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Ispra Establishment
Italy



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Management of Nuclear Materials and Radioactive Waste

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PROGRAMME PROGRESS REPORT

July - December 1979

Programme Progress Report JRC Ispra
July - December 1979

MANAGEMENT OF NUCLEAR MATERIALS AND RADIOACTIVE WASTE

Abstract

This document is the progress report of the programme Management of Nuclear Materials and Radioactive Waste of the Joint Research Centre for the period July - December 1979:

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

In the framework of a Risk Analysis Group the JRC is promoting contacts between people of the Community working in the area of risk assessment of geological disposal.

In the field of the long-term stability of conditioned waste, radiation damage studies and leaching experiments on vitrified and bituminized waste have been performed.

Experiments have been carried out on the migration behaviour of actinides (Pu, Am and Np) leached from a glass matrix.

In the field of actinide monitoring the development of instruments and of interpretation models for non-destructive assay techniques has continued. The methodology developed by the JRC is being tested in nuclear plants.

Project 2 - Chemical separation and nuclear transmutation of actinides

In the field of chemical separation of actinides from HAW, solvent extraction techniques using HDEHP and TBP are investigated.

The assessment studies on nuclear transmutation of actinides include reactor physics calculations, fuel element design, evaluation of risk and of implications on the nuclear fuel cycle.

Project 3 - Decontamination of reactor components

The project includes studies on chemical decontamination and on physico-chemical structure of oxide layers.



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CONTENTS

1.	Introduction.....	3
2.	Projects.....	7
2.1	Evaluation of Long Term Hazard of Radioactive Waste Disposal.....	7
	Waste Hazard Analysis.....	7
	Long Term Stability of Conditioned Waste.....	8
	Interaction of Actinides with the Environment.....	10
	Actinides Monitoring.....	12
2.2	Chemical Separation and Nuclear Transmutation of Actinides.....	15
	Chemical Separation of Actinides.....	15
	Assessment Studies on Nuclear Transmutation of Actinides.....	19
	Actinides Cross-Section Measurements.....	25
2.3	Decontamination of Reactor Components.....	27
3.	Conclusions.....	31
4.	JRC Publications.....	37

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 1979 consists of 62 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

It comprises 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 on Radioactive Waste Management. 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 is 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. 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 other matrices
- Retention of actinides by geological media
- Environmental 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 model development we are passing from generalized 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.

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 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 action programme Radiation Protection. Joint actions have been set up to strengthen the links between the two programmes.

A study on actinide monitoring is also included in this project: this study is important both for safety reasons and for optimizing the waste management.

Waste Hazard Analysis

OBJECTIVES

Aim of the study is to develop a methodology for the assessment of the risk linked to the disposal of radioactive wastes into geological formations. Probabilistic and deterministic approaches are used, in function of the nature of the barriers considered. For the probabilistic approach the Fault Tree Analysis method is utilized; for the deterministic approach the potential levels of environmental radioactive pollution and the corresponding dose-rates to man are calculated, through the development of appropriate models.

For the second semester of 1979 the planned activities were:

- A** To describe in a comprehensive report the application of the probabilistic analysis to a specific clay site.

- B** To publish as EUR report the results on risk analysis for different fuel cycle strategies, described in the previous Programme Progress Report¹.

- C** To continue the collaboration with CEN/SCK, Mol, through the development of a «Consequence Analysis», related to the possible creation of a waste repository in the Belgian clay formation of Boom.

RESULTS

- A** The probabilistic safety analysis for a waste repository in a clay formation was completed during the first semester of 1979, and the main results were illustrated in the previous Programme Progress Report¹.

A detailed report of that study has been prepared in a draft form and is now being revised.

- B** The study of long-term risks linked to throwaway and plutonium recycle strategies (mentioned in the previous Programme Progress Report as Ref. 4) has been published with minor modifications as EUR report².

- C** The deterministic section of our methodology is being applied to a specific site, to verify the applicability of that consequence analysis to a real site. The work is being performed in collaboration with the CEN/SCK of Mol, to assess the environmental consequences of an hypothetical radioactivity release from a repository mined into a clay formation. As a first step, the waste inventory is being defined.

- D** Collaboration with the Indirect Action Programme on Risk Analysis of Geological Disposal.

In order to improve contacts between the Risk Analysis Activity of the JRC and similar activities which have been initiated in the frame of the Activity Sheet 7 (Geological Disposal) of the Indirect Action Programme, a «Risk Analysis Subgroup» of the Activity Sheet 7, was formed two years ago. The group meets periodically to exchange ideas and experience, to discuss risk analysis approaches and to promote supporting experimental research. The JRC keeps the secretariat of the group.

In a meeting held in May 1979, it was decided to promote further contacts between scientists of the various disciplines contributing to Risk Assessments to improve the risk analysis data base. The contacts will take the form of joint meetings between people working in various activity sheets of the JRC and Indirect Action Programmes. Risk evaluators will present their models, discuss on the validity of the assumed data base and, possibly, originate the research which is necessary to decrease the uncertainties of the evaluation.

Interactions between specialists is particularly important at the present time when both the JRC and the Indirect Action Programme are preparing the activities of the next multiannual plan.

A meeting was held in November 19-21, to discuss risk modelling and experimental research in relation to geological disposal. Among the points discussed were:

- Waste arising and waste conditioning data assumed in different countries with geologic disposal programmes.

- The present state of development of risk analysis activities.
- The properties of conditioned waste and their modelling.
- The disposal strategies in hard rock, clay and salt formations.
- The geological data base available and the present knowledge of the evolution of the geological environment of waste repositories.

The conclusions of the meeting are presently being analysed with the purpose of incorporating the recommendations of the meeting in the Community R and D programmes.

A meeting of the Group with DG XII Radiation Protection is foreseen in January 1980 to discuss environmental modelling and risk evaluation criteria.

COLLABORATION WITH EXTERNAL ORGANIZATIONS

- Collaboration with the CEN/SCK, Mol for the application of our methodology to a clay formation.
- Collaboration with DG XII Radiation Protection Programme on the interaction of actinides with the biosphere.
- Collaboration with various organizations of the Community in the framework of the Risk Analysis Sub-group.

CONCLUSIONS

Although several problem areas do exist at present affecting the accuracy of the evaluations, it is apparent both from probabilistic and from deterministic studies that the long-term disposal of radioactive waste presents a large margin of safety; therefore it seems possible to conclude that the problems linked with radioactive waste management, although not completely solved, have not the potential to hamper the development of nuclear energy.

PLANNED ACTIVITIES

Concerning the activity in 1980, it has to be remembered that the JRC proposals for the 1980-1983 programme foresee a considerable increase of effort in the field of risk analysis.

In the first half of 1980, on the basis of the Council decision expected for December 1979, the planning of the activity for 1980-1983 will be prepared in a detailed form, taking into account possible activities of the Indirect Action Programme in this area.

At present for the first semester of 1980 the following activities are planned:

- Publication of a detailed description of the probabilistic analysis performed on the clay formation of Boom.
- Development of the consequence analysis for the same formation.
- Increase of the effort for the management of the Risk Analysis Sub-group.

REFERENCES

1. Management of Nuclear Materials and Radioactive Waste Programme Progress Report of the JRC. January-June 1979, No. 3659
2. F. GIRARDI, G. BERTOZZI, G. BIGNOLI: «Comparison of long-term risks of alternative strategies for the nuclear fuel cycle and of different waste categories», Report EUR 6569 (1979)

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 the studies of Waste Hazard Analysis.

The planned activities for the second semester of 1979 were the following:

Radiation damage on glasses

- Irradiation of capsules (BONI III), containing borosilicate glasses, in the HFR reactor at Petten.
- Measurements by means of optical and electron microscopy techniques, of a series of heavy ions irradiated SiO₂ samples with the following aims:
 1. to study the annealing effect at different temperatures
 2. to study the distribution of the damage in the irradiated area after irradiation and annealing (voids, clusters, precipitates).

Leaching tests

- Termination of leaching tests on glasses with water conditioned by clay.
- Continuation of leaching tests on bitumen conditioning different materials.

RESULTS

Irradiation of capsules (BONI III), containing borosilicate glasses, in the HFR reactor at Petten

The results obtained with the capsules BONI I and BONI II have shown the validity of the damage simulation by fission fragments. The highest dose received by the samples was 0.36 dpa corresponding to a damage period for vitrified HAW of about 10⁵ years.

It appeared convenient to use this simulation technique to reach an equivalent time of the order of 1 million years. Previous experiments have shown that a source of error was the temperature distribution inside the cylindrical glass sample. In fact, the glass cylinders ($\varnothing = 10$ mm) with the highest U-235 content (1.76 w%) had a surface temperature of 50°C and a central temperature of about 75°C. In order to reduce the inhomogeneity in temperature, annular samples are used in the new irradiation experiments.

The capsules will be irradiated in the HFR reactor at Petten for about 8 months. Irradiation started in September 1979.

Radiation damage experiments on silica

Further irradiations with heavy ions and electrons have been performed on vitreous silica to separate the displacement effects from those due to ionization. The irradiation conditions and obtained results are illustrated in Table I, which gives the applied dose in terms of particles/cm² and calculated dpa, the optical absorption at the two characteristic B₂ and E'₁ bands and the corresponding computed number of defect centers. It can be immediately observed that while the E'₁ band is present after irradiation with both types of particles, the so called B₂ band is totally absent after electron bombardment, and therefore it can be attributed to direct atomic displacements. Conversely, the comparison between the concentration of E'₁ and that of displaced atoms seem to indicate that this center is not originated by nuclear collisions: its origin must be sought in other processes such as ionization and consequent strained

Table 1. Typical values of defect centres produced by irradiation at various doses

	(Particles/cm ²)	dpa	B ₂		E' ₁	
			(cm ⁻¹)	N (cm ⁻³)	(cm ⁻¹)	N (cm ⁻³)
Heavy ions (Ni ⁶⁰ 46.5 MeV)	x 10 ¹⁵		x 10 ²	x 10 ¹⁸	x 10 ²	x 10 ¹⁹
	.42	0.05	6.1	6.8	3.8	1.7
	.85	0.1	5.5	5.9	3.3	1.5
	4.2	.5	6.0	6.7	3.8	1.7
	8.5	1.	4.6	5.2	2.6	1.2
Electrons	x 10 ¹⁶	x 10 ⁻⁶			x 10	x 10 ¹⁷
	.55	.25			5	2.3
	1.1	.5			7	3.2
	2.5	1.15			9	4.1
	5.0	2.3			1.1	5.0
	8.0	3.7			1.2	5.5

bonds, as suggested in some theories^{1,2}. The effects of the dose and temperature, during electron bombardment, on the growth of E'₁ have also been studied.

The results of experiments are summarized in Fig. 1 giving the absorption spectra obtained in different irradiation conditions. The intensity of the E'₁ band at 140°C shows saturated values that are much smaller than those obtained for comparable or smaller doses during irradiation at 40°C. At this lower temperature the dose dependence is non-linear with a tendency to saturation. Similar studies on B₂ band, originated by atomic displacements, are in progress.

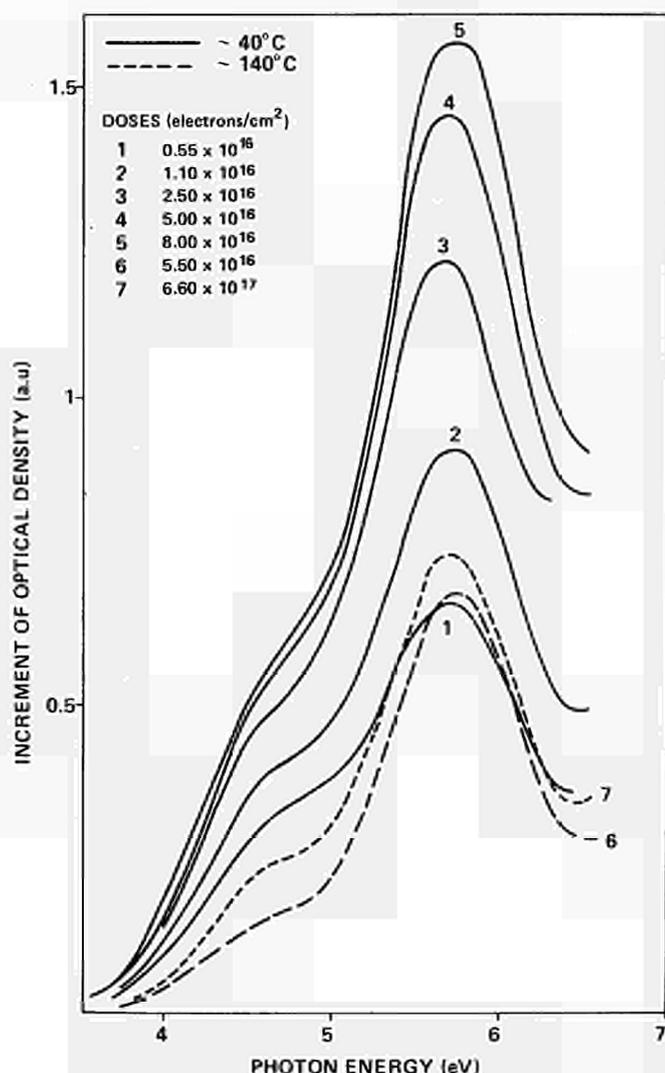


Fig. 1. Change in optical density after 1.5 MeV electron irradiation of SiO₂ at different temperatures and doses

Leaching Tests on Glasses

In order to test glasses in water typical of a clay repository the extractor previously used for leaching tests with pure water was modified. The condensed water enters a vessel containing a clay suspension under agitation. Through a filter the water enters then into the vessel where the glass is located. The clay is changed every 2 weeks in order to have a constant water composition.

