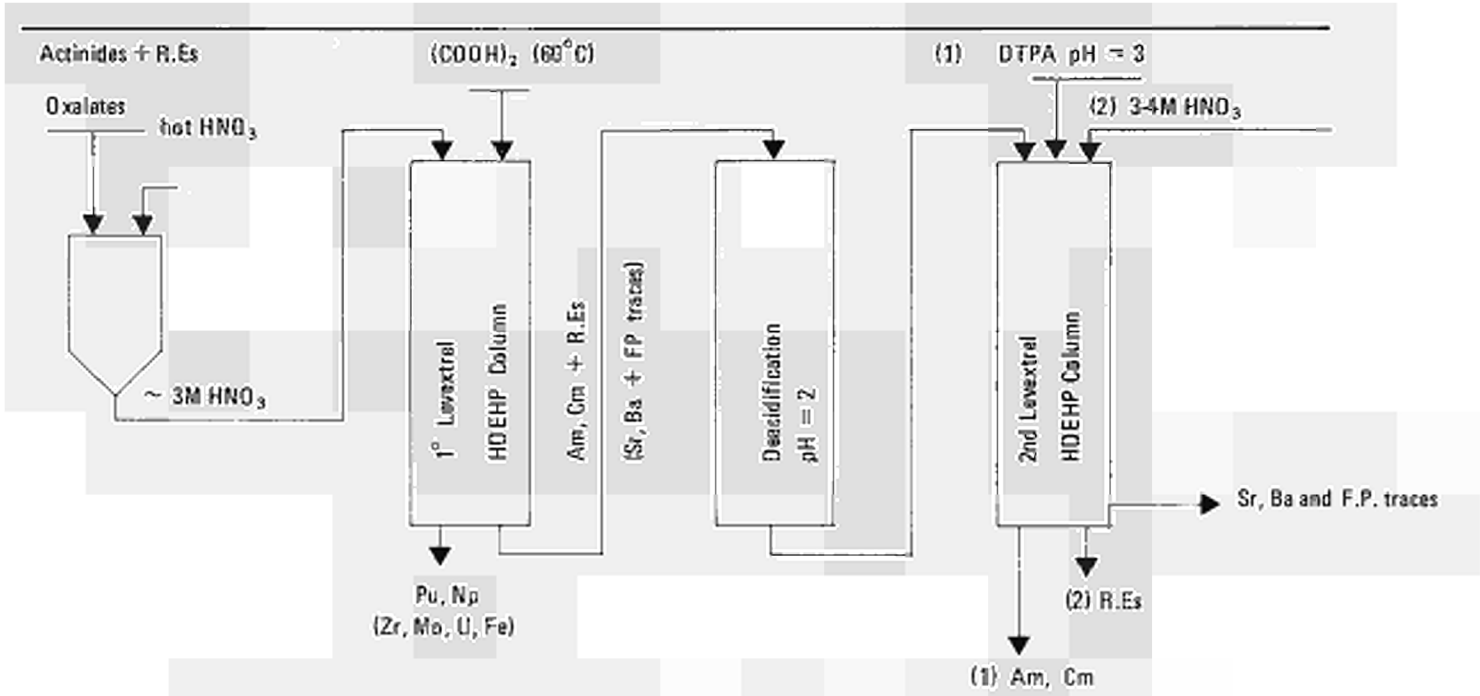

Fig. 1 – Process scheme for Am, Cm/R.Es separation by column extraction chromatography

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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. An overall strategy for transmuting those actinides, produced by the European Community power generating system, should be proposed giving indications as to the reactor type to be preferred as transmutation device and the way of introducing the actinides in the reactor. Taking account of the results for risk and cost analysis, a choice between the various possible recycle strategies should be carried out.

RESULTS

Organizational Work

Considerable effort has been made to reorganize the work due to an increase of staff and to establish contacts with groups inside and outside the European Community. A master plan for the activity has been elaborated defining 20 tasks which have to be investigated before being able to judge this advanced waste management scheme. This plan contains descriptions of each task, suggestions for its technical execution, and time schedules.

