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Management of Nuclear Materials and Radioactive Waste

PROGRAMME PROGRESS REPORT

January - June 1979

Programme Progress Report JRC Ispra
January - June 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 January-June 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

The study for the application of the risk analysis methodology, developed at the JRC, to quantify the probabilistic value of the geological barrier in the Boom (Belgium) clay formation has been completed.

Radiation damage studies and leaching tests have been carried out on borosilicate glasses to assess the value of the barrier provided by waste conditioning.

A set of experiments on the migration of plutonium and americium in columns of the soil which overlies typical clay formations, have been completed.

The Integral Experiment on the plutonium waste measurement system of the Dounreay Nuclear Power Development Establishment (DNPDE) is progressing.

Project 2 - Chemical separation and nuclear transmutation of actinides

In the field of the chemical separation of the actinides from high activity waste (HAW) the studies on the OXAL process have been completed. Several experiments have been carried out on fully active laboratory scale, for the denitration of HAW and for the solvent extraction process using HDEHP.

For the assessment on the nuclear transmutation of actinides, results have been obtained concerning reactor physics calculations in various types of reactors, temperature profile calculations in FBR for fuel elements containing by-product actinides and risk assessment for the radioactivity release from nuclear plants.

Project 3 - Decontamination of reactor components

Studies on the physico-chemical structure of oxide layers and on the efficiency and mechanism of chemical decontamination have been carried out.





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1. INTRODUCTION

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

The Joint Research Centre (JRC) started work in the field of radioactive waste management in 1973. This programme is part of the activity of the JRC in the field of Nuclear Safety which includes also the programme Reactor Safety and the programme Plutonium Fuel and Actinide Research. The staff allocated to the programme for 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 was established.

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



Projects

2. PROJECTS

2.1. Evaluation of Long Term Hazard of Radioactive Waste Disposal

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

The barriers considered are the following:

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

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

In the field of 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 and test a methodology to assess the long-term risk linked to the disposal of radioactive wastes into deep geological formations.

Our methodology, which was illustrated in previous reports¹⁻³, is based on the following steps:

1. Identification of any barrier able to prevent, decrease or delay the radioactivity flow towards the biosphere
2. Modeling of the barriers in such a way that either a probabilistic or a deterministic quantitative description of the behaviour of each barrier against radioactivity escape can be drawn

3. Data collection and model application, to assess containment failure probabilities and radioactivity doses to man
4. Sensitivity analysis, to get information on the relative importance of the various parameters.

The barriers identified are the following:

1. Segregation afforded by the geological formation itself, in which the radioactive waste has been buried. This barrier is treated, in our approach, as a binary system, which only can be either in functional or in failed state. A probabilistic approach is therefore used to model it; to this purpose, we utilize the Fault Tree Analysis (FTA) technique, through which release probabilities as a function of time can be obtained.
2. Physical and chemical stability of the wastes; this barrier takes account of the leachability and physical integrity of the conditioned materials and consequent availability of radionuclides towards transport by water.
3. Geochemical retention of isotopes during their transport by water through porous underground media; this barrier may cause large delays in radioactivity appearance at the land surface; its modeling relies upon mathematical treatment of the retention phenomena which accompany the solution migration through soil columns.
4. Environmental mobility and biological availability of radioelements; this barrier is particularly effective for most of the long-lived alpha-emitters; it is modeled in our approach through the definition of an environmental system composed of compartments in equilibrium, each transfer being governed by constant coefficients.

For the first semester of 1979 the planned activities were:

- A. to complete the test of the FTA to a real site through an application to the Belgian clay formation of Boom
- B. to conclude the comparison of the long-term risks linked to two different nuclear fuel cycle strategies, as described in the previous Programm Progress Report³
- C. to start the application of the deterministic section of the model to the Boom clay formation.

RESULTS

A The probabilistic safety analysis of a waste repository in the Belgian clay formation has been performed in close collaboration with the CEN/SCK, Mol; release probabilities to three different receptors (groundwater, land surface and atmosphere) were estimated over four different time periods (2,000, 25,000, 100,000 and 250,000 years).

The overall release probabilities to groundwater are shown as a function of time in Fig. 1; because of obvious uncertainties in geology predictive capabilities, a probability band has been drawn. Faulting phenomena are among the principal mechanisms having the potential to cause releases to groundwater, while direct releases to land surface may be linked to various glacial phenomena; in the short term, different kinds of human actions may be important.

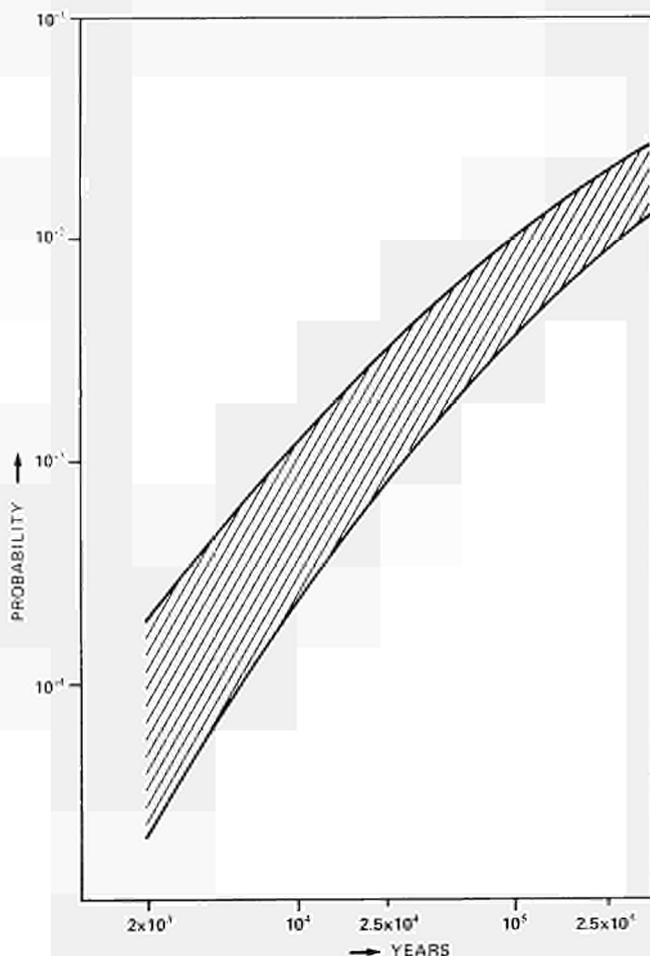


Fig. 1 – Release Probability to Ground Water

The overall failure probabilities appear to be low enough (about 10^{-7} per year) to offer a good margin of safety. As a matter of fact, it is worth noting that the annual failure probability is not a constant, as the integral probability tends asymptotically to unity; however, for geological short time periods, a nearly constant annual probability may be considered.

A report is now being prepared, in collaboration with the CEN/SCK, Mol.

B The comparison of long-term risks linked to throwaway and plutonium recycle strategies, as described in the previous Programme Progress Report³ has been completed⁴.

The following conclusions were reached:

1. The long-term risk linked to the once-through strategy is much higher than that due to the recycle strategy; in fact in the latter option, the plutonium destruction largely exceeds the build-up of transplutonium isotopes.
2. In both strategies the risk would be governed mainly by inhalation of plutonium isotopes, if the release occurs within a few thousands years after repository closure; on the other hand, if the release should occur after more than 10^5 years, ^{226}Ra will be the risk-governing isotope.
3. The dose-rates to individuals appear to be rather small, even in the worst case. Some actions having the capability to decrease even further the long-term risk of radioactive wastes and decrease the uncertainty connected with the quantification of the risk, were identified (Table I).

Table II shows critical parameters and problem areas which are suggested for additional study in the coming programmes.

C Discussions have been held with CEN/SCK, Mol in view of the application of the deterministic section of our model to the Boom clay formation. A collaboration contract is in preparation.

COLLABORATION WITH EXTERNAL ORGANIZATIONS

The collaboration with the CEN/SCK, Mol has already been mentioned.

The contracts of the indirect action programme Radiation Protection (DG XII) relevant for waste hazard analysis are reported in the section «Interaction of Actinides with the Environment».

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 very 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 at industrial levels.

This conclusion is in a fair agreement with the results obtained from similar studies performed in different countries.

Table I – Suggested Actions to Decrease the Long-Term Risk of Radioactive Wastes

	Statement	Element of concern	Suggested actions or options
I	Throwaway strategy implies larger long term risks than plutonium recycle strategy	Pu	Plutonium destruction by recycle
II	Plutonium inhalation governs the risk for releases occurring within 50,000 years	Pu	Minimization of plutonium content in wastes from reprocessing and refabrication plants
III	Ingestion of Ra^{226} governs the risk for releases occurring after 100,000 years	Ra	Minimization of plutonium content in waste. Delayed reprocessing of MOX-fuels to maximize the decay of Cm^{242}
IV	In the Pu-recycle strategy cladding hulls could bear larger fractions of irradiated fuel than assumed in the reference case	Pu Ra	To envisage a decontamination step for cladding hulls

Table II – Critical Parameters and Problem Areas

I	Leach rates and surface-to-volume ratio of vitrified high level wastes; their evolution with time.
II	Conditioning techniques for medium and low-level alpha-waste.
III	Phenomena governing the geochemical retention, physico-chemical state of leached out elements in groundwater, with emphasis on plutonium.
IV	Physico-chemical state of actinides in surface water; their distribution between water and sediments.
V	Equilibrium concentration of actinides in surface soil; their downward migration, resuspension processes.
VI	Environmental behaviour and aging phenomena concerning Pu, Am and Ra.