The test performed at 80°C for 10,000 h is terminated. The test at 50°C will be finished at the beginning of December. The analyses of the surface gel layer are in progress.

In order to evaluate the effects of cracks which may be produced in the glass blocks during the initial cooling, glass samples were water quenched from the temperature of 550°C. After quenching of the samples a pattern of very small cracks was observed at the microscope on the surface of the samples.

The quenched samples were subjected to leaching tests up to 10,000 h with pure water. The results of the tests are compared in Fig. 2 with the results previously obtained on non-cracked samples.

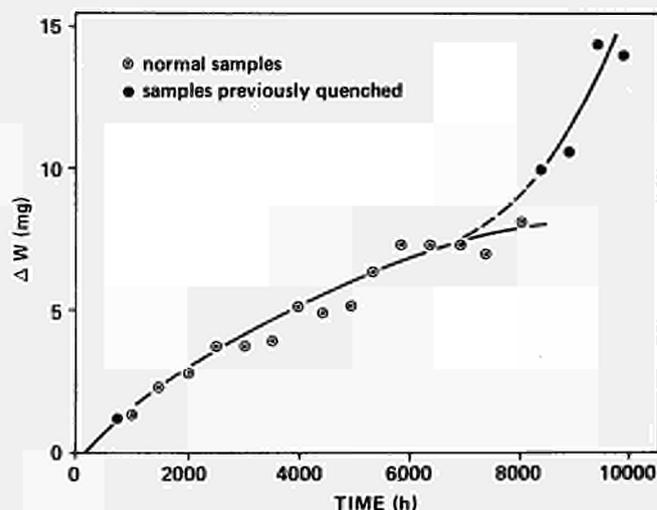


Fig. 2. Leaching tests at 50°C in pure water

From the plot it appears that after about 8,000 h the cracked samples begin to lose weight more and more rapidly.

This can be explained by the fact that the access of water to the new surfaces, produced by quenching, takes a certain time. At the beginning the corrosion of the internal surface is dominated by water diffusion and only after a certain time the surfaces are opened up and corrosion increases abruptly. It is possible also that the acceleration is due to the different conditions of pH existing in the cracks due to preferential leaching of Na.

Leaching Tests on Bitumen

Leaching tests have been performed for about three months on bitumen loaded with 20 w% Fe₂O₃ and CeO₂. The analysis of the liquid failed to show a measurable amount of Fe or Ce in the solution.

Weight determinations show a negligible variation for the CeO₂ loaded samples; this is to be expected due to the high insolubility of this oxide. In the case of Fe₂O₃ loaded samples a leaching rate of the order of 10⁻⁵ g/cm²·d has been detected after 60 days.

Considering also the results previously obtained it seems difficult to define a leaching model of bitumen which is valid for all the possible materials. Water diffuses rather freely in the bitumen following a classical Fick law. If the compound

which is dispersed in the bitumen has a high solubility and a diffusion lower than that of the water, around each grain drops of water are formed which can cause an extensive swelling. As a consequence the interface bitumen-water increases and also the diffusion coefficient increases. It does not seem possible to make a simple model of such a process.

In the case of low solubility or high diffusion no swelling occurs. It is possible then to set up a diffusion model. If we assume that the place of the dissolved grains is occupied by water, the model has to take into account that the water front is displaced as a function of time within the bitumen.

Due to the fact that the stability of the bitumen as a conditioning system is strictly linked to the properties of the waste, it seems necessary to consider more deeply the problem of pretreatment of the waste before the conditioning operation.

COLLABORATION WITH EXTERNAL ORGANIZATIONS

Collaboration with AERE, Harwell, concerning irradiation experiments and theoretical investigations.

Collaboration with the University of Modena concerning examination by SEM (scanning electron microscope) of heavy ion irradiated samples.

Collaboration with CEA, Marcoule, on the leaching of glasses loaded with plutonium.

CONCLUSIONS AND PLANNED ACTIVITIES

Radiation damage on glasses

The investigations on radiation damage are being conducted by means of tests on borosilicate glasses and of more fundamental studies on silica.

In general the planning of the activity has been respected; however, the study of the distribution of the damage in the irradiated areas, planned for the reporting period, has been postponed in order to provide the suitable experimental tool (i.e. H.V. electron microscopy) to observe microstructural changes.

A contract with the Metallurgy Division of AERE, Harwell, for observation by H.V. microscopy is in preparation.

For the first semester of 1980 the following activities are planned:

- Completion of the irradiation of the borosilicate glasses in the HFR, Petten.
- Radiation damage on silica
 1. Studies of annealing effects will be pursued by means of optical techniques
 2. Preliminary assessments for observation of microstructural changes at the H.V. microscope will be performed. The observations may be completed by small angle X-ray scattering and structural measurements of X-ray diffraction in order to demonstrate eventual variation of density and short range order after heavy ions irradiation.

Leaching tests

For the first semester of 1980 the following activities are planned:

- Completion of the analysis of the results on glass leaching in water conditioned by clay
- Continuation of the tests on bitumen
- Leaching tests on borosilicate glasses loaded with Pu (provided by CEA, Marcoule) to verify a possible enrichment of Pu in the gel layer.
- Definition of a leaching test in a system with a restricted amount of water. Probably the case of a glass sample immersed in clay will be taken into consideration.

REFERENCES

- 1 A.R. RUFFA, Phys. Rev. Lett. 25 (1970) 650
- 2 G.M. GREAVES, Proc. Int. Conf. «The Physics of SiO₂ and its interfaces», Ed. S.T. Pantelides, Pergamon Press, 1978, p. 268

Interaction of Actinides with the Environment

OBJECTIVES

The study aims at the development of experimental studies on the behaviour of transuranic nuclides in the environment in order to obtain a better understanding of their interaction with geological media and aquatic systems following an eventual release from a waste repository.

For the reporting period the planned activities were the following:

- continuation of column migration experiments with Pu-238, Am-241 and Np-237
- theoretical modeling study development
- investigation on the development of analytical techniques and valence state evolution of transuranic nuclides.

RESULTS

Experimental studies to investigate the migration behaviour of transuranic elements leached from a glass matrix, under conditions similar to those of natural groundwaters, are being developed in our laboratories. We assume that, as a consequence of a postulated accidental event, water penetrates the repository and partially dissolves the actinide bearing glass, moving afterwards through a surrounding porous formation.

In order to simulate the expected conditions of glass leaching and underground transport in the laboratory, a water pathway is established which flows over the actinide bearing glass and then through columns containing glauconite sand samples. Water fractions are continuously collected and analysed at the column outlet; the water velocity through the columns corresponds to a groundwater movement of about 25 m/yr. The experiments are carried out over periods of 1 to 6 months, and at the end the columns are cut into thin sections and the distribution profile of transuranic isotopes measured.

In Table I results are given on the percentage distribution of actinides leached from contaminated glass. The partitioning was obtained in an experiment over a six month period. As the operating conditions were identical for all three isotopes, the quantity retained by the filters inserted before the soil columns does not appear to depend upon the glass but on the characteristics of the leached radioelements.

Table I. Percentage distribution of actinides after six months of percolation

	10 μm filter	Soil column	Column outlet
²³⁷ Np	1.3	96.4	2.3
²³⁸ Pu	25.3	74.67	0.03
²⁴¹ Am	60	39.96	0.04

The concentrations in the water directly after contact with the alpha-bearing glasses, for americium and plutonium are 10^{-9} M and for neptunium 3.7×10^{-8} M. At the outlet of the soil columns the concentration continually maintained a constant value of 2×10^{-13} M for americium and plutonium and 8×10^{-10} M for neptunium. Two ion exchange resin columns (BioRad AG 50Wx8 and AG 1, 200-400 mesh) were placed in series with the soil column. It was found that all the plutonium and americium released from the soil columns were absorbed on the anion resins.

With the aim of determining the main processes which affect the distribution of activity, the leaching solution was passed through an analytical train made up of a membrane filter, two ion exchange columns and a fraction collector for neutral species.

In Table II are given the results of the distribution of plutonium and americium. These are based on the average of three experiments, each carried out over a 34 day period. Results are given for two cases where the cation and anion resins were inverted in order to determine the eventual filtration effects of the uncharged species on the first column. The soluble charged fraction is considered to be that found on the second column; 34.7% and 0.3% are negatively and positively charged respectively for plutonium. The percentage of uncharged actinide is obtained by subtracting the soluble fraction from the total adsorbed on the corresponding initial column. The data show in fact that between 48-52% of plutonium particles seems to be uncharged and fixed by filtration. The largest «soluble» fraction, however, behaves as an anionic species and only a negligible cationic fraction was detected.

Table II. Percent activity repartition in the analytical train

	Plutonium	Americium
Ultrafilter	12.6	89.2
Cation resin	52.4	10.7
Anion resin	34.7	0.1
Neutral fraction	0.3	—
Ultrafilter	15.1	90.7
Anion resin	83.3	6.0
Cation resin	0.3	3.3
Neutral fraction	1.3	—

For americium it can be seen that the largest amounts of activity are retained on the membrane filter. Of the remaining, some 3% of the total appears to be soluble, having a positive charge, and the remaining 6% is retained by filtration of the resin. Negatively soluble charged forms account for less than 0.1% of the total.

These initial results demonstrate that the soil matrix studied effectively retains the transuranic nuclides; however, the more effective processes seem not to be related to the high exchange capacity of clay minerals but rather due to the highly effective filtering ability of sandy clays. Further the data appears to show that both for plutonium and americium cationic species are only present at very low levels and negatively charged species appear, for plutonium at least to be the main mobile form.

For both plutonium and americium a soluble anionic form having a concentration of 10^{-13} M was detected which was not retained by the columns. Under the present experimental conditions, this concentration is about four

orders of magnitude lower than the Maximum Permissible Concentration for drinking water.

Work is underway in order to understand better the phenomena observed in these laboratory experiments and to develop models of underground migration. Nevertheless, these initial results already indicate that theoretical models of underground transport must take into consideration the filtering effect of porous media. This phenomenon in fact appears, at least for transuranic nuclides, to prevail during the dispersion of solutions leached from α -bearing borosilicate glasses.

Parallel to this development a systematic study in order to determine the amount and kinetics of formation of soluble species is underway. A combination of techniques, as reported in the previous Programme Progress Report (Jan. - June 1979), is being adopted at controlled environmental conditions of pH and Eh, and the distribution of Am-241, Np-237 and Pu-237 isotopes between particulate, cationic, anionic and neutral fractions is being measured at different concentrations of carbonate ions in solution.

COLLABORATION WITH EXTERNAL ORGANIZATIONS

In the framework of the collaboration between the JRC Waste Management Programme and the DG XII Radiation Protection Programme a number of contracts of particular interest for risk evaluation studies has been outlined. These contracts, carried out in various institutes of the Community, are expected to provide the necessary data to validate the environmental models used in long-term waste hazard evaluations.

On the other hand, the JRC's activity on the development of chemical speciation techniques for actinides, can give a basic contribution to studies carried out under contract.

In the framework of this collaboration a graduate student from the Radiation Protection Programme is undertaking at Ispra her doctoral thesis on actinide bioavailability as a function of chemical form in selected fresh water systems.

A meeting with CEA scientists from Cadarache and Fontenay-aux-Roses has been held at Ispra in October with a view to a possible collaboration in the field of actinides speciation and migration through sandy clay columns (artificial or natural barriers).

CONCLUSIONS AND PLANNED ACTIVITIES

The reported results have confirmed the complexity of the studies required for the description of the actinides migration in geological media and aquatic systems.

In the first half of 1980 the following activities are planned:

- filtration experiments with sand columns in parallel with membrane ultrafiltration aiming to identify colloids in the leachate solution
- washing of sandy clay columns after fixation of the leached activity in order to determine the elution behaviour
- continuation of the studies on the physico-chemical behaviour of actinides
- initiation of a study on carbonate complex formation.

Actinides Monitoring

OBJECTIVES

The objective of this study is to develop a methodology for monitoring plutonium contaminated waste streams. The activities are organized in the frame of an Advisory Laboratory.

For the reporting period the planned activities were the following:

A. Integral Experiment

Continuation of the benchmark and calibration measurements. Starting of measurements on real waste.