Radiation Properties of Materials within the Fuel Cycle

The radiation properties of the fuel components essentially determine the lay-out of the transport casks, the way of manufacturing fuel elements, and the reprocessing scheme. In order to evaluate the additional implications due to the presence of actinides other than fuel, it is necessary to quantify at first the radiation levels encountered in the different fuel cycles of existing and advanced reactors and compare them to those arising when minor actinides are recycled. Corresponding data for plutonium-containing fuel of LWRs and FBRs have been calculated as function of the decay time. The main results for decay time equal to zero are summarized in the following table. They are compared to the values characterizing minor actinides in Table 1.

Table 1 - Radiation Properties of Plutonium and Actinides

	Decay Heat (W/g)		Photon Surface Dose Rate (rad/g.h)		Number of Neutrons (n/g.sec)		Neutron Surface Dose Rate (rem/g.h)	
	Pu	Act.	Pu	Act.	Pu	Act.	Pu	Act.
LWR 1st Cycle	1.37-2	1.06	5.1	95.8	1.12+3	7.84+5	2.83-1	1.98+2
LWR Equilibrium	1.92-2	1.04	7.3	55.2	1.62+3	1.70+9	4.10-1	4.30+5
FBR 1st Cycle	1.37-2	1.81	5.1	562.5	1.12+3	6.46+5	2.83-1	1.63+2
FBR Equilibrium	3.94-3	2.01	1.62	333.4	5.17+2	3.49+6	1.31-1	8.83+2

Risk Analysis

A computer program is being developed to quantify the reduction of long-term hazard obtained by recycling actinides other than fuel back to fission reactors.

As parameters are included different recycle strategies, the transmutation rate, the total time interval during which nuclear energy will be generated, the extraction efficiencies for minor actinides and plutonium as well as the routine losses during the individual fuel cycle operations.

Moreover, work has been initiated to set up a methodology to calculate the additional risk due to the presence of actinides other than fuel in the fuel cycle.

At least as far as the operational personnel of the fuel cycle facilities is concerned, the risk will be determined in terms of the dose rate to man. A collection of release data for the various constituents within the fuel cycle has been started, distinguishing between reactor operation, reprocessing and fuel element manufacturing.

Contribution of the JRC-Karlsruhe Establishment

A. Neutron Rates Obtained in the Rapsodie Reactor

The accurate knowledge of fission product neutron absorption in fast reactor fuel is needed to predict reactivity losses which deter-

mine, amongst other things, the breeding gain.
Lists requested by IAEA Working Groups [1, 2] require accuracies between 10 and 30%. Some of these nuclides have been analyzed. The results, when compared with microscopic data of different origin [3, 4, 5], are in some instances in good agreement (Table 2).

The concept of nuclear incineration of minor actinides in a fast reactor needs rather precise neutron cross sections in order to improve the economics of the process. Some of the nuclides of interest were included in the TACO-experiment, and the absorption-, capture- and fission-cross sections are given in Table 3.

B. Methodology of Actinide Prediction

Several methods are used in predicting the actinide build-up in power reactor fuels:

- reactor physics' calculations which do not usually include the calculation of the minor actinides (Np, Am, Cm),
- zero-dimensional, one-group calculations of the ORIGEN-type,
- isotope correlations (IC) which require historical data.

In order to assess the amount of information required to apply these methods and to compare their prediction capabilities, trial calculations on previously analyzed [6, 7] fuel from TRINO VERCELLESE were performed.

Reactor physics' calculations are based on extensive information about reactor operation, usually only accessible to the operator. ORIGEN also requires information on the fuel history and is heavily dependent on a library of neutron cross section data. On the other hand, the IC method does not need information which is not normally available.

Preliminary results from 3 samples show that for most of the nuclides the prediction capabilities of reactor physics and IC are comparable (Table 4). The ORIGEN calculations show the largest deviations from the experimental observations and become progressively poorer for the higher actinides. The original version of ORIGEN which was also used by Haug [8] and Claiborne [9] was employed in these studies.