PLANNED ACTIVITIES

As a logical continuation of the probabilistic assessment already performed, also the deterministic section of our methodology will be validated through an application to the Belgian clay formation of Boom, in collaboration with the CEN/SCK, Mol.

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Long Term Stability of Conditioned Waste

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 first semester of 1979 were the following :

- preparation of a new series of capsules (BONI III) for radiation damage measurement in glasses
- verification of the validity of accelerated tests of radiation damage on vitrified HAW
- continuation of leaching tests on glasses
- continuation of leaching tests on bitumen

RESULTS

Radiation damage on glasses

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 10^5 years.

It seems worthwhile 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 will be used in the new irradiation experiments.

The capsules will be irradiated in the HFR reactor at Petten for about 8 months in order to reach about 1.2 dpa. The capsules have been prepared and delivered to Petten. The irradiation will start in June.

A paper¹ on the simulation of radiation damage in glasses has been presented at the International Symposium on Ceramics in Nuclear Waste Management (Cincinnati).

Verification of the validity of accelerated tests of radiation damage in vitrified high activity waste

Results of stored energy measurements, performed at the JRC Ispra, on borosilicate glasses irradiated with heavy ions are compared in Table I with the results obtained on similar glasses irradiated with internal sources (fission fragments at Ispra¹, Pu recoil at Harwell²).

Differences in the stored energies measured in different samples irradiated with heavy ions, may be due to the effect of the metal coating deposited on the irradiated surface of some of the samples. In fact the metal coating keeps the irradiated surface near the temperature of the sample holder, i.e. the temperature of the cooling water, while the uncoated surface is at a higher temperature due to the low thermal conductivity of the glass. Differences in the results obtained using external or internal irradiation may be due to an imperfect control of the temperature and/or to a different distribution of the damage.

In spite of these uncertainties the data reported in Table I make it possible to draw the following conclusions:

- A saturation effect is evident at the dose of 0.1 dpa.
- The use of displacement per atom (dpa) as a unit of dose seems appropriate for the radiation damage of glasses, since the values of stored energies for glass samples of BONI type as obtained by external irradiation and fission fragment internal irradiation, are comparable at a comparable number of calculated displaced atoms.
- When considering the large variation of dpa/sec (a factor of $10^3 - 10^4$ between external and internal irradiations) the dose rate does not appear to have a considerable influence on the measured stored energy. Detailed results of this work are reported in Reference ¹.

Table I – Measurements of Stored Energy in Borosilicate Glasses

Sample	dpa	dpa/sec	dpa/g	Source of damage
glass BONI*	0.1	$1.35 \cdot 10^{-4}$	40	Ni ⁺⁶ 46.5 MeV
glass BONI	0.1	$1.35 \cdot 10^{-4}$	35	Ni ⁺⁶ 46.5 MeV
glass BONI	1	$1.35 \cdot 10^{-4}$	30	Ni ⁺⁶ 46.5 MeV
189E*	0.1	$1.35 \cdot 10^{-4}$	65	Ni ⁺⁶ 46.5 MeV
189E	1	$1.35 \cdot 10^{-4}$	40	Ni ⁺⁶ 46.5 MeV
glass BONI	0.36	1.10^{-7}	19	fission fragments [1]
189E	0.5	$1.6 \cdot 10^{-8}$	23	Pu recoil [2]

* Coated with a 100 Å silver film

Concerning the fundamental aspects of irradiation damage, annealing studies of heavy ions irradiated silica glasses have been pursued more systematically by means of optical methods. In order to separate in the absorption spectra the band due to displacement (B_2) from that due to ionization (E'_1) a best fit program was employed. The result is illustrated in Fig. 1. These results allow us to determine the ratio between the displacement defects and defects due to the bond breakage in a heavy ions irradiated silica glass.

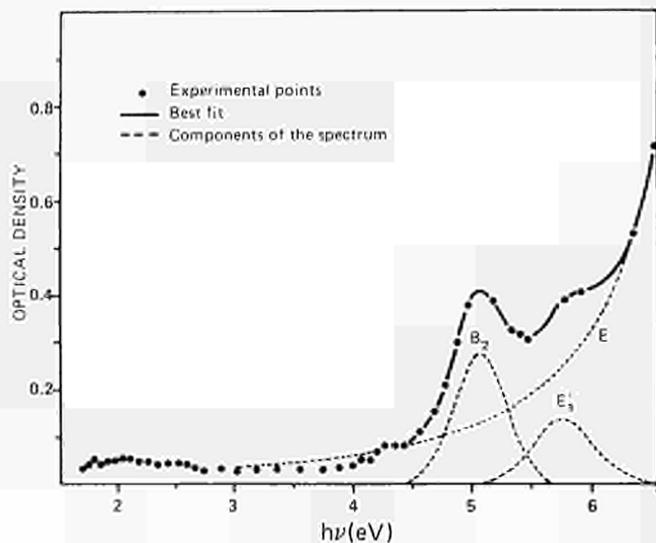


Fig. 1 – Absorption Spectra of Heavy Ion Irradiated Silica

This is a very important parameter to evaluate the complex role of ionization energy in the production of defects during irradiation. In Table II the number of color centers due to displacement (B_2 band) is reported together with the number of color centers due to ionization (E'_1 band) as a function of displacements per atom and of total energy dissipated. A saturation effect is evident.

The research will be pursued in the direction of defining the type of the introduced defects, their distribution in the bulk and their annealing properties.

Table II – Defects Measured by Optical Absorption in Heavy Ion Irradiated Silica

Sample	B_2 Band centers/cm ³	E'_1 Band centers/cm ³	Total Dose dpa	Dissipated Energy Joule/cm ³
1	5.92×10^{18}	1.22×10^{19}	0.5	1.5×10^8
2	6.60×10^{18}	1.75×10^{19}	0.5	1.5×10^9
3	5.18×10^{18}	1.20×10^{19}	1	3.1×10^9
4	5.86×10^{18}	1.52×10^{19}	0.1	3.1×10^7
5	6.54×10^{18}	1.35×10^{19}	0.1	3.1×10^7
6	5.6×10^{18}	1.38×10^{19}	0.1	3.1×10^7
7	6.42×10^{18}	1.94×10^{19}	0.1	3.1×10^7

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 into 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.

Tests were performed at 80°C and 50°C. Fig. 2 shows the results obtained at up to 5,000 hours of leaching.

The comparison between measurements in pure water and in water conditioned by clay, shows that the leaching rates are quite similar in the experiments at 80°C and are slightly lower in water conditioned by clay in the experiments at 50°C.

A paper³ on the leaching tests in pure water has been presented at the International Symposium on Ceramics in Nuclear Waste Management (Cincinnati).

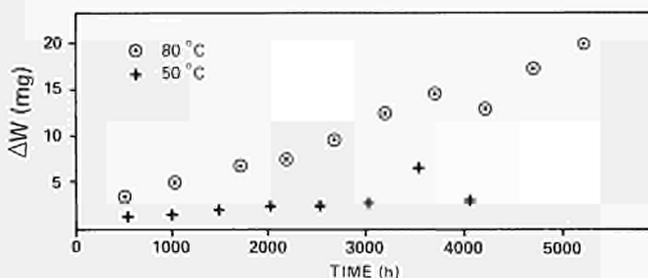


Fig. 2 – Weight Losses of Glass Samples Leached by Water Conditioned with Clay

Leaching tests on bitumen

The tests performed on bitumen loaded with 40% NaNO_3 have shown an extensive swelling. As a consequence the surface exposed to the leaching water has increased considerably.

The amount of Na which is leached at the beginning follows a regular curve of the parabolic type. When the swelling begins to be important the amount leached increases rapidly.

The most reasonable explanation of such a swelling is the retention of large amounts of water in the loaded bitumen. NaNO_3 is strongly hygroscopic so that around the grains which are embedded into the bitumen, bubbles of water, which have diffused through the bitumen, coalesce. Another test using $\text{Ce}_2(\text{NO}_3)_3$ is in progress. Even $\text{Ce}_2(\text{NO}_3)_3$ is hygroscopic but not so strongly as NaNO_3 . This test will allow to decide if it is wise to try to formulate a model of long-term leaching for bitumen conditioning hygroscopic material. Further tests on oxide powders (Fe_2O_3 , CeO_2) will begin shortly.

COLLABORATION WITH EXTERNAL ORGANIZATIONS

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

A collaboration has been started with the University of Modena concerning examination by SEM (scanning electron microscope) of heavy ion irradiated samples.

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 particular, from the comparison of stored energy measurements on borosilicate glasses irradiated in different conditions, it has been concluded that the dose rate has not a considerable influence on the damage.

This is a first proof of the validity of the accelerated tests of radiation damage on vitrified high activity waste.

For the second semester of 1979 the following activities are planned in the field of radiation damage:

- Irradiation of capsules, 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 SiO_2 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).

Some preliminary evidence for the presence of strongly deformed areas along the heavy ions path in irradiated silica samples, obtained by scanning electron microscopy, is particularly encouraging to examine these aspects of the damage.

Leaching tests

The leaching tests on borosilicate glasses, using water conditioned by clay, have shown only slight differences in the leaching rates respect to the previous tests performed with pure water.

The leaching tests using water conditioned by clay will be completed during the second semester of 1979.

In the second semester of 1979 leaching tests will be continued on bitumen conditioned with different materials.

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Interaction of Actinides with the Environment

OBJECTIVES

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

For the first half 1979, the planned activities were the following:

- a) Continuation of long-term column experiments on actinide migration
- b) Initiation of theoretical studies to model the interaction between leached out nuclides and geological media
- c) Development and application of analytical techniques for the determination of actinide chemical forms in aquatic media.