B. Passive Neutron Assay

Set-up and test of the upgraded signal processor unit. Optimization experiments for the upgraded detector unit. Theoretical investigations of the pulse to pulse time correlation analysis.

C. Passive Gamma Assay

Continuation of the experimental verification of the interpretation model.

D. Active Neutron Assay

Design parameter study of the Reference Monitor

E. Organization of the International Meeting on Monitoring of Pu-Contaminated Waste, JRC Ispra, 25-28 September, 1979.

RESULTS

A. The collaboration programme for an Integral Experiment with the plutonium waste management system of the Dounreay Nuclear Power Development Establishment (DNPDE) has been extended for a new period of twelve months. The final report of the foregoing year is in preparation; the results will be given in the next Programme Progress Report.

During the reporting period a considerable number (180) of synthetic waste items were measured with the NDA4 passive neutron monitor of the Dounreay plutonium waste measurement system. The existing interpretation model will be fitted to this waste problem and will be made available to Dounreay as a computer programme.

B. The development of both the signal processing unit and the detector system of the upgraded Reference Monitor for Passive Neutron Assay has been continued. The signal processing unit is in operation. During the next few months reliability tests will be carried out on the electronic circuits. The design of the detector unit is completed, all the components have been delivered and the unit is under construction in the workshop.

The theoretical investigation of the pulse to pulse time correlation analysis was completed. A computer code was elaborated to calculate the detected multiplicity parameters. Fig. 1 demonstrates such a calculation: the measured time distribution of two successive pulses of the first, second, ... order (V) are transformed into the time distributions of the zero, first, second, ... order coincidences (W). The detected neutron multiplicity parameters are determined by a least square fit of the theoretical predictions to the measured time distributions using these parameters as variables. The calculated time distributions of the coincidences (T) are in good agreement with the measured distributions (W).

The pulse to pulse time correlation analysis, developed by the JRC, constitutes a major improvement in comparison with the systems like the VDC and Shift Register which are presently utilized. These systems have been introduced because of their electronic simplicity and ease of implementation rather than the fact that they represent a good algorithm for analysis.

The advent of microprocessors and low volume bulk storage means that a more sophisticated algorithm can be considered as suitable for operational implementation.

C. During the reporting period the experimental verification of the interpretation model for passive gamma assay has been continued. In order to confirm the validity of the radiation transport model, over the whole γ -ray energy range and waste volume, experiments were performed with point sources, simulating a ring source located inside a polyethylene cylinder.

The theoretical predictions of the radiation transport model defined by the equation

$$P \sim \int_V (1/4 \pi d^2) \exp(-\mu \rho l) S(r, \phi, z) dv$$

was evaluated numerically with the MERCURE-3 code. Calculations were performed with appropriate μ and ρ values and compared with experimental results.

The measured and calculated data are in good agreement with each other, thus indicating that the radiation transport model describes correctly the attenuated γ -radiation spectrum detected experimentally.

The planned continuation of this work will involve the elaboration of a mini-computer based program to evaluate numerically the above equation.

The work will be extended to take into account different corrections such as self-shielding, inhomogeneous waste matrices and the use of different γ -ray lines.

D. The layout of the Reference Monitor considered for active neutron assay is given in Fig. 2.

The Sb-Be neutron source (Thick Target) generates a Sub-MeV neutron interrogating flux in the sample. The flux distribution around the sample channel is measured by monitors. Fission neutron leakage from the Pb-pile is measured by ^4He -counters. The sample is rotated and moved along the channel (scanning). The interaction of the experimental results is based on heterogeneous diffusion theory (Nordheim-method). The apparatus is designed to measure prompt fission neutrons induced by photo-neutrons from an antimony-beryllium source. Owing to the energy spectrum of the (Sb-Be)-source neutrons primary induced fission occurs only with fissile isotopes such as U-235, Pu-239, Pu-241. This allows good discrimination of fertile materials such as U-238. Distinction between fissile plutonium and fissile uranium can be investigated measuring the delayed neutron fraction.

The actual and future problems of the Active Neutron Reference Monitor concern the heterogeneous parameters and the verification of the interpretation model.

Some problems exist with the statistics introduced by the Monte-Carlo method. Calculations have been performed using 5,000 neutron histories; this is probably insufficient to draw final conclusions. A new series of calculations will be performed with a greater number of histories. Calculations will also be performed with other two dimensional transport codes so as to reduce the computer time needed which is a limiting factor in the Monte-Carlo method. Another series of calculations will be devoted to the assessment of the effects of axial leaks on the radial flux gradient at the surface of the sample channel, that is on the mean flux ϕ_2 in the sample.

EXPERIMENT	0.0000400	0.0000050	1	16									
CTOT MF-T1	2627.072	1024.320											
V	1	0.0	53.178	49.312	45.958	43.390	40.912	38.912	37.110				
		35.052	34.260	32.699	31.871	30.688	29.711	28.930	27.952				
V	2	0.0	0.865	2.238	3.228	4.142	4.862	5.474	5.858				
		6.367	6.593	6.976	7.136	7.317	7.578	7.602	7.747				
V	3	0.0	0.009	0.082	0.164	0.293	0.392	0.525	0.614				
		0.774	0.864	0.917	1.064	1.176	1.300	1.374	1.466				
V	4	0.0	0.0	0.0	0.009	0.012	0.026	0.046	0.059				
		0.084	0.101	0.120	0.137	0.149	0.164	0.216	0.223				
V	5	0.0	0.0	0.0	0.0	0.001	0.0	0.002	0.002				
		0.009	0.008	0.012	0.012	0.021	0.025	0.033	0.040				
V	6	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0				
		0.0	0.0	0.001	0.0	0.001	0.001	0.005	0.005				
W	0		2573.894	2524.582	2478.624	2435.234	2394.322	2355.410	2318.300				
		2283.248	2248.989	2216.290	2184.419	2153.731	2124.020	2095.090	2067.138				
W	1		52.313	99.387	142.117	181.365	217.415	250.853	282.105				
		310.790	338.457	364.179	388.914	412.285	434.418	455.746	475.951				
W	2		0.856	3.012	6.076	9.925	14.395	19.344	24.588				
		30.181	35.910	41.969	48.041	54.182	60.460	66.688	72.969				
W	3		0.009	0.091	0.246	0.527	0.893	1.372	1.927				
		2.617	3.380	4.177	5.104	6.131	7.267	8.425	9.668				
W	4		0.0	0.0	0.009	0.020	0.046	0.090	0.147				
		0.222	0.315	0.423	0.548	0.676	0.815	0.998	1.181				
W	5		0.0	0.0	0.0	0.001	0.001	0.003	0.005				
		0.014	0.022	0.033	0.045	0.065	0.089	0.117	0.152				
MULTIP-PAR	0	5	1000.000	100.000	10.000	1.000	0.100						
MULTIP-PAR	0	5	2128.045	212.805	21.280	2.128	0.213						
MULTIP-PAR	1	5	2308.592	136.126	12.754	1.457	0.335						
MULTIP-PAR	2	5	2311.076	133.277	14.450	1.194	0.266						
MULTIP-PAR	3	5	2311.062	133.286	14.438	1.202	0.264						
MULTIP-PAR	4	5	2311.089	133.281	14.440	1.202	0.264						
MULTIP-PAR	5	5	2311.037	133.288	14.439	1.202	0.264						
MULTIP-PAR	6	5	2311.060	133.286	14.438	1.203	0.264						
		0	1	2	3	4	5						
T	2	2574.049	0	52.177	-6	0.826	-10	0.012	9	0.000	0	0.000	0
T	3	2524.783	1	99.201	-5	2.997	-2	0.084	-7	0.002	15	0.000	0
T	4	2478.676	0	141.940	-4	6.160	10	0.247	0	0.010	1	0.000	0
T	5	2435.372	0	181.083	-6	10.063	13	0.513	-6	0.025	10	0.001	0
T	6	2394.394	0	217.168	-5	14.527	11	0.885	-2	0.052	8	0.003	10
T	7	2355.517	0	250.637	-4	19.425	5	1.361	-2	0.092	2	0.006	10
T	8	2318.418	0	281.841	-5	24.661	4	1.938	2	0.147	0	0.010	16
T	9	2282.954	-1	311.076	5	30.167	0	2.609	-1	0.218	-2	0.016	5
T	10	2248.893	0	338.578	2	35.892	0	3.372	-1	0.304	-6	0.025	6
T	11	2216.069	-1	364.540	6	41.797	-8	4.220	6	0.408	-7	0.036	5
T	12	2184.322	0	389.121	3	47.853	-8	5.151	6	0.528	-8	0.050	7
T	13	2153.604	0	412.472	2	54.038	-6	6.160	3	0.666	-3	0.067	2
T	14	2123.823	-1	434.712	4	60.336	-5	7.246	-2	0.822	2	0.087	-2
T	15	2094.843	-1	455.929	2	66.732	1	8.406	-2	0.995	0	0.110	-6
T	16	2066.654	-3	476.222	3	73.218	9	9.637	-3	1.187	1	0.137	-13

Fig. 1. Numerical example of the pulse to pulse time correlation analysis. The experiment is simulated by the Monte Carlo pulse train generator, (2500 SF/sec with $\bar{v} = 2.18$; 10000 (α -n) neutrons/sec; the multiplication parameter $k = 0.03$; one fissionable isotope with $\bar{v} = 2.9$ and a detector efficiency of $\epsilon = 15.2\%$ ($1/\lambda = 40 \mu\text{sec}$; $\tau_d = 5 \mu\text{sec}$; $\Delta t = 5 \mu\text{sec}$; $L = 15$)

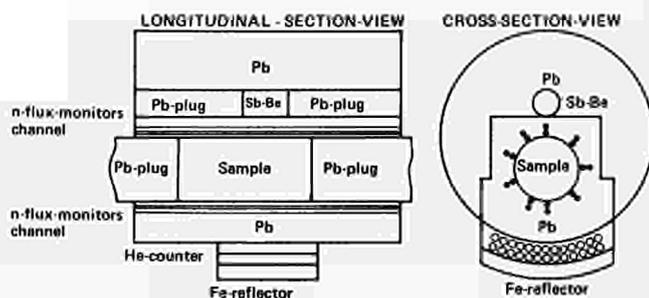


Fig. 2. Lay-out of a reference monitor for active neutron assay

The construction of the apparatus and the experimental part will be considered after the end of the first phase of calculations thus permitting the assessment, within approximately one year, of the feasibility and the sensitivity of this system.

E. Organization of the International Meeting on Monitoring of Pu-Contaminated Waste.

The meeting was held at Ispra on 25-28 September 1979, with the participation of 130 people from both within the Community and outside. 38 papers were presented during 5 sessions:

- General Session
- Destructive Analysis and Reference Materials

- Instruments and Methods
- Theoretical Studies and Nuclear Data
- Operational Experience.

Four working groups were organized in order to draw conclusions. The findings of the working groups were presented and discussed in the final plenary session.

The papers presented and the conclusions of the working groups are being published in the proceedings of the meeting.

Five papers prepared by the JRC staff and by people working under JRC contracts were presented at the meeting¹⁻⁵.

COLLABORATION WITH EXTERNAL ORGANIZATIONS

- Collaboration contract with the Dounreay Nuclear Power Development Establishment (DNPDE) on an Integral Experiment.
- Study contract with the Institut de Physique Nucléaire (University of Lyon) for design parameter study of the Reference Monitor for Active Neutron Assay.
- Collaboration contract in preparation with the Plutonium Fabrication Plant ALKEM (Hanau, F.R.G.) concerning test measurements of waste with equipment development at the JRC.
- Contacts with AERE-Harwell to develop a passive neutron assay system by means of the pulse to pulse time correlation analysis of the neutron pulse train.
- Contacts with the Plutonium Fabrication Plant CNEN (Casaccia, Italy) for setting-up a monitoring system of Pu contaminated solid waste streams.

CONCLUSIONS AND PLANNED ACTIVITIES

The development of the work has been carried out in agreement with the planning.

Concerning the activity in 1980, it has to be remembered that the JRC proposals for the 1980-83 programme, foresee a considerable increase of effort in the field of actinides monitoring.

In the first half of 1980, based on Council decisions, the planning of the activity for 1980-83 will be prepared in a detailed form.

For the first semester of 1980, the following activities are planned:

- Integral Experiment: continuation of real waste measurements

- Passive Neutron Assay: parametric study of the interpretation model for waste measurements with the EURATOM Time Correlation Analyser
- Active Neutron Assay: design parameter study of the Reference Monitor
- Technical Guide for Design and Analysis of Monitoring Systems⁶ : continuation of the revision work
- Consultation and calibration services in the frame of the EURATOM Advisory Laboratory: in particular, test measurements in the ALKEM plant.

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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 for the beginning of 1980 a major report dealing with a critical evaluation of the feasibility of the chemical separation and nuclear transmutation of actinides.

Chemical Separation of Actinides

OBJECTIVES

The experimental studies on the feasibility of the HAW partitioning by OXAL, HDEHP and TBP processes are being concluded.