Table 2 - Comparison Between Measured and Evaluated Neutron Reaction Rates of Selected Fission Products

Isotope	$\phi\sigma t \cdot 10^{-3}$				Accuracy required (%)
	TACO	ref. [1]	ref. [2]	ref. [3]	
Nd-143	7.876	7.874	8.408	5.665	20
Nd-144	2.419	2.998	2.967	-	-
Cs-133	10.850	13.990	12.590	10.986	10 - 30
Nd-146	3.017	3.875	3.720	-	20
Nd-148	3.982	4.247	3.914	-	20
Nd-150	4.794	10.1	7.045	-	-

Table 3 - Neutron Reaction Rates of Selected Heavy Nuclides Measured in the TACO-Experiment

Isotope	$\phi\sigma t \cdot 10^{-2}$		
	Absorption	Fission	Capture
Np-237	7.452	3.635	3.817
Am-241	9.245	3.738	5.509
Am-243	8.330	2.510	5.820

Table 4 - Comparison of Experimental and Calculated Actinide Values

OBS - Observed Values (atoms/initial metal atoms)
IC - Isotope correlation prediction
OR - ORIGEN calculation value
PH - Calculated value using code [10]

n. m. = not measured
n. c. = not calculated
n. c. e. = no correlation exists

	G - 7 - 1 - 1				G - 7 - 1 - 4				L - 54			
	OBS	IC	OR	PH	OBS	IC	OR	PH	OBS	IC	OR	PH
U-236 (10^{-3})	1.64	1.87	1.46	1.53	2.55	2.56	2.40	2.30	3.51	3.72	3.57	n. c.
Pu-239 (10^{-3})	4.19	5.62	3.23	3.95	6.57	5.62	4.46	5.22	6.03	5.94	4.93	n. c.
Pu-240 (10^{-4})	5.95	5.23	4.05	5.66	14.6	12.4	9.90	10.9	17.5	17.5	15.5	n. c.
Pu-241 (10^{-4})	1.95	2.10	0.63	1.80	6.41	5.22	2.65	5.79	10.5	10.5	5.6	n. c.
Pu-242 (10^{-4})	.180	.260	.042	n. c.	1.09	.916	.327	n. c.	2.40	2.41	1.12	n. c.
Am-241 (10^{-5})	1.65	n. c. e.	.154	n. c.	2.06	n. c. e.	.542	n. c.	24.0	n. c. e.	2.71	n. c.
Am-243 (10^{-6})	.872	.985	.223	n. c.	8.19	6.74	3.33	n. c.	n. m.	42.8	19.1	n. c.
Cm-242 (10^{-6})	.57	.01	.10	n. c.	3.07	4.98	.85	n. c.	24.8	30.0	8.60	n. c.
Cm-244 (10^{-6})	.19	.87	.01	n. c.	2.01	.90	.38	n. c.	9.30	9.73	3.55	n. c.

COLLABORATION WITH EXTERNAL ORGANIZATIONS

The contract with the Austrian Academy of Science concerning the development of a computer program for calculating cross sections by models for the nucleus has been prolonged for four months in order to permit the introduction of the "second chance" fission effect.

Preliminary contacts have been established with GfK Karlsruhe, KFA Jülich and CNEN, Casaccia, which should lead to contracts of collaboration to be defined in the near future.

CONCLUSIONS

Apart from the fact that the scientific coordination and the preparation of proposals for collaboration with other institutions has absorbed some manpower, work has proceeded normally. The paper studies concerning the fuel cycle implications, however, should be intensified. Thus it is absolutely necessary that the increase of personnel allocation scheduled for 1978 be kept.

PLANNED ACTIVITIES

During the next reporting period, the following results are expected:

- Updated LWR library of nuclear data which should permit to reproduce experimentally determined isotope distributions of irradiated LWR fuel samples;
- A collection of fuel element performance data;
- Radiation properties of materials within the thorium fuel cycle.

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2.2.3 Actinide Cross Section Measurements

OBJECTIVES

For assessment studies of the nuclear transmutation of actinides as a waste management alternative, fission cross section measurements as function of energy are performed in order to check or improve these data. At present σ_f of Am-241 is measured as function of energy in the range of 0.006 MeV $\leq$ E $\leq$ 1 MeV. Two different techniques are applied. One relates the detected prompt fission neutrons to the fission cross section, which requires the knowledge of ν i.e. the number of prompt fission neutrons emitted per fission. In the other technique the fission fragments are detected [1] which are emitted from thin targets after the fission event.