RESULTS

A Migration experiments have been continued on columns filled with glauconite sand sampled from the Antwerp sands overlaying the Boom clay formation in Belgium

A water pathway has been established which flows over the alpha-bearing glass (^{238}Pu and ^{241}Am) prepared in the laboratory and then through the columns containing soil samples.

The set-up adopted has been illustrated in the previous Programme Progress Report (July-December 1978). First results have been presented in reference 1. In the experiments a water composition typical of the main aquifer of the region has been utilized.

The output from the column is periodically sampled for α -activity and at the end of the experiment the actinide profile within the column is determined.

For the analysis of the soils, each 180 mm column was sliced into thin sections. Americium activity was determined by gamma counting; in the case of plutonium, chemical separation was performed and the α -activity counted by liquid scintillation spectrometry.

Figures 1 and 2 show the ^{238}Pu and ^{241}Am activity profiles of the soil columns after three months of percolation of the leached solution. On the basis of reported distribution coefficients, assuming a reversible ion exchange mechanism, the activity should have been confined in the first few millimeters of the column. However, a continuous small activity output was found to occur and the column profile shows that the long-term interaction between the leachate and the column cannot be described by a simple ion exchange mechanism.

The physico-chemical nature of the migrating species is being studied; the formation of neutral or anionic complexes might explain the existence of mobile chemical forms of transuranic nuclides.

During the present report period, ^{237}Np spiked borosilicate glasses have also been prepared and migration experiments started.

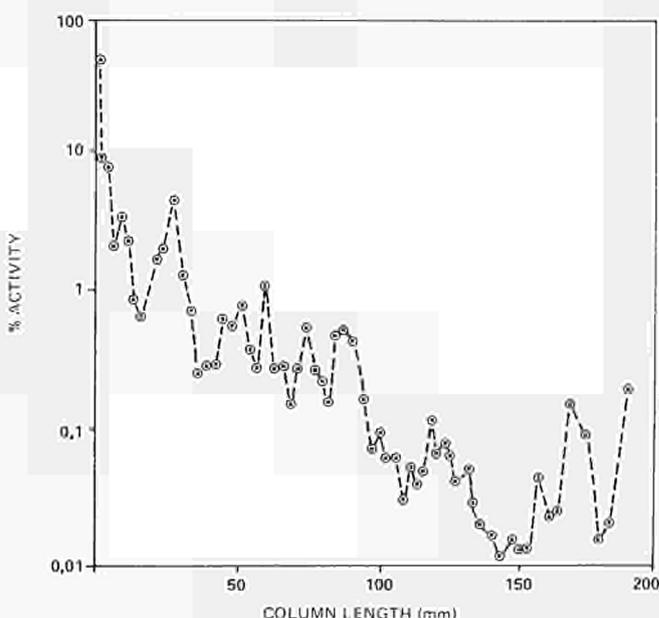


Fig. 1 - Vertical Distribution of Leached ^{238}Pu in Glauconite Sand; Three Months Percolation Experiment

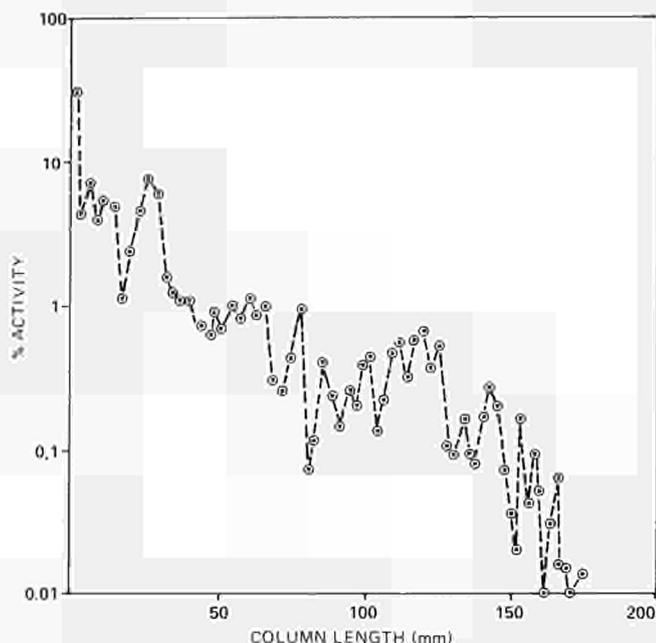


Fig. 2 - Vertical Distribution of Leached ^{241}Am in Glauconite Sand; Three Months Percolation Experiment

B A theoretical study to assess the chemical forms of plutonium in groundwater has been undertaken. On the basis of thermodynamic data on equilibrium constants, different chemical species of plutonium can be shown to be present in natural waters, as a function of redox potential, pH and chemical composition of the medium. In the different valence states they can form a great variety of soluble complexes with the inorganic anions, which are commonly present in groundwater; among them, the most important are the carbonates because of their great stability.

The study² very clearly indicated that an accurate knowledge of such types of complexes is of great importance if reliable modelling of plutonium migration is to be undertaken. The most important equilibrium constants for which reliable values are not yet available are those of Pu (III) and Pu (IV) species with carbonates

C Parallel to these theoretical calculations analytical methods for the estimation at very low concentration of actinide valence states are being developed. These methods should be applied in connection with point B, to the characterization of physico-chemical forms of actinides in leached solutions. The same techniques should be used in order to follow the evolution of chemical species of actinides in selected aquatic ecosystems.

A combination of techniques is being studied for the determination of plutonium (III), (IV) and (VI) and americium (III). These include the use of filtration, cation and anion resins, calcium fluoride columns and solvent extraction by thenoyltrifluoroacetone (TTA).

Experiments were carried out using the gamma emitting ^{237}Pu isotope*. The production of the different oxidation states was carried out using classical procedure. The chemical forms produced (III), (IV) and (VI) were then chemically purified so that the initial species of the isotopes was known. Testing and development of the various techniques described below were then carried out using these solutions.

* The donation of this isotope for the analytical method development work by the Biologische Anstalt Helgoland, Hamburg, is gratefully acknowledged.

Solvent extraction with TTA

It has been shown³ that the oxidation states of tracer quantities of plutonium in aqueous media can be determined using 2-thenoyltrifluoroacetone. This procedure was tested using different chemical forms of ^{237}Pu in water samples (20 ml) containing approximately 1 nCi activity (37 Bq) i.e. $\sim 10^{-14}$ molar solutions. A parallel study was started with ^{241}Am (III). Extraction was carried out using 0.4 M TTA dissolved in xylene. The results obtained agree closely with literature values and are shown in Table 1. For extraction carried out at pH 4.3 sufficient ammonium acetate was added to render the final solution > 10 M with respect to acetate. The acetate complexes the (VI) ions and prevents them being extracted. It was noted that Am (III) behaved as Pu (III), being extracted at pH 4.3 but not at pH 0.3.

Table 1 - Percentage Extraction of Plutonium and Americium by 0.4 M Thenoyltrifluoroacetone in Xylene Under Different Conditions of pH, from 1 Day Old Samples

Chemical species	pH	Distilled water	Fresh water	Seawater
Pu III	4.3*	100	93	93
	0.3	8	5	7
Pu IV	4.3*	94	98	98
	0.3	97	96	80
Pu VI	4.3*	7	10	20
	0.3	11	5	8
Am III	4.3*	91	90	92
	0.3	2	1	1

* Aqueous solution > 10 M in ammonium acetate
pH Adjusted with HCl acid
Shaking time about 10 mins.

Adsorption on CaF_2 and ion exchange with resins

Initial studies on the behaviour of different Pu valence states on cation and anion exchange resins and CaF_2 columns for aqueous solutions having hydrogen ion concentrations in the range pH 1 - 12 are underway.

Calcium fluoride columns containing the crystallized salt (200 - 400 mesh) were used to compare the chemical form of plutonium to those obtained by TTA solvent extraction techniques. It appears possible to distinguish between Pu (IV) and (VI) without altering the natural conditions of the solution. Detailed results will be reported at a later date.

Application of chemical speciation techniques to the study of the behaviour of actinides in simulated aquatic environments

The interest and applicability of the chemical speciation techniques described above to studies of the behaviour of actinides in aquatic environments is being tested in a comparative study on the behaviour of Pu and Am in vitrified form vs. the behaviour of Pu and Am added as a solution.

Two different sources of actinides have been simulated containing ^{237}Pu and ^{241}Am . In two aquariums actinide contaminated borosilicate glasses (~ 2 g) were suspended about 2 cm above the sediment surface; in other two aquariums solutions of plutonium (IV) and americium (III) were introduced into the surface waters. The four aquariums contained about 20 l lake water overlying a 4-6 cm deep sediment layer. The systems further contained a certain biomass made up of fish (2 species), snails (2 species) and plants. The microcosms were allowed to become well established (about 1 year) before the start of

the experiment. Growth and reproduction of all components of the aquarium occurred, one of the best indices for comparison to a natural system.

The uptake, distribution and behaviour of plutonium and americium were followed for a period of three months. Samples of each compartment of microcosms were measured approximately every 10 days. Using the analysis train described in the previous section, the evolution of the chemical forms originating from the two different source terms was followed in the aquariums.

Although the water activities are low in the aquariums containing the contaminated glass, measurable quantities were detected throughout the three months period. The formation of soluble forms having both positive and negative charges can be deduced by their absorption on the ion exchange resins. In each case a non-negligible fraction of non charged forms could also be detected. This was particularly noticeable in the case of contamination originating from ²⁴¹Am glass.

The fact that the formation of soluble forms of actinides can be identified and that differences occur between not only the radioelements but also perhaps due to the source term underlines the importance of characterizing the species in the environment.