For the reporting period it was planned:

- to conclude HDEHP batch-extraction and denitration tests on a fully active laboratory scale
- to conclude batch-countercurrent experiments on fully active HAW by HDEHP as well as on simulated HAW by TBP.
- to conclude engineering evaluation studies
- to start the preparation of the final report on the chemical and engineering feasibility of the HAW partitioning.

RESULTS

Denitration experiments

Fully active denitration experiments under the operating conditions foreseen by the HDEHP process flow-sheet has been concluded. A last denitration experiment was performed on the actual (4000 l/t) HAW solution prepared in Ispra hot cells¹ and stored under acidic conditions (4.6 M HNO₃) during about 11 months. After a suitable clarification step this solution was denitrated by formic acid to a pH of about 2 and then filtered. The denitration precipitate was washed and redissolved by 70% HNO₃. The composition of the various liquid fractions was determined by alpha and gamma spectrometry. Process conditions and results are reported in Table I where the percent distribution values for each element refers to its total amount present in the initial HAW solution after clarification.

Table I. HAW denitration. Percent distribution of actinides and fission products
HAW dilution ~ 4000 l/t HAW storage = 11 months
HNO₃ conc. = 4 M/l HCOOH/HNO₃ molar ratio ~ 2

Solution	Element Pu-238 Pu-239	Am-241 Cm-244	Ce-144	Eu-154	Ru-106	Cs-137
Supernatant (~ 30 ml)	24.4	83.9	82.2	80.9	73.1	84.6
0.5 M HCOOH wash (1)	3.7	9.0	14.6	13.7	12.6	14.5
5M HNO ₃ wash (2)	46.4	5.4	2.9	3.8	12.8	0.8
From dissolution of precipitate	25.5	1.7	0.3	1.6	1.5	0.1

(1) by a single (~ 20 ml) wash step

(2) by four successive (~ 20 ml) wash steps

Table I shows that Am behaves as observed in the preceding denitration tests, while a rather large fraction of the initial Pu amount remains in the denitration precipitate. This unexpected Pu behaviour could depend on the insufficient amount of noble metals acting as catalyst of the formic acid decomposition which in turn would be insufficient to promote an adequate Pu reduction. The lower degrees of the Pu retention in the denitration precipitate, separated after the preceding denitration processes, were in fact attributed to the reduction of Pu to its trivalent state. If during the denitration the Pu reduction is incomplete, Pu IV may in fact hydrolyse and coprecipitate with other hydrolysable ions (Zr, Mo).

Another possible effect could be the presence of phosphate ions generated by the radiolysis of TBP traces. No dodecane wash of the HAW raffinate was in fact performed at the end of Purex process operations so that non negligible traces of TBP, mechanically entrained in the HAW solution, may degrade by radiolysis up to phosphoric acid.

The precipitation of Zr phosphates and of hydrous zirconia could also explain the high Pu retention (18%) measured in the precipitate separated from the HAW solution after 9 months storage under acidic conditions. This possibility seems confirmed by the presence in the same precipitate of about 18% of the initial amount of Zr. More difficult to explain is the presence of a large portion of Pd (24%) that could, however, support the hypothesis above reported

(insufficient noble metal content) to explain the large fraction of Pu (25%) adsorbed on the denitration precipitate.

On the basis of the above results it is evident that to minimize the presence of phosphate the HAW raffinate must be washed by the organic diluent just after being produced. As it is likely that also the ageing of the HAW raffinate could promote the formation of precipitate the storage of the HAW solutions has to be avoided as much as possible. The organic wash of the HAW solution is a plant operation normally accomplished without difficulties. On the contrary even if the HDEHP process does not include any storage step («direct partitioning») occasional HAW storage periods cannot be «a priori» excluded.

According to the TBP process flow-sheet (see solvent extraction experiments) a partial HAW denitration to 0.5-1.0 M HNO₃ has to be carried out. Partial denitration experiments were performed using a synthetic concentrated (~ 13-fold) HAW solution containing phosphate-ions. The obtained results indicate that actinide content increases in the precipitate as the storage time of the concentrated HAW increases. After not more than three weeks of storage in fact the Pu content in the denitration precipitate increased from 1 to about 3% of its initial amount.

Also in this case the HAW storage in presence of phosphates appears to be an important factor in determining the actinide retention in the overall precipitate produced by the HAW concentration, storage and denitration steps.

Solvent extraction experiments

HDEHP Process

Batch extraction experiments on fully active laboratory scale carried out under acidic (4 M HNO₃) and low acidity (pH 2) conditions have been described in the preceding progress report². The set of experiments relating to the HDEHP process has been concluded by testing on fully active laboratory scale the back-extraction of trivalent actinides from 0.3 M HDEHP - 0.2 M TBP in n-dodecane. The solvent was previously loaded by extracting trivalent actinides, Pu and RE from a 3500 l/t fully active HAW solution at pH ~ 2. The measured percent back-extraction yields are reported in Table II along with those obtained under similar conditions using a simulated 5000 l/t.

Table II. Selective back-extraction of trivalent actinides from HDEHP loaded with synthetic and fully active HAW

Element	Solvent	Strip Solut.	% Back-Extraction	
			Synthetic HAW	Fully Active HAW
Pu	0.3 M HDEHP 0.2 M TBP in n-dodecane	0.05 M DTPA 1M glycolic acid pH~ 3 O/A = 1	29	20
Am			97	96
Eu			38	40
Ce			20	23

From the comparison of the results obtained from the two batch-experiments (Table II) it appears that the Am back-extraction efficiency does not show significant variations. Nearly the same percent back-extraction yields were obtained for Am using synthetic and fully active HAW solutions.

HDEHP Radiolysis

The efficiency of the HDEHP back-extraction could be negatively affected by the actinide and RE radiation effect on the solvent.

In order to determine the extent of this effect a fraction of HDEHP solvent (0.3 M HDEHP - 0.2 TBP in n-dodecane) containing actinides and RE previously extracted from a fully active HAW solution (3500 l/t) was stored during 5 months in a sealed tube (self-irradiation test). No formation of precipitates being observed at the end of the storage time, the loaded HDEHP was successively volume to volume back-extracted with the following aqueous solutions:

1. 0.05 M DTPA + 1 M glycolic acid at pH 3 for the selective stripping of Am and Cm
2. 5M HNO₃ for the selective stripping of RE
3. 0.08 M H₂C₂O₄ for the selective stripping of Pu and
4. 1 M Na₂CO₃ at 60° for the solvent clean up (U)

The distribution coefficients (D_A⁰) measured for the various elements seem to indicate a certain reduction of the back-extraction efficiency probably due to the self-irradiation effect. A factor 3 increase of the D_A⁰ was in fact measured for Am corresponding to a decrease of the Am percent back-extraction yield from 96 to about 88%.

As to the subsequent stripping steps mentioned at points 2 and 3, the back-extraction efficiency of RE and Pu remained practically unchanged. During the alkaline clean-up of the solvent (point 4) carried out at 60°C no emulsion or interfacial precipitate were observed.

After reconditioning to the acid form, the HDEHP concentration was determined by titration with tributylammonium hydroxide and sodium hydroxide. A reduction of about 10% of the initial HDEHP molarity was measured.

The extraction capacity of the regenerated solvent controlled under simulated condition did not show significant variation of the Am and RE distribution coefficients with respect to the expected values.

A set of more intensive irradiation tests were also performed using a Cobalt-60 source at a dose rate of 1 M rad/h. Different types of solution were irradiated at variable radiation exposures, namely:

Pure solvent (0.3 M HDEHP-0.2 M TBP in n-dodecane)

The purpose of this irradiation test was to determine whether radiolysis products having high extractive capacity for trivalent actinide could be produced during the irradiation and could thus prevent a complete actinide back-extraction. According to the results obtained with irradiated (120 Mrads) and unirradiated solvent fractions the above hypothesis can be excluded.

Loaded solvent

This irradiation test was aimed at determining to what extent the Am and RE back-extraction efficiency may be affected by irradiating the loaded HDEHP with an external beta-gamma radiation source.

The obtained results show that the Am and RE retention in the irradiated HDEHP show a certain tendency to increase as the radiation exposure increases. These results are therefore fairly consistent with those obtained by self-irradiation tests.

After a few weeks from the end of the irradiation the formation of a precipitate was however observed in the already irradiated HDEHP fraction, the amount of which was directly proportional to the radiation exposure.

HCOOH and DTPA solutions

These irradiation tests were performed in order to investigate the possible formation of oxalic acid as well as the radiation stability of the DTPA solution in view of its possible recycling.

The obtained results indicate no formation of oxalic acid and an increase of the Am and RE retention in the solvent as the DTPA radiation exposure increases.

TBP Process

TBP batch-countercurrent experiments with the simulated HAW solution have been completed during the present reporting period. The option 1, previously proposed for TBP process³ was slightly modified according to the following process sequence:

1. Partial denitration of 13-fold concentrated HAW and final adjustment to 1 M HNO₃.
2. Selective extraction of U, Pu at 1 M HNO₃ (instead of extracting at 4-5 M HNO₃ before the denitration step as originally proposed)
3. Final acidity adjustment (to ~ 0.2 M HNO₃) and salting by addition of Al³⁺/Al(OH)²⁺ nitrates, NaNO₃ and NaOH in amounts compatible with the vitrification (instead of the direct denitration to 0.2 M HNO₃).
4. TBP extraction and back-extraction of Am and RE.
5. HDEHP selective extraction of RE.

These process step modifications were aimed at achieving the following objectives:

- to attain the reduction of the HAW acidity by means of a procedure which is undoubtedly less critical for the remote acidity control than a specific denitration process.
- to promote favorable conditions for converting to an extractable form the Pu fraction coprecipitated during the HAW concentration and storage. The possibility of achieving this latter objective is however still to be demonstrated on fully active laboratory scale.

A simulated HAW solution, concentrated by a factor 13, partially denitrated and then adjusted to 1 M HNO₃ was therefore used as initial feed. The operating conditions applied for the batch-countercurrent extraction and back-extraction steps are summarized in Table III.

Typical percent separation yields are reported in Table IV for Pu, Am, Eu, Ce and some FP. The obtained results are consistent with the results of batch experiments reported for Am in a preceding progress report⁴. They can be summarized as follows:

- Pu, Am, Eu and Ce behave as expected in the various TBP extraction and back-extraction steps. The actinide separation from the concentrated HAW is obtained with DF higher than 10³ for Pu and 10⁴ for Am.
- The separation Ce and Eu from actinides is also satisfactory; DF of about 2000 and 133 for Ce and Eu being measured respectively after their selective extraction by HDEHP.

Therefore this experiment leads to conclude that the TBP process is suitable to remove actinides from a concentrated synthetic HAW solution with quite satisfactory actinide separation efficiencies. Obviously the validity of this statement is presently still restricted to the employment of simulated conditions and needs to be demonstrated also for actual HAW solutions.

OXAL process experiments

They have been completed with the actinides/RE partitioning experiments on fully active laboratory scale illustrated in the preceding progress report².

Process Engineering Assessment

The stage has been reached concerning the engineering assessment of actinide separation whereby the laboratory data collection is correlated and assessed in practical nuclear chemical engineering terms in order to establish the feasibility of a flow-sheet for the subsequent stage of a preliminary design.

Previous PPR's have indicated some significant factors concerning the proposed flow-sheets submitted for feasibility assessments. Generally these factors have been based on the following considerations:

- I. solvent choice
- II. HAW-concentrated or original
- III. «Direct» or «Delayed» partitioning
- IV. Proven nuclear fuel reprocessing technology
- V. The overall waste balance must be of the same order or at the most only fractionally greater than that for a comparable vitrification step alone
- VI. Precipitates - solid formation
- VII. Decontamination factors achieved, compared to requirements
- VIII. Process safety - criticality, chemical and control
- IX. Corrosion
- X. Cost estimate considerations

The above list is by no means exhaustive but it does illustrate simply the magnitude of the problem. It was foreseen that the actual conception and possible realization of a HAW partitioning plant would present a formidable task because of the function the plant is required to do in terms of performance and decontamination factors (DF's of the order 10³, 10⁴ compared to 200 for normal PUREX type reprocessing).

In short the conception is of a highly advanced technology industry requiring very small limits of operational variation for successful «production» of α -free waste streams.

Integration with the PUREX reprocessing facility has always been considered an essential requisite and so compatibility has been a conditional criterion on which to base the processing sequence.

The necessary criteria originally envisaged of, 5 year cooled HAW produced from LWR fuel, must therefore be considered in a much broader context in terms of

1. storage time out of reactor prior to reprocessing
2. reprocessing itself
3. HAW treatment after reprocessing - «direct» or «delayed» which is dependent on (1) above
4. activity levels in the extraction and stripping sections.

The considerations of these factors all influence in some way the choice of one of the proposed flow-sheets. For example the TBP solvent instead of the HDEHP solvent would be the preferred choice. However, in order to enhance the extraction of Am/Cm/RE's the TBP solvent requires addition of salting agents (nitrates of aluminium). Further the quantity of salting agent required would be considerable and totally unacceptable from the point of view of solidified waste arisings if the unconcentrated HAW was considered. Consequently the TBP process can only be applied to the concentrated HAW since the salting agent requirements are then of an order of 10-15 less in quantity and at the same time amenable to the vitrification process.