In both methods the 3 MeV Van de Graaff accelerator of the Institute for Applied Nuclear Physics of the GfK, Karlsruhe has been used as neutron source. The first cross section measurements were started during 1976 with the first technique. The fission fragment detection method is being applied during late 1977.

RESULTS

The Neutron Detection Method

The analysis of the experimental data has been terminated for the determination of the fission cross section, the neutron energy and its resolution. Work has still to be done for the analysis of the errors. Work was continued with the measurement data obtained with thick Li targets (6 keV $\leq$ E $\leq$ 120 keV). These targets produced a white neutron spectrum burst of about 0.8 ns duration for irradiation of the test targets [2, 3]. The obtained background and isotope corrected time-of-flight spectra permitted the analysis of about 26 energy intervals which were condensed to 7 cross section values. These condensed cross section values with increased energy resolution are rather insensitive to the analysis procedure (unfolding, unfolding and smoothing, no unfolding, no unfolding and smoothing) of the time-of-flight spectra. The data obtained agree well with those of Shpak [4] but are up to a factor of 40 smaller than those listed in the ENDFB-IV data file.

For this reason all fission rates of Am-241 using these data files are overestimated in the case of LMFBRs with peak spectra around 100 keV. Our earlier results of the Am-241 fission cross section in the energy range of 200 keV to 500 keV were greater up to a factor of 3 compared with the ENDFB-IV data file, which could lead for extremely hard fast breeder spectra to an underestimation of the Am-241 fission rate.

Fission Fragment Detection

The gas scintillation chamber has been installed at the Van de Graaff of the GfK, Karlsruhe during July 1977. First measurements were performed during September 1977. The analyses of the data suggested some modifications on the electronics and a new measurement series was started during late 1977.

COLLABORATION WITH EXTERNAL ORGANIZATIONS

The activity is based on a collaboration between the GfK, Karlsruhe, Institute for Applied Nuclear Physics and the JRC-Ispra. The GfK provides the 3 MeV Van de Graaff as a neutron source, its operation instrumentation and computer soft- and hardware. The execution of the experiments is a common effort and the data analysis is in charge of the JRC-staff.

CONCLUSIONS

The measurement of the Am-241 fission cross section was justified due to the large discrepancies existing between ENDFB-IV data and those of ref. [4]. Our measurements confirm rather the data of ref. [4].

PLANNED ACTIVITIES

It is envisaged to complete during the first semester of 1978 the fission cross section measurements on Am-241. During this period a decision will be taken on a possible continuation of the cross section measurements on other actinides.

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2.3. DECONTAMINATION OF REACTOR COMPONENTS

OBJECTIVES

The main objective is to improve the knowledge of the mechanisms of reactor component contamination and of the techniques used for their decontamination.

At the last meeting of the Advisory Committee for the Programme Management (October, 1976), the members suggested to orient the activity towards the waste problems related to decontamination processes and decommissioning operations. These suggestions have been taken into account in the definition of the 1978 activities.

RESULTS

General Orientation

In order to be able to better define where the main interest lies of utilities, a contract has been given to the firm Laborelec (Belgium). The scope of the contract is to perform an inquiry in the different power stations in order to define the need in Europe of decontamination and the scope of the decontamination operations. The final report has been received and is now underway to be published as a EUR-report.

A bibliographic review on the state of the art in the field of decontamination of primary circuit components of LWR is almost concluded and a report is in preparation. This report also takes into account the papers presented at the Conference on Water Chemistry of Nuclear Reactor Systems (Bournemouth, October 1977), such as to permit a very recent updating.

Effect of Thermal and Redox Cycling

The scope of this activity is to analyze the effect of thermal cycling and of variation of oxygen concentration on the release of material from the layer deposited on the surface.

A high-temperature water loop, which was used previously for the study of the influence of the water chemistry on microsuspension generation, has been adapted to these new tests.

Some tests have been performed under conditions typical of a BWR, which means with a certain amount of oxygen present. Whereas in the preceding tests without oxygen the solid corrosion products were

mainly magnetite, under these conditions even ferric oxide was present. A more detailed analysis of the surface layer is underway.

Preliminary Tests on Various Decontamination Techniques

Some preliminary tests have been performed on the electrochemical method. If an intimate contact between the brush and the surface is maintained, a good decontamination can be obtained. The conditions are those typical of electropolishing.