The development of separation techniques and their further application will continue.

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 have been outlined.

- 1) Institute of the Association EURATOM-ITAL, Wageningen, The Netherlands :
«Behaviour of radiocontaminants in soils and vegetation of specific sites»
- 2) Commissariat à l'Energie Atomique, Centre d'Etudes Nucléaires de Cadarache, France :
«Transfer of plutonium in the environment»
- 3) Université Catholique de Louvain, Belgium :
«Translocation of radionuclides absorbed by the roots (redistribution) and subsequent release from leaves into the atmosphere (biological resuspension)»
- 4) Biologische Anstalt Helgoland, Hamburg, FRG :
«Investigation of the distribution of actinides in marine and estuarine environments»
- 5) Radiobiology Dept. SCK/CEN Belgian Nuclear Center :
«Biological availability of transuranics in freshwater systems»
- 6) Delta Institute for Hydrobiological Research Yerseke, The Netherlands :
«Plutonium in the Rhine-Meuse-Scheldt Delta: analysis of estuari sediment, saltmarsh and vegetation samples»
- 7) Ministry of Agriculture, Fisheries and Food, Lowestoft Laboratory, U.K. :
«Distribution and behaviour in the marine environment of radionuclides of waste disposal significance from the utilization of nuclear energy»

Discussions have also been held with scientists of the CNEN Fiascherino Laboratory, Italy (chemical speciation of actinides in aquatic systems); CEA Cadarache, France (in situ study of actinides in marine environment) and CEA Fontenay-aux-Roses, France (assessment of accidental or continuous low-level discharge of radioactivity into the environment).

These contracts are expected to provide the necessary data to validate the environmental models used in long-term waste hazard evaluations.

CONCLUSIONS AND PLANNED ACTIVITIES

The programme on actinide interaction with the environment is now well established and is following closely the timing of the planned objectives. However, owing to the complexity of the development work necessary for the columns under pressure this project is at present in abeyance.

In the second half of 1979 the subsequent points are foreseen:

- Continuation of column migration experiments with ²³⁸Pu, ²⁴¹Am and ²³⁷Np.
Results for experimental periods up to six months should then be available.
- Continuation of theoretical modelling studies.
- A more detailed investigation both on the development of analytical techniques and valence states evolution will be performed.

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3. S.C. FOTI, C. FREILING : «The determination of the oxidation states of tracer uranium, neptunium and plutonium in aqueous media», Talanta, 11, 385-392, 1964.

Actinides Monitoring

OBJECTIVES

The study aims at the development of a methodology for plutonium waste monitoring. For the reporting period theoretical and experimental work is planned concerning development of reference instruments and methods.

Moreover mathematical modeling of the Pu waste measurement system of the Dounreay Nuclear Power Development Establishment (DNPDE) was foreseen.

RESULTS

Integral Experiment

In the frame of a collaboration programme an integral experiment is in execution at the Pu waste measurement system of the Dounreay Nuclear Power Development Establishment. The first Progress Report on the integral experiment has been prepared¹.

The most important achievements during the reporting period are the following:

- a) Delayed neutron measurement technique with a 14 MeV neutron generator.

In this technique the waste item is irradiated for 2 min. and after 5 sec. waiting time the delayed neutrons are counted for 1 min. The detection process of these neutrons is modelled by means of Monte Carlo calculations with the TRIPOLI 02 code in the framework of a subcontract with the University of Lyon. The calculations help to understand the neutron transport in the prompt and delayed neutron domain.

In addition the calculation with the TRIPOLI 02 code provides few group constants which are required for a simple interpretational model. This model, developed at Ispra, includes:

- A set of mathematical expressions for fission rate, prompt and delayed neutron production, etc.
- Calibration procedures
- Corrections for self-shielding and matrix effects.

b) Passive neutron assay.

The effective escape probabilities have been determined for the NDA4 monitor of the Dounreay Pu waste measurement system. The measurements with polyethylene and graphite blocks have been carried out at Dounreay and interpreted at the JRC Ispra. Data were obtained on:

- mean escape probability
- escape probability spectra
- escape probabilities from item surfaces.

It should be mentioned that the results are sensitive to thermal neutron absorption properties of the sample. This effect could be eliminated by cadmium shielding of the detector assembly as provided in the EURATOM modular system.

Reference Monitor for Passive Neutron Assay

In addition to the already existing and prevalently applied correlation methods (V.D.C., autocorrelation) a more exacting pulse to pulse time correlation analysis was elaborated. This method allows the complete reconstruction of the detected neutron multiplicity.

The time distribution of two successive pulses of the first, second, ... order are measured.

A mathematical model was developed for theoretical prediction of the measured time distributions. The model is based on probability theory and on the assumption that the correlated fission neutrons decay in the ground-mode (described by a single exponential function). The theoretical predictions are fitted to the measured time distributions, varying the detected neutron multiplicity parameters.

Both hardware and software development on the signal processing unit has been continued. All of the purchased units were delivered. This new computerized system will be completed and set up in the next few months.

Parts of the upgraded detector system are under construction. When these parts are ready, an extensive series of measurements will be started in order to find an optimum for the efficiency and the flattening of the spatial response of the neutron detector assembly.

Reference Monitor for Passive Gamma Assay

The γ -spectrometer consists of a Ge(Li) detector (10% efficiency). The detector pulses are fed to a multichannel analyzer coupled to a PDP 11/5 computer. The measured spectra are analyzed by computer programs.

After the assembling and testing of the apparatus an extensive series of measurements have been started in order to determine the basic properties of the spectrometer:

- the energy resolution is about 2 keV at 1.33 MeV

- the intrinsic photo-peak efficiency falls from 10 to 1 between 100 keV and 2 MeV
- the Ge(Li) detector can be treated as a point detector. That is the measured intensity falls according to $I = I_0 (1/4 \pi d^2)$.
- the attenuation follows rigorously the theoretical prediction, that is, the measured intensity falls according to $I = I_0 \exp(-\mu \rho l)$. The attenuation coefficient of polyethylene was measured.
- same measurements have been carried out along a line at 21° away from the detector axis. Both the point source characteristics and the intrinsic efficiency did not show any change.

It follows that the escape probability from a waste item to a point spectrometer in case of homogeneous material and uniform source distribution is given by:

$$P \sim \int_V (1/4 \pi d^2) \exp(-\mu \rho l) dV$$

An essential part of the theoretical analysis in Chapter III of our Guide² «Passive Gamma Assay» is based on Mercur-3 calculations which uses this expression. Thus these calculations can be regarded as valid.

Active Neutron Assay

Chapter V of our Guide «Active Neutron Assay» has been written. It is going to be published as a EUR report². Design parameter studies on the Reference Monitor for Active Neutron Assay were started recently at the University of Lyon in the frame of a collaboration programme.

The calculations will be carried out by means of the TRIPOLI 02 code.

A technical meeting of specialists (International Meeting on Monitoring of Pu-Contaminated Waste) will be held on 25-28 September 1979 at the JRC Ispra. The aim of the meeting is to assess progress and to establish the direction of future work in the evaluation of practical systems for monitoring plutonium contaminated waste. The waste systems include both solid and liquid waste and systems which contain either high or low level gamma activity. The meeting seeks to promote an international exchange of information and results relevant to the successful implementation of Pu waste monitoring systems.

The four day meeting will include sessions for oral presentation, followed by sessions of working groups to assess the state of the art in particular areas of interest. The subjects for the working groups will be established after the contributed papers have been received. The findings of the working groups will be presented in a final plenary session. Individual contributions should cover specific fields of the Pu-waste monitoring complex or should outline national activities and programmes.

COLLABORATION WITH EXTERNAL ORGANIZATIONS

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

Contract with the Institut de Physique Nucléaire (University of Lyon) for design parameter study of the Reference Monitor for Active Neutron Assay.

Contacts with the Plutonium Fabrication Plant (CNEN, Casaccia, Italy) for setting up a monitoring system of Pu contaminated solid waste streams.

CONCLUSIONS

As far as the progress in interpretation of active and passive neutron measurement in the Integral Experiment at Dounreay is concerned, we think that we resolved the fundamental problems in an adequate manner. Therefore we shall be able to apply our interpretation model as soon as calibration measurements are executed and real waste measurements started.

The gamma spectrometer was operated all the time except during a short shut down period. The unit will be equipped with a fast printing unit in the near future. More progress was made than expected in the experimental verification of the interpretation model for passive gamma-assay.

The development of the upgraded reference monitor for passive neutron assay has been continued. Good progress was reached in the field of the active neutron assay as far as the theoretical model is concerned.

PLANNED ACTIVITIES

For the second semester 1979 the following activities are planned:

- 1) Integral Experiment
Continuation of the bench-mark and calibration measurements. Starting of measurements on real waste.
- 2) 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.
- 3) Passive Gamma Assay
Continuation of the experimental verification of the interpretation model.
- 4) Active Neutron Assay
Design parameter study of the Reference Monitor.

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(To be published in 1979)

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 were continued.

For the reporting period it was planned:

- to conclude denitration tests under simulated conditions (5000 l/t and concentrated HAW) and on fully active laboratory scale (5000 l/t HAW)
- to conclude HDEPH batch-extraction tests on fully active laboratory scale
- to continue the experiments on the separation of the actinides from RE on fully active laboratory scale
- to continue the engineering assessment of the proposed HAW partitioning processes as well as of the conceptual design of a partitioning plant.

RESULTS

Denitration Experiments

Fully active laboratory scale experiments were carried out using HAW solutions prepared at Ispra from UO_2 fuel samples irradiated at 26,000-36,000 MWd/t and cooled for 4 years.