Concentration of the HAW which is already a routine plant operation evidently means a large increase in the activity level per unit volume of the HAW. This could present serious problems particularly from Zr-Nb, Ru-Rh in high acidity exhaustive extraction steps and from the RE's mainly Ce-Pr 144, in the low acidity separation step. It is already known

Table III. Operating conditions applied to the various process steps of the TBP flow-sheet

Process steps L. stream constituents	U - Pu Select. extraction			Am, Eu, Ce Extraction			Am, Eu, Ce Stripping			Eu, Ce Select. extraction		
	Compos.	Stage No.	Relat.* flow	Compos.	Stage No.	Relat.* flow	Compos.	Stage No.	Relat.* flow	Compos.	Stage No.	Relat.* flow
TBP/n-dodecane	30%	10	770	30%	8	1800		3	1800			
FEED : HNO ₃ (M/l) Al ³⁺ (M/l) Na ⁺ (M/l) NO ₃ (M/l)	1	8	385	0.20 0.54 1.08 2.90	8	600						
SCRUB : HNO ₃ (M/l)	1	2	385									
STRIP : HNO ₃ (pH)							1.5	3	900			
HDEHP / n-dodecane TBP										0.3 M 0.2 M	8	4200
FEED : HNO ₃ (pH) DTPA (M/l) Glycolic acid (M/l) Na ⁺ (M/l)										3.0 0.05 1 >0.25	5	3500
SCRUB : DTPA (M/l) Glycolic acid (M/l) pH										0.05 1.0 3.0	3	700

(*) RELATIVE FLOW : litres per metric ton of U

Table IV. TBP batch - counter-current experiments. Percent separation yields of actinides and some FP obtained by operating the various process steps. Process conditions: see Table III

Process steps Element	% Separation yields			
	U, Pu Select. extr.	Am, Eu, Ce Extraction	Am, Eu, Ce Stripping	Eu, Ce Select. extr.
Pu	≥ 99.9	≥		
Am	≤ 0.1	≥ 99.99	≥ 99.97	~ 0.07
Eu	≤ 0.1	≥ 99.99	≥ 99.97	99.25
Ce	≤ 0.1	≥ 99.98	≥ 99.97	≥ 99.95
Ru	~ 13	~ 90.0	~ 26	
Rh	~ 1			
Pd	≤ 5			
Zr	≤ 5			
DF	(Pu) = 10 ³	(Am) ~ 10 ⁴		(Eu) = 133 (Ce) ≥ 2000

from the PUREX process that the solvent TBP does suffer radiation damage forming various compounds which retain U, Pu and FP's due to the low aqueous solubility of the complexes formed; precipitation of insoluble compounds can form a «fixed» radiation source in the contactor and will deteriorate anyway the hydrodynamic performance of the process. These effects would be amplified further in the higher radiation fields of concentrated HAW (depending on the time of storage). Thus a necessary criterion is to follow the concentration step with storage of the HAW in order to allow the activity level to decay. The TBP process therefore offers itself for the «delayed partitioning» of the actinides. The solvent in this case will be subjected in the extraction of Am/Cm/RE's to the order of 100 Ci/l whereas the HDEHP solvent as proposed for «direct» partitioning would be subjected to 3-4 times this value. Further since the RE's are retained in HDEHP during the stripping of Am, Cm, then the contact time of solvent with these levels of activity will be far greater. This needs to be compared to activity levels at which the solvent is known to maintain its required properties. The criterion to be established here therefore is the optimum time of storage prior to treatment of the HAW together with the terms of acceptable radiation dose to be received by the solvent.

The conceptual design would be fairly incomplete without some form of cost estimate. Such a cost estimate signifies the additional cost for the integration of a similar technology, i.e. the actinide separation plant, to a reference nuclear reprocessing facility operating on the PUREX cycle. This approach helps to offset the difficulty of retrieval of information regarding individual costs of unit operations. Thus a relative cost index with respect to the PUREX cycle is achieved together with the requirement of already existing technology and, as mentioned previously, compatibility of the processes.

In summary the process schemes are being analysed and examined in relationship to the considerations of I. - X. and 1.-4. above. The results and implications are being assessed qualitatively and as far as practicable quantitatively in order to formulate a final overall assessment.

Assessment studies will be completed for the end of 1979.

Preparation of the final report

The preparation of the final report concerning the feasibility study on the chemical separation and nuclear transmutation of actinides has been initiated. The subject of the different chapters relating to the chemical separation section have been already discussed and provisionally identified.

COLLABORATION WITH EXTERNAL ORGANIZATIONS

Collaboration contract between JRC and CEA (CEN, Fontenay-aux-Roses, France) concerning the verification on fully active laboratory scale of the proposed HDEHP and TBP flow-sheets (continuous countercurrent mixer-settler battery experiments).

CONCLUSIONS AND PLANNED ACTIVITIES

Most of the project activities have been accomplished according to the planning.

During the present reporting period the following experimental activities have been in fact concluded:

- denitration experiments on the fully active HAW (unconcentrated)
- HDEHP batch extraction experiments on the fully active laboratory scale

- radiation damage tests on HDEHP solvent
- TBP batch-countercurrent experiments with a simulated HAW solution

However, some experiments on fully active HAW are presently still not ended. Thus was due to unexpected difficulties causing a delay in obtaining the operating licence for the new two cells. As consequence it was not possible to accomplish in these cells the denitration of concentrated HAW and the HDEHP batch countercurrent experiments as it was originally planned.

During the next six months it is planned :

- to conclude the preparation of the final report on the chemical and engineering feasibility of the HAW partitioning. The contents of this report will be illustrated at the 2nd Technical Meeting on the Nuclear Transmutation of Actinides to be held in Ispra on April 21-24, 1980
- to complete, if necessary, the set of fully active experiments originally planned at Ispra. The decision on whether this will be still convenient and fruitful will be taken on the basis of results of fully active experiments to be performed under the collaboration contract with CEA.
- to prepare the planning of the activity for 1980-1983, on the basis of the Council decision. Studies for the separation of plutonium from medium activity liquid wastes and solid waste are planned.

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Assessment Studies on Nuclear Transmutation of Actinides

OBJECTIVES

The aim of this activity is to evaluate the neutron-physical and technological feasibility of the transmutation of actinides other than fuel in fission power reactors and to propose a suitable recycle strategy.

For the reporting period activities in the following areas were planned:

- Generation of data on formation and transmutation rates of by-products actinides in FBR, LWR and HTR reactors
- Design of fuel elements for FBR and LWR reactors
- Parametric evaluation of additional fuel cycle costs due to by-product actinide recycling
- Risk assessment for by-product actinide recycling
- Evaluation of the technical implications on the fuel cycle
- Elaboration of a recycle strategy
- Preparation of the final report on the nuclear transmutation of actinides.

RESULTS

Reactor Physics Calculations for LWRs

Reactor physics calculations for the assessment of actinide transmutation in LWRs have been continued with the use of the reactor physics zero-dimension code RIBOT-5A and the two-dimension code EREBUS.

Homogeneous recycling

A typical BWR has been studied with respect to two basic recycling schemes.

The first scheme involved the homogeneous recycling of all self-generated by-product actinides in a U-Pu fuelled BWR. The chosen fuel composition corresponded to a loading of 1/3 of the fuel elements charged with 5% of equilibrium recycled plutonium (fuel plutonium) with the following composition:

Pu-238:	2.9%
Pu-239:	45.5%
Pu-240:	26.5%
Pu-241:	15.8%
Pu-242:	9.3%

The fuel plutonium was maintained constant at the beginning of each cycle.

However, in the case of Pu-238 the total quantity was composed of fuel Pu-238 and recycled actinide Pu-238. It should be noted that U-236 was not recycled.

The U-235 beginning of cycle (B.O.C.) content was determined such as to ensure an end of cycle K_{eff} practically equal to 1.

Improvements with respect to the recycling scheme already adopted¹ have been carried out. All by-product actinides (including Cm-242) have been recycled applying decay times between cycles of 0, 1, 5 and, for completeness, 100 years. Fig. 1 shows the U-235 B.O.C. enrichment for different decay times and different cycles.

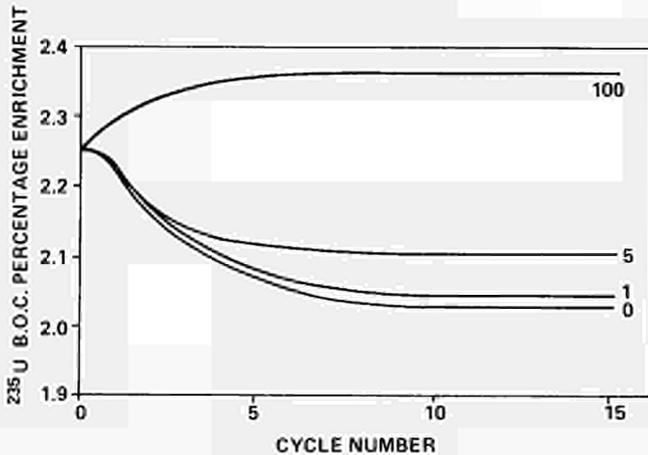


Fig. 1. Beginning of cycle percentage ^{235}U enrichment for the U-Pu BWR with different decay times (years)

The second scheme involved the homogeneous recycling of all self-generated by-product actinides in a U fuelled BWR. In this case all the Pu-isotopes and U-236 were recycled. The U-235 B.O.C. enrichment was adjusted as described above.

Table I outlines some results for both recycle schemes and for cycles 1 and 10. Fig. 2 shows the corresponding U-235 B.O.C. enrichment for both schemes. The results shown in Fig. 2 and in Table I refer to a zero decay time.

The important gain in the first cycle of the U-fuelled BWR is due to the build-up of fresh plutonium (nearly 50% of the equilibrium Pu-239 is formed in the first cycle). The smooth

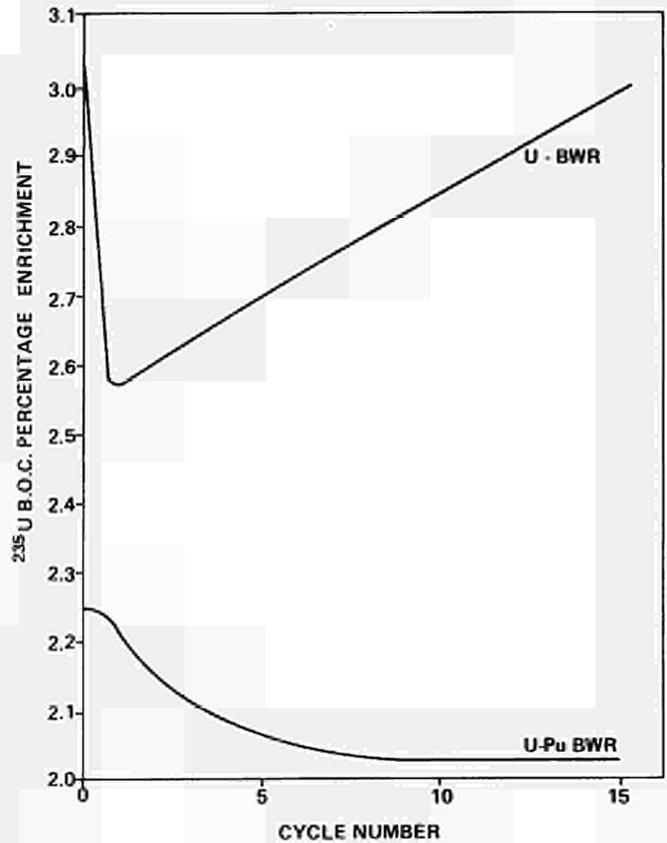


Fig. 2. Beginning of cycle percentage ^{235}U enrichment for U-BWR and U-Pu BWR

rise of the curve thereafter is due to the continuously increasing quantity of U-236 with each recycle. For the U-Pu fuelled BWR an equilibrium U-235 B.O.C. enrichment was obtained at about the 8th cycle.

The positive influence on reactivity, already pointed out¹ for the U-Pu fuelled BWR is here confirmed for this recycle scheme.

Heterogeneous recycling

The improvements of the EREBUS computation, carried out with respect to the last Programme Progress Report¹, are essentially the following:

- the geometrical description of the reactor core pattern is now more complete, taking into account nine fuel bundles instead of one. This permits the insertion of a more complex spatial structure of fuel elements around the target element,
- the cross sections and the nuclear data have been tested and in some cases modified,
- the self-shielding factors of by-product actinides have been evaluated and introduced.

Cell homogenization and fuel isotope cross section collapsing in three groups (one thermal, one epithermal and one fast) have been carried out by means of LASER (a multigroup, one-dimension, burnup, cell code). The cross-section data were used as input in EREBUS.