Some preliminary tests with chemical methods (citrox and alkaline permanganate) have shown that the efficiency of the treatment depends strongly on the composition and age of the sample and on the concentration of the reagents. A systematic study seems necessary.

PLANNED ACTIVITIES

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

- Effect of Thermal and Redox Cycling

  The effect of thermal cycling on well-consolidated layers will be examined, following the release of soluble cations and suspended particles. The surface composition will be followed.

- Physico-Chemical Structure of Oxide Layers

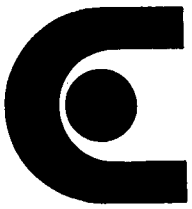
  The corrosion and deposited layers follow a continuous evolution. A study will be performed trying to define the change arising during the life of the layer. The study will be performed also on contaminated layers.

- Systematic Study of Chemical Decontamination

  A systematic study will be initiated in order to define what are the conditions which allow to minimize the waste arising.

- Preliminary Evaluation of the Decommissioning of the ISPRA-I Reactor

  An evaluation of the possibilities and interest to execute the decommissioning of the ISPRA-I reactor as a test case will be performed.



Conclusions

3. CONCLUSIONS

In the second semester of 1977 satisfactory progress has been made in the development of the programme.

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 waste hazard analysis the collaboration with CEN, Mol - scientists made possible the utilization of the methodologies set up during the previous years to quantify the probabilistic value of the geological barrier in the Boom (Belgium) clay formation.

Collaborations have also been established with laboratories involved in the indirect action programmes Radiation Protection and Management and Disposal of Radioactive Waste. In particular the formation has to be mentioned of a working group on Risk Analysis in Geological Disposal in the framework of the fiche 7 of the indirect action programme. The JRC assures a permanent secretariat of the group.

In the field of the long-term stability of conditioned waste, directed to provide input data for the waste hazard models, most of the post-irradiation experiments on the glasses irradiated in the HFR of Petten have been carried out. The delay in the delivery of the instrument for stored energy measurements did not permit to complete the experiments. The experiments on the glass leaching in pure water are almost terminated.

In the field of actinides monitoring a new chapter of our Guide has been completed. Collaborations have been established with scientific institutes and nuclear plant operators.

In order to achieve the final goal of setting up an Advisory Laboratory for plutonium waste monitoring we plan to reinforce the staff allocated to this activity.

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

Chemical Separation and Nuclear Transmutation of Actinides

In the field of the chemical separation of actinides, the batch tests on simulated HAW solutions have been completed for three flow-sheets based on oxalate precipitation and solvent extraction with TBP and HDEHP. The separation yields determined are quite satisfactory.

The three flow-sheets have now to be tested on real HAW solutions in hot cells.

Preliminary investigations were started on the engineering implications of actinides separation. The recruitment of a chemical engineer at the beginning of 1978 will offer the possibility of a more important effort in this matter.

In the field of the assessment studies on nuclear transmutation an activity has been started on the evaluation of the technological and risk implications of this advanced strategy.

Due to the complexity of the studies, we plan to reinforce, during 1978, the staff allocated to this activity by a reduction of the effort on the actinides cross section measurements. We also plan to conduct part of the assessment studies in collaboration with national organizations.

The planning of the future activities (see Table 2) is directed to produce for the end of 1979 a major report dealing with a critical evaluation of the feasibility of the chemical separation and nuclear transmutation of the actinides.