In Table I are reported results of denitration tests carried out on simulated and fully active HAW solutions (5000 l/t) according to option 2 of the HDEPH process scheme¹.

Table I – Behaviour of Pu During Denitration of Unconcentrated HAW Solutions by Formic Acid

Run nr.	1	1	3	4
HAW solution (5000 l/t)	simul.	simul.	fully active	fully active
chemical form of Pu	ionic	poly- meric*	prob- ably ionic	prob- ably ionic
HCOOH/HNO ₃ molar ratio ³	2	2	~ 2	2.2
% Pu left in the supernatant solution	86	61	70	56
% Pu recovered by formic and nitric acid washings	14	39	28	43.4
% Pu left in the precipitate	< 0.1	< 0.1	?	0.6

* prepared according to reference [2]

By operating the denitration of simulated HAW with a HCOOH/HNO₃ molar ratio of about 2, it seems possible to maintain almost quantitatively the Pu in the extractable form. The following mechanism is tentatively proposed to explain the Pu behaviour during denitration: the lower retention of Pu into denitration precipitates at pH = 2 may be due to the reduction of Pu (IV) to Pu (III), even if Pu is initially present under polymeric form. This reduction is probably promoted by the formic acid decomposition in presence of Pd and Rh black acting as catalysts. Preliminary results of fully active runs (Table I) are comparable with those of simulated solutions although some Pu was lost to the precipitate. The higher Pu fraction adsorbed on the precipitate separated in the run N. 3 was probably due to the incomplete dissolution of Pu formate by the 4M HNO₃ wash.

The behaviour of Am and Cm was similar to that observed for Pu. Experiments are still in progress to optimize the operating conditions.

Denitration experiments on concentrated HAW solutions (about 500 l/t), as proposed by option 2 of the TBP process scheme³, were carried out until now using only simulated solutions. The low acidity value required for the separation

of the actinides from HAW could be reached by means of the denitration process provided an accurate acidity control is carried out during the process. The latter appears however to be a rather critical operation due to the accurate measurements to be remotely operated and requiring specially developed instrumentations.

As an alternative procedure a partial denitration up to about 0.5-1 M HNO₃ followed by a final adjustment to the required acidity value (0.1-0.2 M HNO₃) can be carried out.

By operating the denitration of the concentrated HAW solution (4-5 M HNO₃) with HCOOH/HNO₃ molar ratios in the range 1.3-1.5 a denitrated HAW solution, 1.5-0.6 M in nitric acid, is finally obtained.

Results from two tracer runs are reported in Table II. They show that, as in the case of unconcentrated HAW, even if polymeric Pu is added to the HAW, the latter is converted almost quantitatively during the HAW denitration to an extractable form. The acidity of resulting denitrated HAW solution has been further reduced to 0.1-0.2 M HNO₃ by addition of calculated amounts of Al(OH)₃ together with the usual salting agents (Al(NO₃)₃ and NaNO₃) required for TBP extraction. Fully active runs are scheduled for the second half of 1979.

In view of a possible utilization on an industrial scale, TBP and n-dodecane were substituted to mesitylene for experiments on fully active HAW.

Pu, Am and Cm separation yields, which appear to be sufficiently high considering that they correspond to a single extraction stage, confirm the values obtained with simulated solutions.

Solvent Extraction Experiments

HDEHP Process

Batch extraction tests on fully active laboratory scale have been continued. Typical results, obtained for the HDEHP process and reported in Table III, do not show significant differences between extraction tests performed on simulated and fully active HAW solutions.

Table III – Percent Separation Yields Obtained from HDEHP Batch Extraction Tests on Simulated and Fully Active HAW Solutions

Option	Elem.	Solvent	Simulated HAW		Fully active HAW	
			Acidity	% Extr. yield ¹	Acidity	% Extr. yield ¹
1	Pu	0.25 M HDEHP in mesitylene	4 M HNO ₃	>99.8	4 M HNO ₃	} ₃
	U			>99.9		
	Np			80 ²		
	Mo			92		
	Zr			99.5		
2	Pu	0.3 M HDEHP in n-dodecane	pH=2.11	>99	pH=2.76	} ₃
	Np			≥98		
	Ru			~0.5		
	Am			≥98		
	Cm			≥99		
	Ce			≥98		
	Eu			>99		

¹ after a single extraction stage

² cumulative, after 4 extraction stages

³ not measured

⁴ 0.3 M HDEHP-0.2 M TBP in n-dodecane as solvent

The pH dependence of the actinide and lanthanide extraction by the HDEHP/TBP solvent has been experimentally tested on fully active laboratory scale. In Figures 1 and 2 the measured values of distribution coefficients (D_A⁰) of Pu, Am, Cm and Ce, Eu are plotted versus pH values. The shape of resulting isotherms shows the considerable influence of the pH variation on the extraction degree which is practically of the same order of magnitude for all the considered elements.

As to the back-extraction of loaded HDEHP/TBP solvent (option 2), only simulated HAW solutions have been so far tested. Results have been reported in reference 4. Hot cell batch extraction experiments are continuing.

Instead of continuous countercurrent experiments, initially planned, HDEHP batch-countercurrent experiments have been started under simulated conditions to better precise some extraction and back-extraction parameters such as phase ratios, minimum number of stages of extraction/back-extraction and scrubbing steps etc.

Table II – Behaviour of Pu, Am, Eu and Ce During the Denitration of Concentrated HAW Solution by Formic Acid

HAW concentration factor	13.6			13.5			
Initial acidity (M/l)	4.6			4.7			
Final acidity (M/l)	0.64			1.12			
HCOOH/HNO ₃ molar ratio	1,5			1.5			
chemical form of Pu	polymeric			polymeric in presence of phosphates			
	Pu	Am	Eu	Pu	Am	Eu	Ce
% element in the supernatant solution	91.1	94.6	82.8	89.3	92.2	88	88
% element recovered by formic and nitric acid wash	7.4	5.38	7.18	9.85	7.75	12	12
% element left in the precipitate	1.5	0.02	0.02	0.85	0.05	n.d.*	n.d.*

* not detected

For this purpose an extraction system based on interchangeable extraction units has been developed at the Ispra laboratories with the aim of essentially reducing the volumes of organic and aqueous phases involved in each counter current experiment.

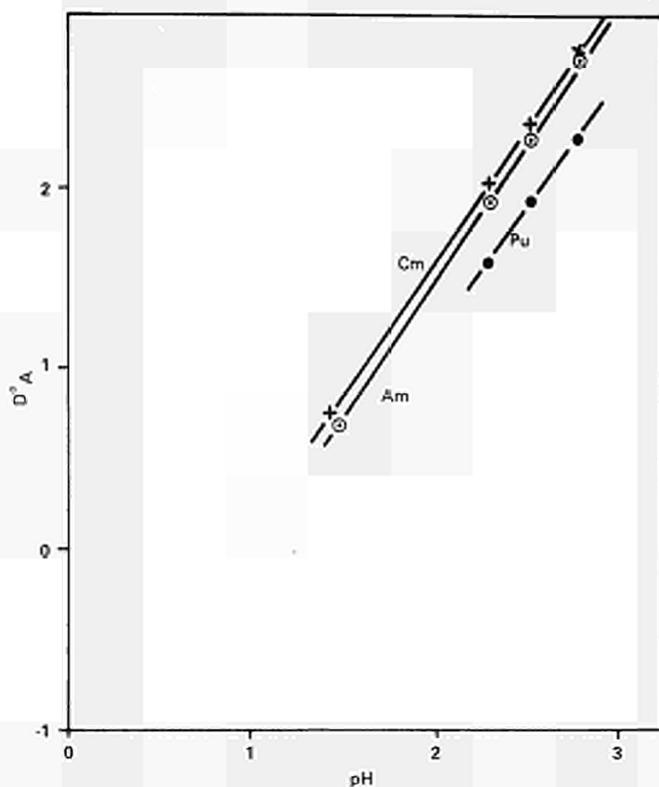


Fig. 1 – pH Dependence of the Am, Cm and Pu Extraction (D^0_A) by 0.3 M HDEHP - 0.2 M TBP in *n*-Dodecane from Fully Active HAW Solution (3,500 l/t)

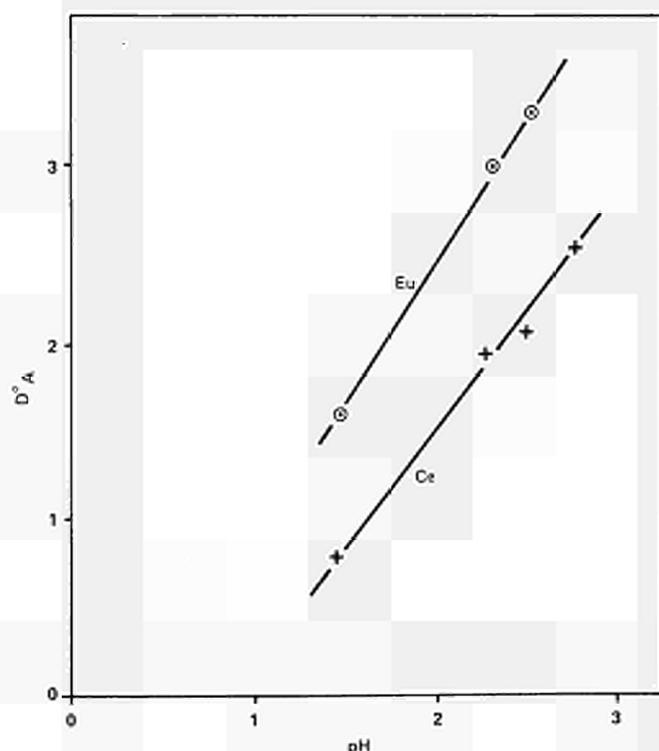


Fig. 2 – pH Dependence of the Ce and Eu Extraction (D^0_A) by 0.3 M HDEHP - 0.2 M TBP in *n*-Dodecane from the Fully Active HAW Solution (3,500 l/t)

Preliminary results indicated that Am (and RE) can be separated almost quantitatively from the simulated HAW solution (5000 l/t) using four extraction stages ($O/A = 1$) with two aqueous scrub stages ($O/A = 0.5$). More than 99% of Am can be subsequently stripped from the loaded HDEHP-TBP solvent using five back extraction stages ($O/A = 4$) with two organic scrub stages ($O/A = 1$).