For the by-product actinide isotopes a cross section set was prepared by adjusting the Benjamin² set according to comparisons performed with RIBOT results.

With this cross section set various calculations have been performed in order to investigate the flux and power perturbation caused by the presence of actinides in the fuel assembly.

Table 1. Build up (kg/THM) of some actinides for two recycling schemes in a BWR

ISOTOPES	U BWR		U - Pu BWR	
	CYCLE 1	CYCLE 10	CYCLE 1	CYCLE 10
^{236}U	$3.78 - 0^*$	$1.86 + 1$	—	—
^{237}Np	$3.70 - 1$	$4.33 - 0$	$2.51 - 1$	$5.08 - 1$
^{238}Pu	$4.50 - 2$	$4.98 - 1$	$6.33 - 2$	$1.13 - 1$
^{239}Pu	$5.51 - 0$	$9.55 - 0$	—	—
^{240}Pu	$2.18 - 0$	$5.35 - 0$	—	—
^{241}Pu	$1.26 - 0$	$3.59 - 0$	—	—
^{242}Pu	$3.66 - 1$	$1.65 - 0$	—	—
^{241}Am	$3.99 - 2$	$2.75 - 1$	$1.52 - 1$	$1.78 - 1$
$^{242\text{m}}\text{Am}$	$6.60 - 4$	$5.23 - 3$	$2.75 - 3$	$3.27 - 3$
^{243}Am	$1.14 - 1$	$1.24 - 0$	$8.96 - 1$	$1.18 - 0$
^{242}Cm	$1.37 - 2$	$8.68 - 2$	$5.56 - 2$	$6.94 - 2$
^{243}Cm	$1.13 - 3$	$1.58 - 2$	$6.71 - 3$	$1.16 - 2$
^{244}Cm	$4.37 - 2$	$3.76 - 0$	$7.08 - 1$	$3.95 - 0$
^{245}Cm	$1.86 - 3$	$3.33 - 1$	$4.50 - 2$	$2.90 - 1$
^{246}Cm	$2.66 - 4$	$7.29 - 1$	$9.37 - 3$	$1.01 - 0$

* $3.78 - 0 = 3.78 \times 10^{-0}$

— indicates isotopes not involved in recycling

The first calculations considered a target fuel rod completely filled ($\rho = 10.5$) with actinides of equilibrium isotopic composition.

The power in the target rod increased by a factor of approximately 5 with respect to the unperturbed regions. In a second case, the actinide charge was reduced by a factor of more than 5 (exactly 5.48) giving a density of approximately 2. No diluent was added.

The third calculation was made with the density reduced again by a factor of 2 ($\rho \sim 1$). A negative power peak factor of nearly 0.85 was found in this case. The same density ($\rho \sim 1$) had been inserted in an annular region (Fig. 3) with the important flux and power depression shown also in Fig. 3.

It may be concluded that heterogeneous recycling in isolated rods or in 'islands' within target fuel assemblies would be a very complex operation.

Further study is necessary, should one want to support heterogeneous recycling, in order to find compromise configurations able to give high actinide burning ratios (i.e. high flux) without strong power peakings and together with reasonable actinide charges.

Solutions which have already been suggested by several authors^{3,4} involve the variation in the enrichment of the rods neighbouring the target rods, or in having water in certain zones of the target assemblies. However, these solutions are somewhat cumbersome and unpractical.

Reactor Physics Calculations for FBRs and HTRs

Reactor physics calculations for FBRs and HTRs are carried out by CNEN, Casaccia and KFA, Jülich under study contracts.

The second progress report⁵ was presented by Casaccia containing a detailed description of the calculation technique to be applied for computing the generation of by-product actinides in the FBR. A block diagram of the

calculation scheme is shown in Fig. 4. The characteristics of a representative fast breeder reactor are summed-up in Ref. 6. Starting from the design criteria of various fast breeders, a notional fast reactor for investigations in the frame of by-product actinide transmutation has been proposed.

KFA Jülich has supplied the 2nd and 3rd progress reports^{7,8}. They contain information concerning the design of a large pebble bed reactor of 3000 MWth, charging and discharging isotope vectors, and power as well as flux distributions for the reactor. In order to make possible corresponding burnup calculations by means of ORIGEN, a set of microscopic cross sections for the heavy metal isotopes has been derived from the RSYST library available at KFA Jülich. Energy-dependent disadvantage factors for the neutron flux for spherical fuel elements vary in a range of plus and minus 5%. This means that there is only a weak spatial flux depression in the fuel which will hardly influence the transmutation rate and the isotope evolution.

Design of fuel elements for FBRs

The calculated internal temperature of 1840°C previously reported¹ for a ring pellet in a fast flux, has been considered to be compatible with the melting point of Np-Am-Cm oxide (2440°C) and the expected temperature ($\sim 2000^\circ\text{C}$) of the eutectic between actinide oxides and magnesium oxide introduced as mechanical support.

On the other hand this configuration implies technological problems arising from:

- fabrication of a ring 1.35 mm thick over 5.85 mm external diameter;
- possible shape deformation in the sintering process;
- accommodation of a MgO kernel pellet.

A possible alternative to avoid these difficulties is the preparation of normal pellets with actinide content for

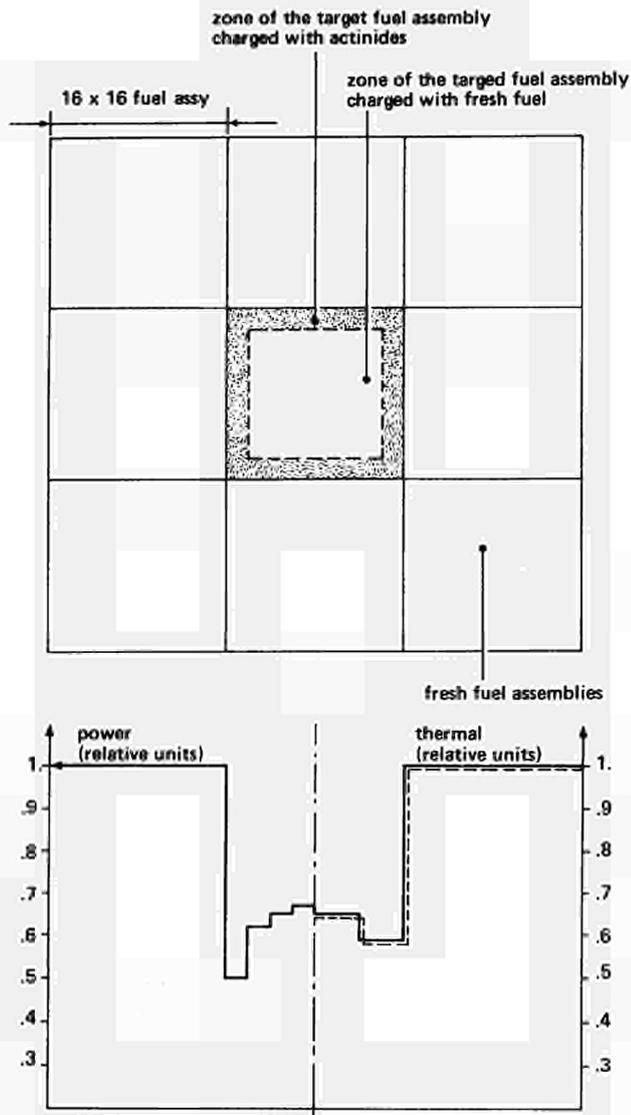


Fig. 3. Power and flux distribution

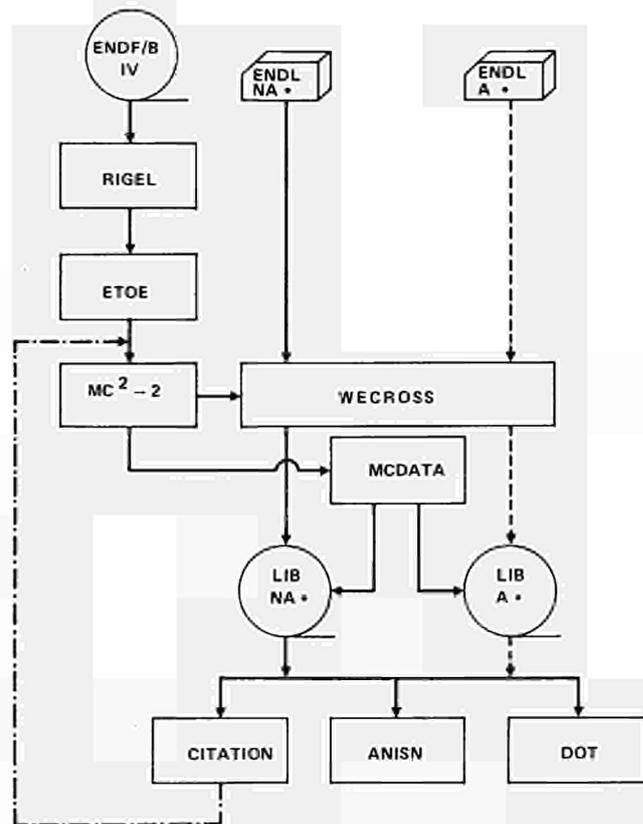


Fig. 4. Block diagram for computing the LMFBR

- * NA refers to a library containing cross sections without self-shielding
- * A refers to a self-shielded library

length unit smaller than in ring pellets. As inert diluent MgO has been considered.

Central temperature (T_c) calculations have been carried out for various mixtures of actinide oxides (Np-Am-Cm or Am-Cm in the ratios given in a previous Programme Progress Report⁹) with MgO. The T_c calculations have been carried out for gaps of 75 and 150 μ . The linear Vegard's law has been applied to calculate the thermal conductivity of the oxide mixtures.

From the curves of the central temperature vs actinide oxide content per length unit reported in Fig. 5, it appears that the temperature of 2000°C is obtained for a mixture of 0.545 g/cm MgO + 1.20 g/cm Am-Cm oxide (linear power rating 290 W/cm) and for a mixture of 0.492 g/cm MgO + 1.33 g/cm Np-Am-Cm oxide (389 W/cm). These data refer to a gap of 75 μ .

Though the use of ring pellets permits operation with higher actinide oxide content for unit length, the use of normal pellets containing MgO as inert diluent appears preferable for the following reasons:

- the fabrication method is considerably simplified;
- the possibility to blend with MgO, Np-Am-Cm mixture as well a Am-Cm mixture, maintaining acceptable T_c values, allows flexibility in the partitioning scheme;

- MgO used as diluent, seems acceptable in the reprocessing plants;
- the decrease of linear power does not create problems in the reactor operations.

Design of fuel elements for LWRs

Orientative temperature profile calculations have shown a little influence on T_c by 5 w% actinide oxide blended in U-5 w% Pu fuel in a representative PWR for homogeneous actinide recycling. The plutonium and actinide compositions correspond to those given in Ref. 1. The results summarized in Table II show that the introduction of actinides in uranium-plutonium MOX fuel for LWR, depresses the central temperature.

The situation is quite different when actinides are irradiated in a fast flux for which the central temperature depends on the linear power rating proportional to the actinide amount.

Risk Assessment

In the field of risk assessment two different studies were initiated.

1. Modification of the MARYON II programme.
2. Long term hazard analysis of LMFBR fuel cycle waste with fission products.

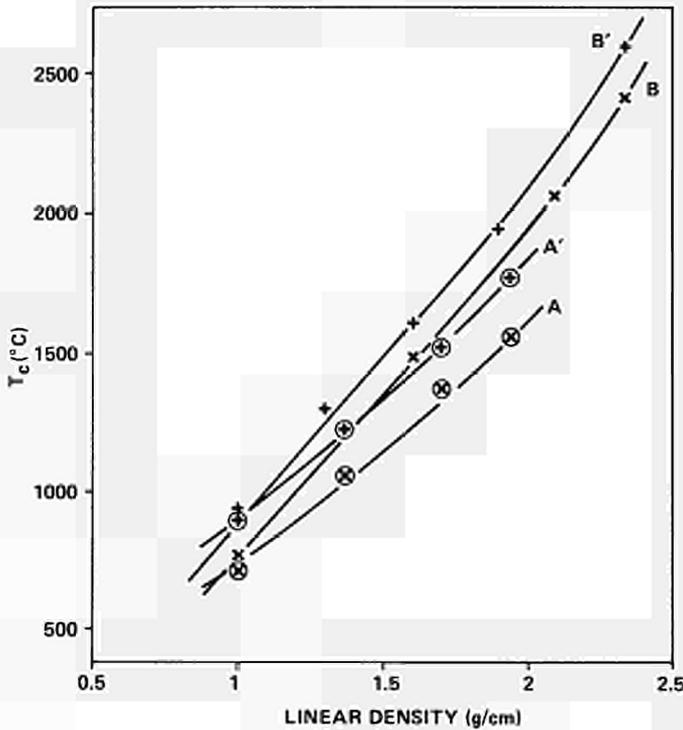


Fig. 5. Central temperature vs actinide oxide linear density for pellets of 5.85 mm diameter in FBR
 Curve A = Np - Am - Cm - Mg oxides with gap 75 μ
 Curve A' = Np - Am - Cm - Mg oxides with gap 150 μ
 Curve B = Am - Cm - Mg oxides with gap 75 μ
 Curve B' = Am - Cm - Mg oxides with gap 150 μ

Table II. Central temperature, T_c , vs fuel composition
 Pellet $\phi = 8.192$ mm
 Linear power = 438 W/cm
 Cladding temperature = 312°C

Fuel composition	Central temperature T_c (°C)
U-2.7W/o ^{235}U	2140
U-5W/o Pu	1450
U-5W/o Pu - 5W/o actinides (Np - Am - Cm)	1400
U-5W/o Pu - 5W/o actinides (Am - Cm)	1390

The modification of the MARYON II programme was performed in order to obtain a larger freedom in the specification of the loss fractions to the waste and the environment. These loss fractions are now defined as rectangular matrices which attribute to each fuel cycle for each isotope a different loss fraction. The losses to the environment are now summarized and are no longer specific to a fuel cycle facility. The waste arisings will be given according to the categories: total waste, high-level waste originating from the high-level liquid waste of the reprocessing plant and the rest of the waste defined as medium and low-level waste. A preliminary description of this code is given in Ref. 10. The code was supposed to be ready by September 1979 but unforeseen delays have occurred during the debugging.