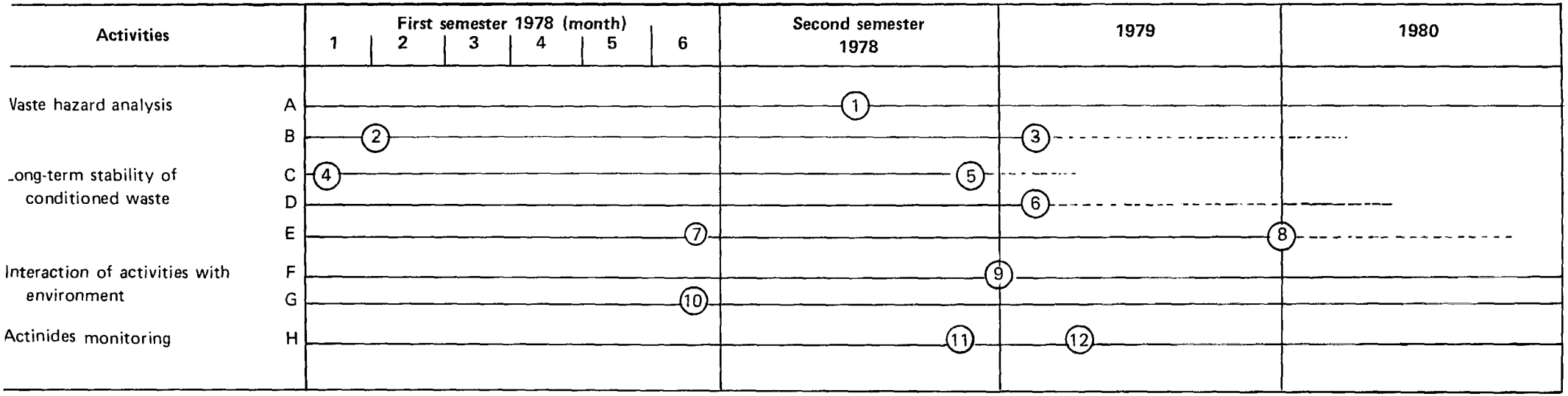
Decontamination of Reactor Components

On the basis of a literature review and of the preliminary experiments the activity has been oriented on some points (chemical methods, physico-chemical structure of oxide layers,effect of thermal and redox cycling) of interest for decontamination processes in power station operations.

The planning for future activities has decision points at the end of 1978. In this planning (see Table 3) also an evaluation of the possibility and interest to execute the decommissioning of the ISPRA-1 reactor as a test case, has been included.

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

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 terrestrial environment, introduction of marine environment)

③ Model revision

④ Start-up and completion of the experiments with water in conditions of geological disposal

⑤

⑥ Completion of the experiments on bituminized waste and possible start-up of experiments on other matrices

⑦ Decision point: completion of the experiments on the radiation damage simulation by fission fragments or start-up of a new irradiation test

⑧ Completion of the experiments on the validity of accelerated tests

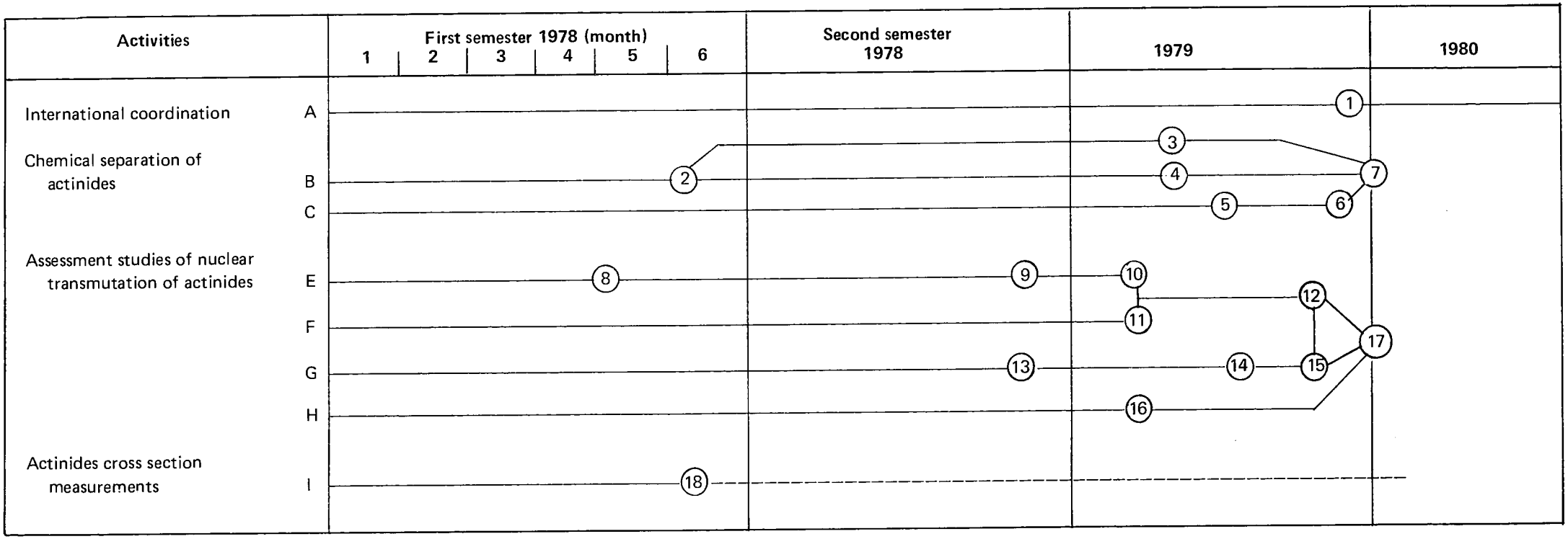
⑨ Conclusion of initial column (soil and sediments) experiments with leached actinides and development of further experimental programme