TBP Process

Only batch experiments on simulated HAW solutions have been until now used to investigate the chemical feasibility of the extraction steps of the TBP process. Results have been already reported⁵.

The preparation of TBP batch-counter-current experiments using simulated HAW solution is in progress. In the experiments the concentrated HAW solution resulting from denitration tests will be used as feed. Tracer level runs are scheduled for the end of this reporting period.

OXAL Process Experiments

Actinide/RE partitioning experiments on fully active laboratory scale have been performed. Three extraction chromatography-based procedures were experimentally investigated, all involving column adsorption of Am-Cm-RE from nitrate solution, and subsequent selective elution of the actinides with an actinide complexing medium.

The procedures were as follows:

Scheme A (HDEHP-Levextrel):

- adsorption of actinides and rare earths from nitrate solution at pH 2
- elution of Am + Cm by 0.05M DTPA, 1M glycolic acid at pH 3
- elution of RE by 3M HNO₃

Scheme B (HDEHP-Levextrel)

- adsorption as in scheme A
- elution of Am + Cm by 3M LiNO₃ - 0.065M DTPA at pH 1.8
- elution of RE as in scheme A

Scheme C (TBP-Levextrel)

- adsorption of Am-Cm-RE from nitrate solution, suitably buffered by addition of an acid deficient Al nitrate solution (2M)
- elution of Am + Cm + heavy RE fraction by 5.4 LiNO₃, 0.05 M DTPA, 1 M lactic acid (pH = 1.8)
- elution of RE as in scheme A

Figure 3 shows the elution curve of scheme A as an example. Rather large elution volumes are necessary to obtain a high recovery yield for actinides, with considerable overlapping of the actinides and rare earths fractions. Although results are not conclusive, extraction chromatography does not appear as a promising technique for actinides-rare earths partitioning on an industrial scale.

Process Engineering Assessment

Solvent extraction processes

From the two proposed flowsheets that rely completely on countercurrent techniques for the actinide separation, the TBP process appears highly attractive for the well known and widely proven excellent chemical and technical performances of this solvent in the nuclear field. Its good hydraulic behaviour even at high loading rates makes it suitable for the processing of concentrated solutions. This process is therefore proposed to treat HAW after concentration to about 0.5 m³/t U.

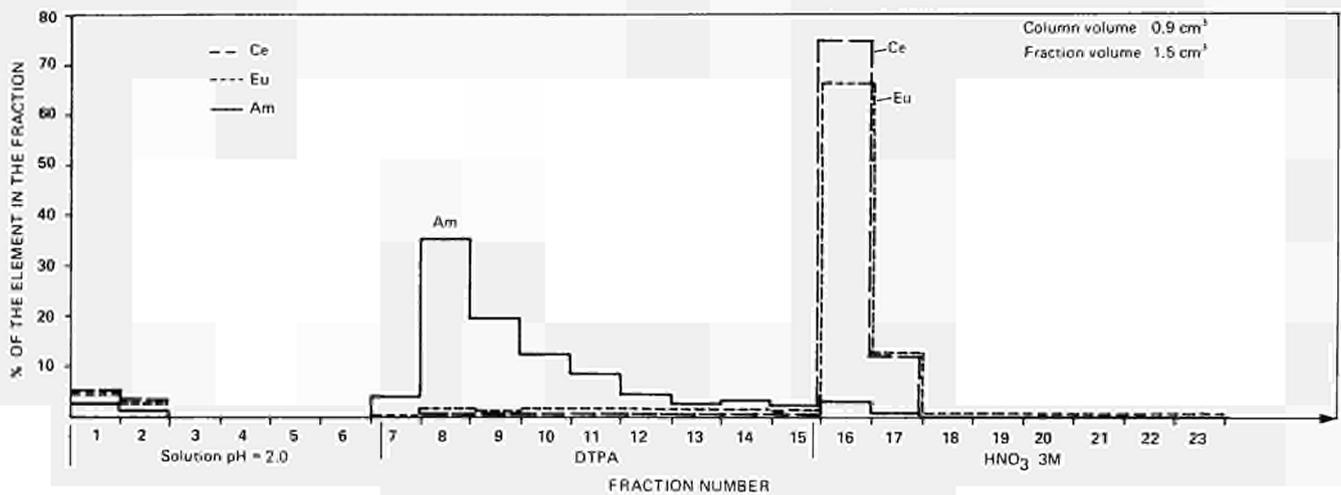


Fig. 3 – Separation Am/R.E. Using LEVEXTREL - HDEHP

In each of the two extraction-back-extraction steps for exhaustive high valent actinides and Am/Cm/RE separation, overall flowrates of about 1/20 of those of the corresponding Purex HA co-decontamination cycle would be required, resulting in considerable savings for equipment and plant layout. If reprocessing is performed after relatively short cooling an interim storage of the HAW is however required to allow for a sufficient FP decay. Virtually no active Zr/Nb should be present, since these elements are heavily extracted in the exhaustive extraction step, whereas in the Am/Cm/RE separation an advanced decay of Ce-144 would limit the radiation damage.

The latter step can be performed in all three proposed processes only at high pH, i.e. at low nitric acid concentrations. Denitration with formic acid in a batch mode operation is proposed.

This step must be considered critical as to corrosion risks on the equipment and to process control, to avoid on one side the reaction to run out of control (feed rates and temperature) and on the other side to reach too high a pH at the end (heavy precipitation). The TBP process requires at this point the addition of Al and Na nitrates as salting agents to enhance the Am-Cm extraction. The apparent draw-back of this process turns out as an advantage for the denitration step, since the latter can be stopped at about 0.5-1 M residual HNO₃ and this solution can then be adjusted to low free acidity by introducing the salting agents as an acid deficient Na-Al nitrate solution. Operating on concentrated HAW the added amount of these salts is fairly well compatible with that required in the glass making step. Therefore any undue dilution of the fission products is excluded.

Another unit operation required in this process is the separation of solids (feed clarification). The chemical conditions worked out should provide at least in the denitration step for an alpha-free precipitate ready for vitrification. The very straight targets (overall DF of the FP actinides 10³) would however require a very thorough washing of these precipitates, leading to important volumes of wash solution and to complex operation procedures. The analytical control for characterization of practically each batch of precipitate would be necessary in order to avoid the carry over of alpha contamination by a sub-standard batch. Certainly there is no reasonable way today to achieve this. It appears therefore advisable to consider these precipitates a priori alpha contaminated and to foresee their treatment together with other contaminated solids from the reprocessing operation like dissolver sludges etc. In this case the precipitates can simply be separated without washing.

The only serious draw-back of TBP in a waste management scheme including transmutation is the impossibility to separate the rare earths from the actinides. For this final step of partitioning other techniques or solvents must be applied.

The most popular among the latter for such a purpose is HDEHP, the solvent proposed by the JRC Ispra for the other, alternative countercurrent solvent extraction partitioning process.

Conceptually this process is very similar to the TBP process. Therefore only the main differences are pointed out here. They concern:

- The flow rates. The treatment of unconcentrated HAW means about 10-fold higher aqueous flow rates. The factor for the organic extractant is still more unfavourable taking into account the limited loading capacity of the HDEHP with respect to TBP. The consequences on equipment dimensions and plant layout appear evident.
- This process is foreseen for DIRECT HAW partitioning, i.e. for potentially short cooled HAW containing still considerable levels of ⁹⁵Zr and ⁹⁵Nb. Both elements are strongly extracted by HDEHP, a fact that will lead to heavy radiation damages, hence to operational difficulties and to a separation efficiency's reduction.
- No salting is required. Therefore any possibility of a less extended denitration with a final pH adjustment is excluded.
- Selective back-extraction operations on the loaded organic phase are required in the last step. In order to remove the RE in the second stripping section to the required specifications for residual alpha contaminants, it is essential that in the first stripping section Am-Cm have been back-extracted quantitatively and that residual Pu and Np are kept also quantitatively in the organic layer. Retention and/or entrainment phenomena would seriously compromise the feasibility of this process.

OXAL Process

The most significant difference of this process with respect to the foregoing processes is the possibility of separating an important fraction of FP from actinides, mainly the couples Zr-Nb and Ru-Rh, without requiring solvent extraction operations. This makes the process highly attractive for the early DIRECT partitioning. On the other side, the handling of highly active solids in sludge form is considered one of the most difficult problems in the reprocessing field. A thorough analysis of the problem shows, however, that by operating batch-wise, the denitration-precipitation as well as the

dissolution of the precipitates could be made both in the same reactor, avoiding any solids transfer and counter-washing operations.

The further separation of the actinides from residual FP, mainly Sr, Ba, and RE, or vice versa, can be performed in the most appropriate way by one of the extraction processes described above.