In Ref. 11 the hazard accumulation of the total waste was studied in an LMFBR fuel cycle with and without recycling of by-product actinides including the fission products.

Fig. 6 shows for actinide loss fractions of 10^{-3} till 10^{-4} the ingestion hazard, as function of decay time, of the fission products and of the actinides with and without by-product actinide recycling (nuclear power duration 400 y).

After a decay time of 200 years the contribution of the actinides is the dominant part of the ingestion hazard if the by-product actinides enter completely into the waste. The by-product actinides dominate in these conditions the ingestion hazard and a reduction of the fuel losses to the waste far below 10^{-2} is not of big interest from the waste management point of view.

A recycling of actinides however would require low actinide loss fractions to the waste and in addition recommend a recycling of I-129 and possibly of Tc-99 in the reactor.

Fig. 7 gives the ingestion hazard waste ratio for a loss fraction of actinides of 10^{-3} . With no recycling of I-129 and Tc-99 the waste hazard is only reduced by by-product actinide recycling to about 30% or 40% after a waste decay of 10^4 years. With a recycling of I-129 and Tc-99 however the waste hazard is reduced in the worst case to 20%, but reaches values for times greater than 2.10^5 years below 10%. Actinide loss factors of 10^{-4} would even reduce the waste hazard with I-129 and Tc-99 recycling below 5%.

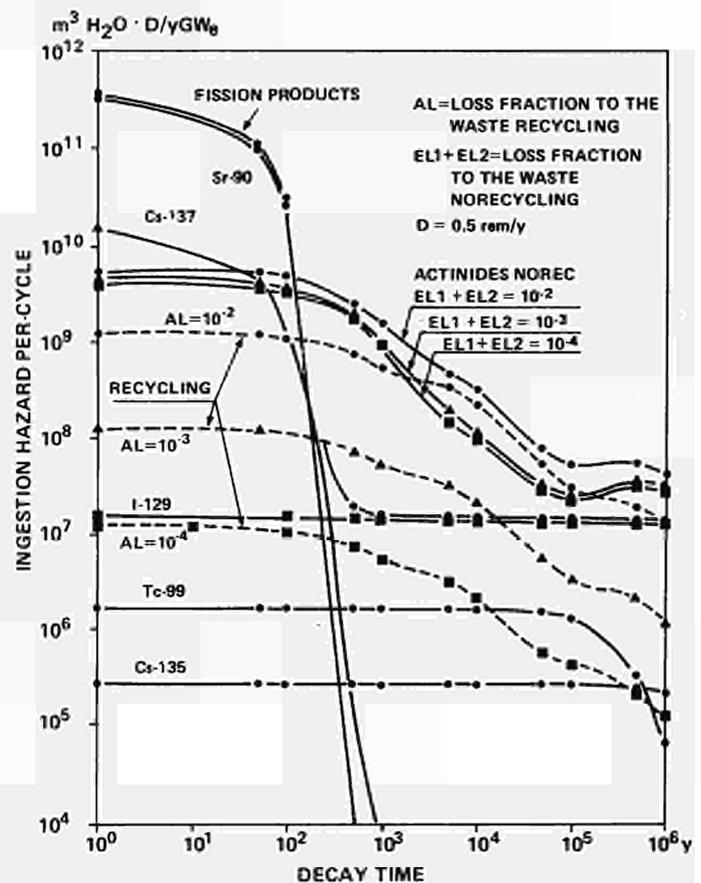


Fig. 6. Accumulated fission product and actinide ingestion hazard waste

Cost evaluations

The assessment of the implications on the fuel cycle costs caused by recycling by-product actinides through power reactors will be performed comparing fuel cycle cost with and without the recycling scheme.

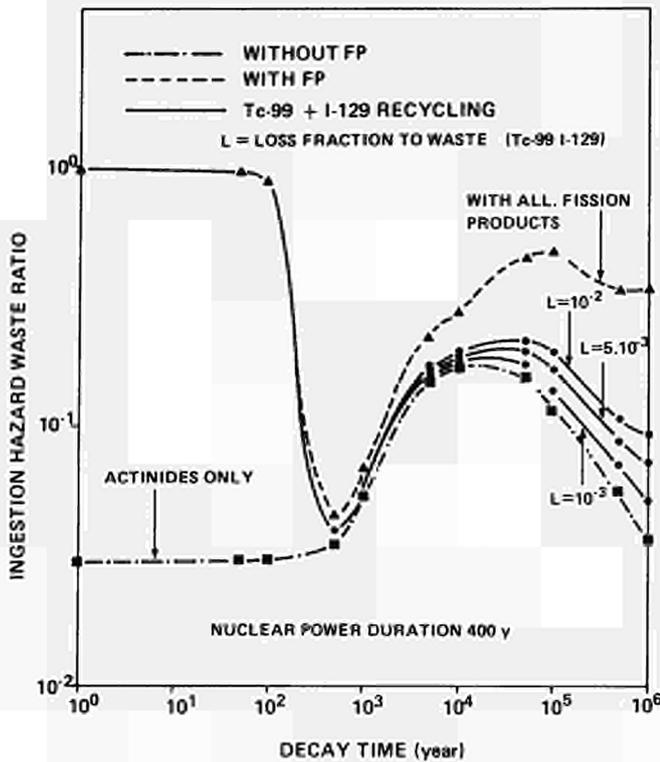


Fig. 7. Ingestion hazard waste ratio with recycling of Tc-99 and I-129

A base for the unit costs as cost of raw materials, conversion cost, enrichment cost, credits for spent fuel material, costs for reconversion, fabrication, reprocessing and shipment as well as waste disposal costs has been acquired. Since reliable unit costs for the fabrication of target fuel elements and the recovery of by-product actinides from the HAW stream do not exist, it is intended to vary these two parameters within a reasonable range, and to observe their influence on the fuel cycle costs expressed in mills per kWh(e) for individual reactors and for a given power economy.

Implications on the fuel cycle

For the assessment of the impacts on the fuel cycle facilities due to recycling actinides, the heat source, ingestion and inhalation hazard, and the neutron source strength of an average subassembly was determined for the reference reactor, for a subassembly with a homogeneous mixing of self-generated by-product actinides in the equilibrium recycle conditions and for a case with heterogeneous recycling of these actinides in 3 special target subassemblies. These data will be communicated for each fuel cycle facility in a separate report at present in preparation.

Future plans are to perform similar studies with a pressurized Pu-fuelled light water reactor.

The problem of the fabrication of fuels containing actinides is under investigation. The possible utilization of sol-gel processes in order to reduce the difficulties related to the handling of powders, is evaluated in the framework of a contract with Agip Nucleare.

Elaboration of a recycle strategy

Based on a forecast for the nuclear power economy system of the member states of the EC, a proposal shall be made as to how the by-product actinides generated by this system can be kept in the fuel cycle. The best-suited reactor type

for transmutation will be indicated, and time-dependent mass flows of heavy metals through the various plants will be given. Criteria for the choice of the reactor type will be neutron-physical arguments, cost and risk consideration.

COLLABORATION WITH EXTERNAL ORGANIZATIONS

Study contract with KFA Jülich and CNEN Casaccia concerning reactor physics calculations for thorium-fuelled reactors and fast breeder reactors.

Study contract with Agip Nucleare concerning fuel fabrication by means of sol-gel processes.

CONCLUSIONS AND PLANNED ACTIVITIES

It is expected that all the background material needed for writing the final report will be available at the end of February, such that only in March the final recycle strategy can be established. However, some chapters of the report may be already written starting at the end of January 1980.

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Actinide Cross-Section Measurements

OBJECTIVES

The foreseen activity of this period included a further development of the experimental methods, the preparation of the instrumentation for the measurements of fast neutron $\sigma(n,\gamma)$ and $\sigma(n,f)$ of Am-243 and initial measurements at the pulsed Van de Graaf neutron generator at the KFK Karlsruhe, using a massive target of Am-243. The first measurement concerns $\sigma(n,\gamma)$. In this experiment an appropriate gamma detector allows to measure gamma-rays discriminated from the very intense low-energy gamma-ray spectrum emitted by spontaneous decay of Am-243. At the same time a correction for the gamma-rays associated with fast fission events in the target is required. For this purpose the experiment includes measurements of the fission rate, at different neutron energies, that will be determined by detecting the fission neutrons using a liquid scintillation detector with gamma PSD. The fission neutron measurements should give, at the same time, preliminary data on $\sigma(n,f)$ of Am-243 interesting also in view of the specific $\sigma(n,f)$ experiment, where the fission neutron spectrum from Am-243 will be accurately measured and normalized to U-235.

RESULTS

The Am-243 sample has been finally received at the KFK near the end of the last semester. The preparation of a suitable sample for the experiments had been considered and discussed also with the JRC-Karlsruhe and Geel, but hard problems have to be solved for the handling of this very active material. Due to the foreseen delay in the sample preparation work at the KFK, in the meantime we have worked to improve and to complement the detection system.

A second scintillation neutron detector system with gamma PSD has been realized to be used in combination with the former one for coincidence work. The improved sensitivity now reached (only 10 pico-Coulomb/MeV at the PM anode)

allows to withstand a very high gamma background, with a negligible gain drift in the detected neutron spectrum. The minimum threshold for fission neutron spectrum is about 100 keV with an effective PSD, and the resolving time is less than a microsecond.

Liquid scintillator containers of different sizes have been made, filled and tested to find the best fission neutron efficiency relative to gamma discrimination. In addition, the PSD circuitry has been provided with a fast timing discriminator (leading edge technique), allowing a time resolution (of some ns) sufficient for neutron TOF measurements in the low energy range (up to 100 keV, or 220 ns/m) and for fission neutron coincidences. We expect an improvement using faster photomultipliers, which have been ordered. A manuscript describing the original part of the instrumentation is in preparation.

COLLABORATION WITH EXTERNAL ORGANIZATIONS

The Am-243 cross-section measurements are being prepared and will be performed in close collaboration with the staff of the Institute for Applied Nuclear Physics of the KFK - Karlsruhe.

CONCLUSIONS

The difficulties encountered in the preparation of the Am-243 sample at the KFK did not allow to start the measurements in this period. However, in the meantime, more activity has been spent for designing and realizing new parts of the experimental equipment and for improvements and tests.

PLANNED ACTIVITIES

The plans for the next period depend on decisions concerning the extension of this activity in the next pluriannual programme. In any case, it is intended that the measurements for which the instrumentation has been prepared will be performed and concluded as soon as the Am-243 sample will be ready.

2.3 Decontamination of Reactor Components

OBJECTIVES

The main purpose of the activity is to contribute to a better knowledge of the decontamination processes for reactor components. The waste management aspects are particularly considered. For the reporting period studies were planned in the following areas:

- A. Chemical decontamination
- B. Physico-chemical structure of oxide layers
- C. Decontamination by redox cycling
- D. Decommissioning of the Ispra I reactor

Due to the fact that this activity will be interrupted at the end of the present year most of the effort has been devoted to the analysis of the data collected and to the preparation of final reports.

RESULTS

A - Chemical decontamination

The systematic study conducted on the effect of various parameters on the effectiveness of the chemical decontamination has been concluded. Tests are in progress to investigate the mechanism of the decontamination by SEM analysis of contaminated surfaces (from power stations) at different decontamination stages.

B - Physico-chemical structure of oxide layers

The physico-chemical structure of oxide films formed in high temperature water on austenitic alloys were studied.

Some results of the analyses performed by means of AES (Auger Electron Spectrometry), SIMS (Secondary Ion Mass Spectrometry) and SEM (Scanning Electron Microscope) including microprobe analysis, on samples of austenitic steel oxidized in hydrogenized water at 300°C had been described in previous Programme Progress Reports^{1,2}.