⑩ Definition of the experimental programme

⑪ Completion of the chapter V of the guide "Application of active neutron assay"

⑫ Revision of the Guide

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
- 1 Organization of an international meeting

B OXAL process
- 2 Completion of the experiments on precipitation of actinides + rare-earths oxalates
- 3 End of laboratory scale experiments on OXAL (separation of actinides from rare-earths)
- 4 Completion of preliminary engineering evaluations

C Solvent extraction
- 5 Completion of the fully active scale batch experiments with HDEHP and TBP
- 6 Completion of the countercurrent experiments with HDEHP and preliminary engineering evaluations
- 7 Report on feasibility of actinides separation. Tentative cost evaluation. Proposal for further actions

E Reactor physics calculations
- 8 Establishment of reactor physics calculation methods
- 9 Generation of nuclear data set
- 10 Completion of the reactor physics calculations

F Collection of chemical and physical data for fuel element design
- 11 Proposal for fuel element designs containing actinides other than fuel
- 12 Elaboration of an overall recycle strategy

G Cost and risk analysis
- 13 Establishment of cost calculation procedure
- 14 Establishment of the methodology of risk assessment
- 15 Results of cost and risk assessment

H Study of the implications of actinide recycling on the fuel cycle
- 16 Proposal of changes in the fabrication plant
- 17 Preparation of a report on the feasibility of the actinides transmutation

I Differential cross section measurements
- 18 Completion of the measurements on Am-241

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

Activities		First semester 1978 (month) 1 2 3 4 5 6	Second semester 1978	1979	1980
Decontamination of reactor components	A	①			
	B	②	③		
	C	④	⑤		
	D		⑥		

A Systematic study on chemical decontamination
1 Comparative tests on contaminated samples

B Study on the physico-chemical structure of oxide layers
2 Electron microscope examination of radioactive samples
3 Decision point

C Partial decontamination using thermal redox cycling
4 End of the experiments on the thermal cycling
5 End of the experiments on the redox cycling - Decision point

D Preliminary evaluation of the decommissioning of the Ispra 1 reactor
6 Redaction of an operational proposal - Decision point

4. JRC PUBLICATIONS

Published or Presented

1) GIRARDI, F., BERTOZZI, G., D'ALESSANDRO, M., "Long-Term Risk Assessment of Radioactive Waste Disposal in Geological Formations", paper presented at the 1977 Annual Meeting of the American Nuclear Society, New York, 12-17 June 1977; EUR 5902e (1977)

2) BERTOZZI, G., D'ALESSANDRO, M., GIRARDI, F., VANOSSI, M., "Safety Assessment of Radioactive Waste Disposal into Geological Formations: A Preliminary Application of Fault-Tree Analysis to Salt Deposits", paper presented at the "Workshop on Risk Analysis and Geologic Modelling in Relation to the Disposal of Radioactive Wastes in Geologic Formations", Ispra, 23-27 May, 1977; EUR 5901e (1977)

3) MANNONE, F., CECILLE, L., "Removal of Long-Lived Actinides from Purex Type HAW Raffinates: Development of Conceptual and Experimental Studies on Solvent Extraction"; EUR 5816e (1977)