COLLABORATION WITH EXTERNAL ORGANIZATIONS

To verify on fully active laboratory scale the proposed HDEHP and TBP flow-sheets by continuous countercurrent experiments, based on the use of mixer-settler batteries, a collaboration contract between CEC and CEA (CEN, Fontenay-aux-Roses, France) is being set up. The starting of the experimental work to be performed at CENFAR laboratories is foreseen around September 1979.

CONCLUSIONS

The project activities are generally in progress according to the planning. However, a certain delay is being accumulated in some hot cell experiments on fully active HAW solutions. This is in general due to the frequent time consuming operations performed inside remote handling facilities, the cadence of which cannot be accelerated for reasons of security and health physics protection. As consequence the completion of HDEHP batch extraction and denitration experiments on fully active laboratory scale is postponed to the next period.

PLANNED ACTIVITIES

During the next six months it is planned:

- to conclude HDEHP batch-extraction and denitration tests on fully active laboratory scale
- to conclude batch-counter-current experiments on fully active HAW by HDEHP and 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 actinide separation.

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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 and the cost and risk implications of the transmutation of actinides other than fuel in fission power reactors and to propose a suitable recycle strategy.

The activity is subdivided into the following main studies :

- Reactor physics calculations
- Fuel element design
- Cost and risk analysis
- Study of the implications on the fuel cycle

In this report the progress in reactor physics calculations for LWRs, fuel element design and risk analysis are discussed. The results of the studies will be included in a summary report ready for the beginning of 1980.

RESULTS

Reactor Physics Calculations for LWRs

The reactor physics calculations for the assessment of the actinide transmutation in LWRs have been continued with the use of the zero-dimensional RIBOT-5 code¹.

Aim of these calculations is to determine by means of parametric studies the actinide build-up vectors and transmutation rates under different recycling conditions and to verify the calculational results derived by ORIGEN. Parameters as the reactor cycle length, specific power (i.e. flux density), neutron spectrum, fuel temperature, cooling time, spatial burn-up distribution, nuclear data uncertainties, etc., are being investigated in order to assess their influence on the actinide vectors and on the reactor balance.

The results so far achieved have been reported in internal working papers^{2,3,4} and are only partly presented here, together with some new results.

In Table I the characteristics of the considered light water reactors are shown.

Table I – Characteristics of the Considered Light Water Reactors

	BWR	PWR
Thermal power	2650 MW	3000 MW
Average core power density	55 kW/l	93 kW/l
Fuel inventory (heavy metal)	104.4 MTU	82.8 MTU
Average ²³⁵ U enrichment	2.74%	2.7%
Discharge burnup	27.5 GWD/MTU	33 GWD/MTU

In Table II the actinide isotope vectors from the 1st and 35th cycles of a BWR-U and a PWR-U are indicated, as calculated by RIBOT. The recycling scheme is very simple : all self-generated by-product actinides are recycled homogeneously, with the exception of ²⁴²Cm. The beginning of cycle (B.O.C.) core reactivity is kept constant by varying the ²³⁵U B.O.C. enrichment.

Together with actinide build-up the equilibrium factors are given, defined as the ratios between equilibrium cycle build-up values divided by 1st cycle build-up values of by-product actinides.

Table II – Build Up (kg/MTU) of Some Actinide Isotopes from BWR-U and PWR-U

	BWR-U			PWR-U		
	Cycle 1 Output	Cycle 35 Output	Equilibrium Factor: Weq/W1° cycle	Cycle 1 Output	Cycle 35 Output	Equilibrium Factor: Weq/W1° cycle
²³⁷ Np	3.61-1*	8.00-1	2.22	3.45-1	7.16-1	2.07
²³⁸ Pu	4.58-2	1.14-1	2.49	4.85-2	1.17-1	2.41
²⁴¹ Am	3.97-2	5.21-2	1.31	2.61-2	3.21-2	1.23
^{242u} Am	6.57-4	8.85-4	1.35	4.12-4	5.14-4	1.25
²⁴³ Am	1.31-1	2.02-1	1.54	1.64-1	2.54-1	1.55
²⁴³ Cm	1.25-3	2.48-3	1.98	1.00-3	1.77-3	1.77
²⁴⁴ Cm	3.35-2	4.25-1	12.7	6.06-2	5.74-1	9.47
²⁴⁵ Cm	2.28-3	3.08-2	13.5	1.88-3	2.71-1	14.4
²⁴⁶ Cm	3.60-4	2.72-1	755.5**	4.05-4	3.67-1	897.3**

* 3.61-1 = 3.61 × 10⁻¹

** ²⁴⁶Cm not yet at equilibrium

In Table III the build-up of by-product actinides are given for the 1st and 14th cycle for a Pu-fuelled BWR. Reactor characteristics are the same as those indicated in Table I. The recycling mode is slightly different, being in this case the cycle time kept constant at 1090 days with a burn-up of 27.7 GWD/MTU. The chosen fuel composition corresponds to a loading of 1/3 of the fuel elements charged with 5% of equilibrium recycled plutonium (fuel plutonium) of the following composition, kept constant at the beginning of each cycle :

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

The ²³⁵U B.O.C. composition is determined such to have an end of cycle K_{eff} practically equal to 1, in order to have a good fuel utilization.

Table III – Build Up (kg/MTU) of Some Actinide Isotopes from BWR-Pu

	BWR-Pu		
	Cycle 1 Output	Cycle 14 Output	Equilibrium Factor: Weq/W1° cycle
²³⁷ Np	2.51-1*	5.10-1	2.03
²³⁸ Pu	6.33-2	1.12-1	1.77
²⁴¹ Am	1.52-1	1.78-1	1.17
^{242u} Am	2.75-3	3.26-3	1.19
²⁴³ Am	8.96-1	1.18	1.32
²⁴³ Cm	6.71-3	1.08-2	1.61
²⁴⁴ Cm	7.07-1	4.01	5.67
²⁴⁵ Cm	4.50-2	2.93-1	6.51
²⁴⁶ Cm	9.37-3	1.25	133.4 **

* 2.51-1 = 2.51 × 10⁻¹

** ²⁴⁶Cm not yet at equilibrium

The B.O.C. ²³⁵U enrichment is plotted in Fig. 1, showing a net decrease from the 1st to the 14th cycle, due to the high and increasing content of by-product fissile actinide (mainly ²⁴⁵Cm).

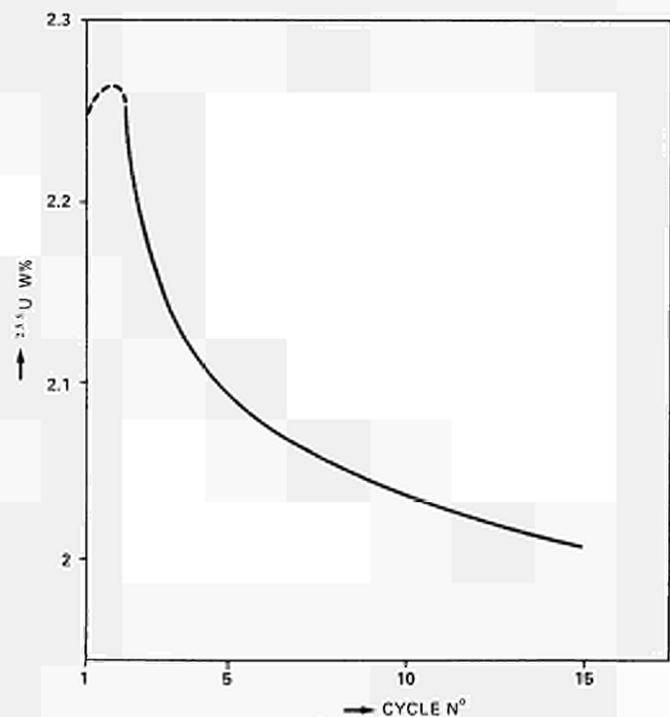


Fig. 1 – BWR - Pu Recycling: Beginning of Cycle ²³⁵U Enrichment with Constant Cycle Length

Spatial flux calculations within a PWR fuel element, presented in Fig. 2, were performed by the aid of EREBUS. The result is shown in Fig. 3. It indicates that heterogeneous recycle of by-product actinides in the centre of a subassembly is hardly a good choice. In fact, the strong flux depression in the centre of the fuel element would result in a very low transmutation rate. An alternative could be to recycle by-product actinides heterogeneously in single pins situated at the edge of the fuel element.

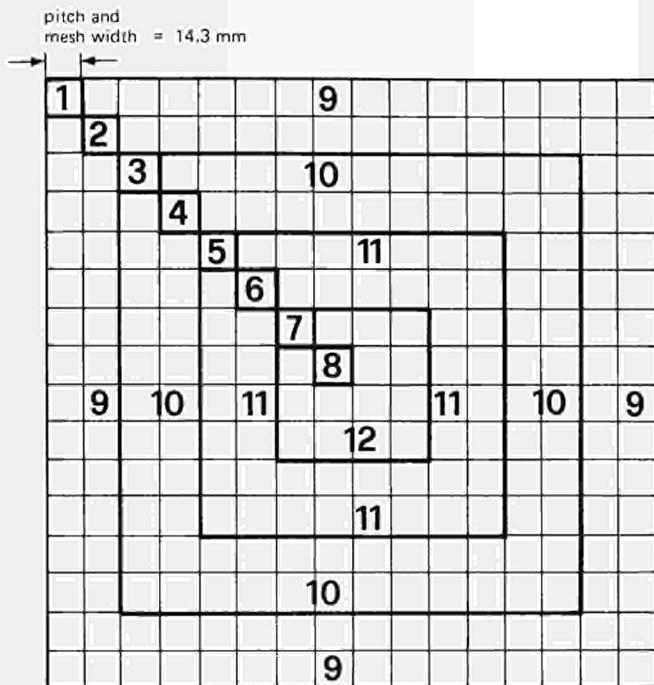


Fig. 2 – PWR Fuel Element Geometry for EREBUS Calculations. Regions are Numbered from 1 to 12

Temperature and Oxygen Profiles in FBR Target Pins

Several geometric configurations of target pins for actinide recycling in a fast flux have been proposed in previous Programme Progress Reports. Because of the hard neutron spectrum, the actinide recycling in FBRs does not impose the limitation presented by the reactivity penalty in LWRs.