The SIMS and AES analyses were performed under contract by Dornier System GmbH. The final report of the contract is in preparation.

The preparation for metallographic and microprobe analysis of the specimen investigated by Dornier has been to a great part concluded.

The evaluation of the data and the preparation of conclusive reports is in course.

In some cases inconsistencies among results obtained with different methods have been observed. Two papers have been already completed^{3,4}. The observations during loop operation for 1800 h at 300°C in flowing water treated in one of these papers⁴ can be summarized as follows:

- 1) Quantities and compositions of corrosion products released to the water during loop operation with standard cycles varied considerably.
- 2) The average Fe/Cr/Ni ratio in the released matter, in dispersed and particulate form, over the whole operation period is rather similar to that of the base metal.
- 3) The formation of zones of different composition in the system austenitic steel/water after 1800 h oxidation at 300°C, as already reported in the previous paper³

concerning oxidation in oxygen free water, was confirmed. The different zones are:

- a) The base metal
- b) The metal at the interface with the oxide: decrease of iron and chromium content, increase of nickel
- c) An inner oxide at the interface with the metal: increase of chromium content, decrease of iron
- d) The outer oxide: slight increase of iron, decrease of chromium
- e) The oxide released as particulated to the water: increase of iron content and decrease of nickel.
- f) The composition of total released matter, i.e. particulate and dispersed matter: almost equal to the composition of the base metal.

As far as the contaminated specimen coming from Grundremmingen is concerned, the preparation of the metallographic specimens in the LMA, has been achieved and the investigations are going on.

C - Decontamination by redox cycling

Thermodynamical calculations, performed during the reporting period on the basis of the literature data of chromite spinels (publication in preparation), suggest that redox cycling could have similar effects on austenitic alloys as on certain types of monel.

Because of the displacement of part of the staff to another programme, loop operations cannot be continued so that this activity was dropped.

As far as thermal cycling is concerned we have observed during the loop operations that the total quantity of released matter during the whole period of 1800 h corresponds to about 3% of the quantity of oxide formed on the tube surfaces. Considering that numerous thermal cycling operations have been carried out during this period we cannot expect high efficiency of decontamination of austenitic steel surfaces by cycling techniques.

D - Decommissioning of the Ispra I reactor

The activity is being concluded with the preparation of a technical report giving an evaluation of the radioactivity inventory of the reactor and of the state of the structure and services of the installation.

CONCLUSIONS AND PLANNED ACTIVITIES

The work in the field of decontamination of reactor components is planned to be interrupted at the end of 1979.

Some analyses on contaminated surfaces and the publication of conclusive reports will engage a limited part of the present staff for the first months of 1980.

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Conclusions

CONTENTS

1.	Introduction.....	3
2.	Projects.....	7
2.1	Evaluation of Long Term Hazard of Radioactive Waste Disposal.....	7
	Waste Hazard Analysis.....	7
	Long Term Stability of Conditioned Waste.....	8
	Interaction of Actinides with the Environment.....	10
	Actinides Monitoring.....	12
2.2	Chemical Separation and Nuclear Transmutation of Actinides.....	15
	Chemical Separation of Actinides.....	15
	Assessment Studies on Nuclear Transmutation of Actinides.....	19
	Actinides Cross-Section Measurements.....	25
2.3	Decontamination of Reactor Components.....	27
3.	Conclusions.....	31
4.	JRC Publications.....	37

3. CONCLUSIONS

In the second semester of 1979 the programme has been developed 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 waste hazard analysis it appears of particular interest the application of the methodology developed at Ispra, to the Boom (Belgium) clay formation. After the conclusion of the work for the quantification of the probabilistic value of the geological barrier, the application to the same geological formation of the deterministic section of the JRC model, has been initiated. The work, which is carried out in collaboration with CEN/SCK, Mol, is directed to assess the environmental consequences of an hypothetical radioactive release from a repository mined into the clay formation.

During the reporting period a considerable effort has been spent by the JRC to promote, in the framework of a Risk Analysis Group, further contacts between people of the Community working in the area of risk assessment of geological disposal.

Aim of the Group is to discuss risk analysis approaches, to improve the risk analysis data base and to promote supporting experimental research.

The contacts will take the form of joint meetings between scientists of different disciplines, working in various activity sheets of the JRC and of the Indirect Action Programme.

In the field of the long-term stability of conditioned waste radiation damage studies and leaching experiments on vitrified and bituminized waste have been continued.

Borosilicate glasses containing uranium are being irradiated in the HRF reactor (Petten), in order to simulate, by fission fragments damage, the long-term damage produced by alpha-emitters in vitrified wastes. The simulation technique using fission fragments which has been developed by the JRC makes possible to reach an equivalent time of the order of one million years.

In order to investigate the basic mechanisms of the damage, irradiations by heavy ions of silica samples have been carried out. Measurements by means of optical and electron microscopy techniques will provide information on damage distribution and annealing effects.

Leaching tests on borosilicate glasses (using water typical of a clay repository) and bitumen loaded with different oxides have been carried out.

In the field of the interaction of actinides with environment, experiments have been carried out on the migration behaviour in columns of glauconite sand of actinides (Pu, Am and Np) leached from a glass matrix. In order to interpret the results of the migration experiments, investigations on the chemical form of the actinides have been performed. The results obtained confirm the complexity of the migration phenomena and the major role of the physico-chemical nature of the migrating species. For the glauconite sands the retention of the actinides seems

not to be related to the exchange capacity of the minerals but rather due to the highly effective filtering ability.

In the field of actinides monitoring, setting-up of instruments and design parameter studies have been carried out for the non-destructive assay techniques utilized for plutonium monitoring: passive gamma, passive neutron and active neutron assay techniques.

The methodology developed by the JRC is being applied in the framework of an Integral Experiment on the plutonium waste monitoring system of the Dounreay Nuclear Power Development Establishment (DNPDE).

The International Meeting on Monitoring of Pu-Contaminated Waste has been held at Ispra on 25-28 September 1979 with the participation of 130 people. The meeting was very useful to assess progress and to establish the direction of future work.

The planning of future activities for the project Evaluation of the Long-Term Hazard of Radioactive Waste Disposal is shown in Table I. It should be noticed that on the basis of the JRC programme proposals 1980-1983 it is planned to reinforce considerably the activities of this project. However, the planning given in Table I refers only to the continuation of the activity employing the present staff. A complete planning, taking into account staff redistribution will be prepared only after the Council decision on the new JRC programme.

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 for the beginning of 1980 a major report dealing with a critical evaluation of the feasibility of the chemical separation and nuclear transmutation of actinides.

The Second Technical Meeting on the Nuclear Transmutation of Actinides to be held at Ispra in April 1980, under the sponsorship of NEA/OECD will provide a very useful opportunity for a discussion of the results obtained at the JRC and for a comparison with the conclusions drawn by other organizations.

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

The studies on the OXAL process have been completed during the first semester of 1979.

During the second semester of 1979 the activity was directed to investigate some aspects of the processes based on the use of solvent extraction. Concerning the HDEHP process, denitration experiments and batch extraction tests on fully active scale have been carried out. A series of radiation damage tests have been performed on HDEHP in order to evaluate the behaviour of this solvent under plant conditions. Concerning the TBP process, batch-countercurrent experiments on simulated HAW solutions have been carried out.

The flow-sheets proposed by the JRC Ispra, based on HDEHP and TBP solvent extraction are being verified in the laboratories of CEA (CEN, Fontenay-aux-Roses, France) by continuous countercurrent experiments using mixer-settler batteries. This work is carried out in the framework of a collaboration contract between CEA and JRC.

The laboratory work on chemical separation of actinides from HAW is complemented by an engineering assessment directed to evaluate the feasibility of the processes at the plant scale.

On the basis of the laboratory work and engineering assessment, the conceptual design of a plant for HAW partitioning will be prepared.

The assessment studies on the nuclear transmutation of actinides include reactor physics calculations, fuel element design, evaluation of cost and risk and of implications on the nuclear fuel cycle.

The reactor physics calculations provide the basic data on the formation and transmutation of actinides in nuclear reactors. Calculations for LWR, FBR and HTR reactors are being completed at the JRC and in national laboratories in the framework of study contracts.

The work on the design of fuel elements for the actinides recycling in FBR and LWR reactors has been continued.

In order to be in position to judge on the interest of the actinides transmutation, evaluations are being completed on the reduction of the long-term waste hazard and on the various implications on the nuclear fuel cycle.

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

On the basis of the JRC programme proposals for 1980-1983 it is planned to reduce considerably during 1980 the activity in the field of the chemical separation of the actinides from HAW and nuclear transmutation. The staff made available should be employed in studies of risk analysis, actinides monitoring and plutonium separation from various waste

streams. However, during 1980 sufficient staff will be allocated to the present activities on chemical separation and nuclear transmutation of the actinides, to make possible the best exploitation of the results produced by the project.

Decontamination of Reactor Components

The activity in this area includes studies on chemical decontamination and on physico-chemical structure of oxide layers. The systematic study conducted on various parameters, influencing the effectiveness of the chemical decontamination, has been completed. Tests are in progress to investigate the mechanism of the decontamination by SEM (scanning electron microscope) analysis of contaminated surfaces from nuclear power stations. The physico-chemical structure of oxide layers prepared in a small water loop has been investigated by various analytical techniques.

Part of this project was also the evaluation of the possible interest of the decommissioning of the ISPRA-I reactor as a source of information of general interest for reactor decommissioning. Due to the decision not to include this activity in the research proposal of the JRC for 1980-1983, the work in this area is being concluded with the preparation of a technical report giving an evaluation of the radioactivity inventory of the reactor and of the state of the structure and services of the installation.

On the basis of the JRC programme proposals for 1980-1983 it is planned to interrupt the project Decontamination of reactor components at the end of 1979. As a consequence the results obtained are being assembled in final publications. During 1980 a minor effort will be spent for some conclusive experimental tests. It is planned that part of the staff of this project should reinforce the experimental work connected with risk evaluation.

For further information please contact the Programme Manager.

PLANNED ACTIVITIES AND IMPORTANT MILESTONES (JANUARY - JUNE 1980)

Table 1. PROJECT 1: Evaluation of the long-term hazard of radioactive waste disposal

Activities	first semester 1980 (month)						second semester 1980	
	1	2	3	4	5	6		
Waste hazard analysis	A							
	B							1
Long-term stability of conditioned waste	C				2	-----		
	D							3
	E						4	5
Interaction of actinides with the environment	F				6			7 8
	G							
Actinides monitoring	H						9	10 11

- | | | | |
|---|--|----|---|
| A | Coordination activity in the framework of the risk analysis group | | |
| B | Actinide distribution in the environment following failure of geological barrier | 1 | Completion of the study for the application of the deterministic model to the Boom clay formation |
| C | Leaching tests on vitrified waste | 2 | Completion of the experiments with water typical of clay formations |
| D | Leaching tests on bituminized waste | 3 | Completion of the experiments on bitumen charged with dispersed oxide powders |
| E | Experiments of radiation damage in glasses | 4 | Completion of the glass samples irradiation in the HFR reactor |
| F | Interaction with abiotic environment | 5 | Start-up of the post-irradiation tests |
| | | 6 | Completion of the literature survey and choice of the analytical methods for stability constant determination |
| | | 7 | Completion of a first series of tests with americium on sand columns with different filtration efficiency |
| 8 | Completion of experiments on elution behaviour of leached plutonium and americium through glauconitic columns | | |
| G | Collection of data and promotion of activities in a strict link with the Indirect Programme Radiation Protection | | |
| H | Plutonium waste monitoring | 9 | Completion of test measurements in the ALKEM fabrication plant |
| | | 10 | Completion of the integral experiment at Dounreay |
| | | 11 | Completion of the theoretical assessment on the active neutron reference monitor |

PLANNED ACTIVITIES AND IMPORTANT MILESTONES (JANUARY - JUNE 1980)

Table II. PROJECT 2: Chemical separation and nuclear transmutation of actinides

Activities	first semester 1980 (month)						second semester 1980
	1	2	3	4	5	6	
International coordination A	-----						-----
Chemical separation of actinides B	-----		-----		-----		
Assessment studies on nuclear transmutation of actinides C	-----		-----		-----		
Actinides cross-section measurements E	-----						-----

- | | |
|--|---|
| <p>A International coordination in the framework of the OECD Nuclear Energy Agency</p> <p>B Chemical separation of actinides from HAW by solvent extraction and OXAL process</p> <p>C Nuclear transmutation of actinides in FBR, LWR and HTR reactors</p> <p>D Measurement of cross-sections of Am-243</p> | <p>1 Organization of the 2nd Technical Meeting on the Nuclear Transmutation of Actinides, JRC-Ispra, April 1980</p> <p>2 Summary report on the chemical separation and nuclear transmutation of actinides</p> <p>3 Definition of studies requiring further development during 1980</p> <p>4 Completion of the experiments for (n, γ) and (n, f) reactions</p> |
|--|---|

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