4) CECILLE, L., LANDAT, D., MANNONE, F., "Séparation des actinides des solutions de déchets nucléaires à haute activité (HAW) par extraction par solvant. I: Description d'un schéma de séparation faisant appel au TBP", Radiochem. Radioanal. Letters, 31 (1) (1977) 19-28

5) CECILLE, L., LE STANG, H., MANNONE, F., "Séparation des actinides des solutions de déchets nucléaires à haute activité (HAW) par extraction par solvant. II: Description d'un schéma de séparation faisant appel à l'HDEHP", Radiochem. Radioanal. Letters, 31 (1) (1977) 29-38

6) MOUSTY, F., TOUSSAINT, J., GODFRIN, J., "Séparation des actinides des déchets liquides de haute activité - Le procédé OXAL", Radiochem. Radioanal. Letters, 31 (1) (1977) 9-18

7) CECILLE, L., LANDAT, D., LE STANG, M., MANNONE, F., "Séparation des actinides des solutions de déchets nucléaires à haute activité par extraction par solvant - Etat des travaux expérimentaux réalisés au CCR-Ispra", Proceedings of the First Technical Meeting on Nuclear Transmutation of Actinides, JRC-Ispra, 16-18 March, 1977; EUR 5897e,f (1977) 145

8) MOUSTY, F., TOUSSAINT, J., GODFRIN, J., GIRARDI, F., "Séparation des actinides des déchets liquides de haute activité - Le procédé OXAL", ibidem, 163

9) HAGE, W., SCHMIDT, E., "Reactor Physics Aspects of Burning Actinides in a Nuclear Reactor", ibidem, 13

10) SCHMIDT, E., CAMETTI, J., "On the Neutron Physical Feasibility of Transmutation of Actinides other than Fuel in Nuclear Power Reactors", ibidem, 177

11) SOLA, A., "Some Preliminary Results on Actinide Incineration and Transmutation in a Thermal and in a Fast Reactor", ibidem, 281

12) SOLA, A., CARUSO, K., "Sensitivity Studies and Actinide Nuclear Data Requirements", ibidem, 337

13) SCHMIDT, E., "Influence of Nuclear Data Uncertainties on Results when Recycling Actinides other than Fuel", ibidem, 359

14) KOCH, L., ERNSTBERGER, R., KAMMERICHS, K., "Formation of Minor Actinides and Requirements of Nuclear Incineration", ibidem, 247

15) HAGE, W., HETTINGER, H., KÄPPELER, F., KUMPF, S., WISSHAK, K., "Measurements of the Fission Cross Section of Am-241", ibidem, 423

16) LANZA, F., "Simulation of Alpha-Damage in Glasses by Means of Fission Induced by Epithermal Neutrons", paper presented at the HFR Users Meeting, Petten, October, 1977

Submitted for Publication or Presentation

1) ZAMORANI, E., "Surface Dose Rate Contribution by Actinides Mixed in Uranium-Plutonium Fuel Elements", to be published as EUR-Report

2) ROOFTHOOFT, R., "La Décontamination dans les Centrales Nucléaires Refroidies à l'Eau Légère - Résultats d'une Enquête dans les Centrales Européennes", to be published as EUR-Report

3) LANZA, F., PARNISARI, E., "Evaluation of Long-Term Leaching of Borosilicate Glasses", to be published as EUR-Report.

LIST OF AUTHORS

Programme Manager: M. Bresesti

Assistant to the Programme Manager: J. Ley

Scientific Coordinator: F. Girardi

Project 1:
A. Avogadro
G. Bertozzi
G. Bignoli
G. Birkhoff
L. Bondar
M. D'Alessandro
F. Girardi
F. Lanza
P. Lensi
A. Manara
N. Murray
J. Schneider
P. Van Rutten
T. Visani

Project 2:
L. Anselmi
L. Cecille
F. Girardi
W. Hage
H. Hettinger
S. Kumpf
F. Mannone
F. Mousty
E. Schmidt
A. Sola
E. Zamorani

L. Koch (JRC - Karlsruhe)
F. Käppeler (Institute for Applied Nuclear Physics, GfK,
K. Wisshak " " " " Karlsruhe)

Project 3:
O. Gautsch
F. Lanza
F. Van Rutten
P. Weisgerber

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