As far as the fabrication is concerned, heterogeneous recycling, a strategy requiring the fabrication in ad hoc facilities of special pins or sub-assemblies containing actinides, should be an incentive because the fabrication of special fuels could be limited only to a fraction of the total charge of the reactors. When actinides (Np-Am-Cm) are placed in a fast neutron flux, the heat generation can produce a central temperature prohibitive for the life of the targets. Such a situation is linked to the thermophysical characteristics, as thermal conductivity, λ , of the actinide oxides and porosity of the material. The temperature will influence phenomena as mechanical stresses in the fuel and cladding, fuel restructuring, oxygen redistribution and fuel-cladding chemical interactions (FCCI).

When Np-Am-Cm oxides are considered to be recycled in a FBR in which the severe operation conditions (linear power, W/cm, and surface cladding temperature) can influence the integrity of the fuel element, calculations of radial temperature profile and oxygen distribution is a prerequisite in order to predict the behaviour of the targets. Calculations of temperature profile and oxygen distribution have been performed by TPROF and CODACT computer programs.

Temperature profiles calculations

The scope is to guarantee that the maximum temperature, T_m , in the center of the target does not exceed the melting point of the fuel material.

Various geometric configurations have been considered. The annular configuration presents an inner temperature, T_i , of 1840°C. All other geometric configurations have given unacceptable values of T_i .

In Table IV the geometry, thermophysical properties and composition for the annular configuration are reported. The annular configuration for actinide recycle is similar to the LOWI design developed for LWR fuel pin, shown in Fig. 4⁵. This arrangement in which the inner pellet is made by natural or depleted uranium and the ring element by enriched uranium oxides, presents an improved performance with respect to mechanical interaction and central temperature. Therefore the ring containing actinide oxides could surround a co-axial pellet made by an inert material such as MgO.

The temperature profiles of Fig. 5 have been calculated for the value of thermal conductivity given in Table IV and for two other values (-20% and +10% respectively) to take into account the uncertainties deriving from the lack of experimental data for the thermal conductivity as a function of oxide mixture and oxygen-to-metal ratio. The detailed results on the radial temperature profiles are reported in an internal working paper⁶.

Oxygen redistribution

In the UO_2 - PuO_2 system the oxygen released during irradiation can increase the concentration at the outer part of the pellet reaching the oxygen potential, ΔG_{O_2} , of the stainless-steel used as cladding material.

Oxygen and caesium a fission product, are responsible for the intergranular attack of the cladding inducing chemical reactions between the nuclear materials and the elements present in the cladding (caesium chromates, caesium

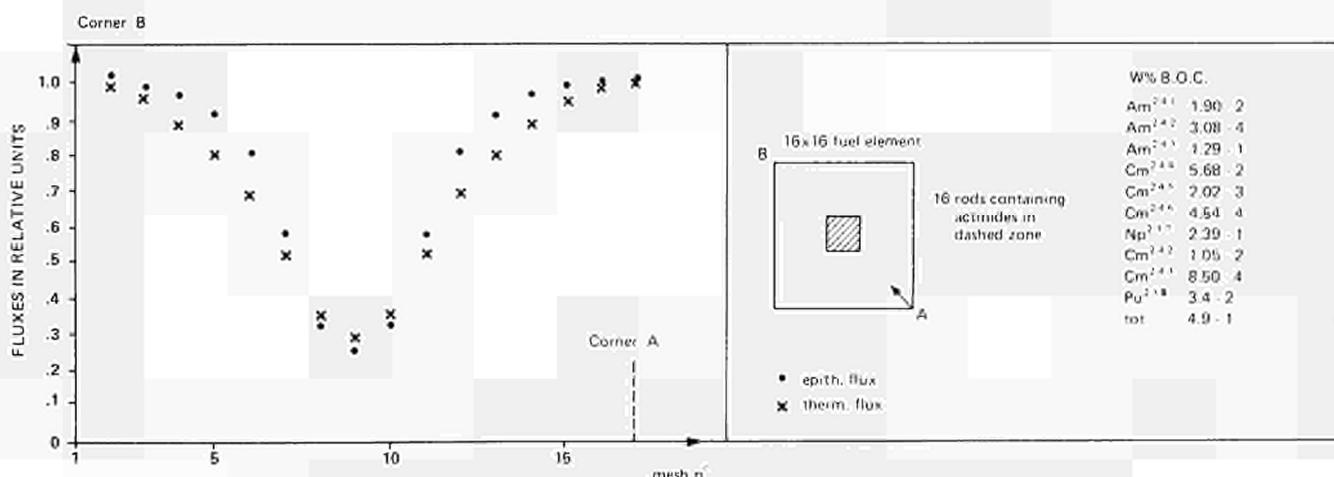


Fig. 3 – Flux Depression in PWR Fuel Bundle due to the Presence of 0.49% of Actinides

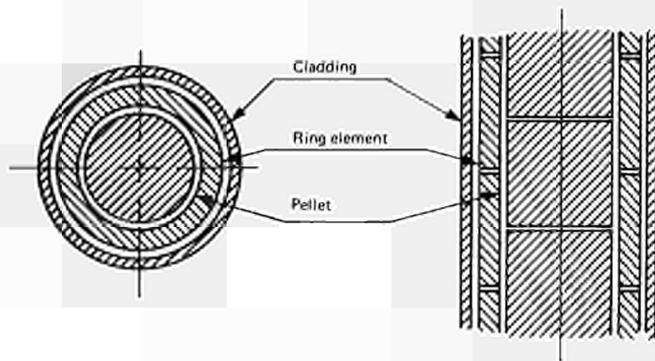


Fig. 4 - The New LOWI Fuel Design

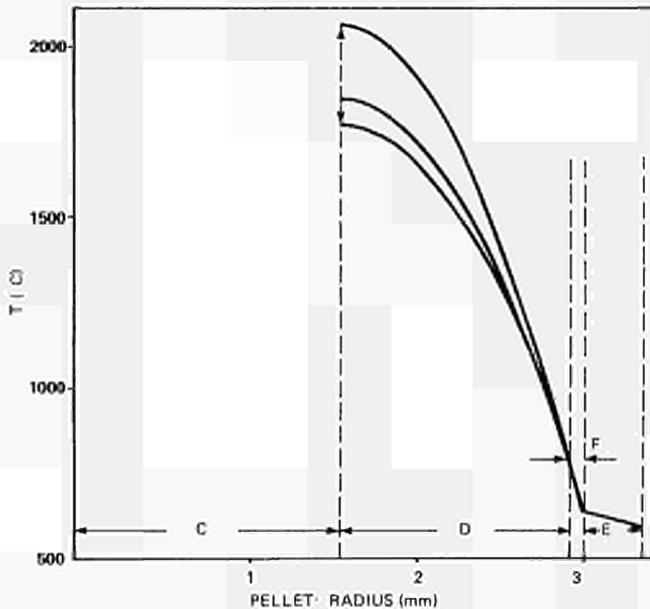


Fig. 5 - Annular Pellet Configuration for Np-Am-Cm Oxides: Radial Temperature Profiles Calculated Using Different Values of Thermal Conductivity. C = hole radius; D = pellet thickness; E = cladding thickness; F = pellet-cladding gap

uranate and caesium plutonate). The penetration of chemical products leads to a reduction of the wall thickness of the cladding, weakening the mechanical properties by a stress corrosion cracking (SCC) mechanism. The oxygen potential above a threshold value is a condition for cladding attack and interaction between fuel and cladding. This interaction is initially negligible in the case of hypostoichiometric fuel obtained by lowering the Pu⁴⁺ valency in the sintering process.

Radial oxygen profiles have been calculated for the annular pellet configuration. The results are shown in Fig. 6 as a function of burnup. The maximum value of the oxygen-to-metal ratio (O/Np + Am + Cm) is 1.973. At this value of O/M, the (U-Pu)O_{2-x} system presents $\Delta G_{O_2} = -163$ Kcal/mole O₂ at the temperature of 800°C calculated at the outer part of the pellet compared to the -120 Kcal/mole O₂ for cladding oxidation.

Thus, if the thermodynamic properties of Np-Am-Cm oxides are similar to those of the U-Pu oxides, the cladding corrosion would be negligible.

Table IV - Composition, Thermophysical Properties and Geometry of Actinide Pellet Annular Configuration

Atomic fraction	Np	= 0.727
	Am	= 0.216
	Cm	= 0.057
Oxygen-to-metal ratio		= 1.927
(O/Np + Am + Cm)		
Oxyde weight (g/cm)		= 1.933
Thermal conductivity		= 0.0393-1.55E-05t+4.907E-09t ²
Melting point (°C)		= 2440
Theoretical density		= 11.24
(g/cc)		
Radial density (%TD)		= 90
Cladding outer diameter		= 6.7 mm
Cladding inner diameter		= 6.0 mm
Cladding thickness		= 350 μ
Pellet outer diameter		= 5.85 mm
Pellet inner diameter		= 3.146 mm
Gap		= 75 μ
Linear power (W/cm)		= 566
Cladding outer temp		= 590
(°C)		

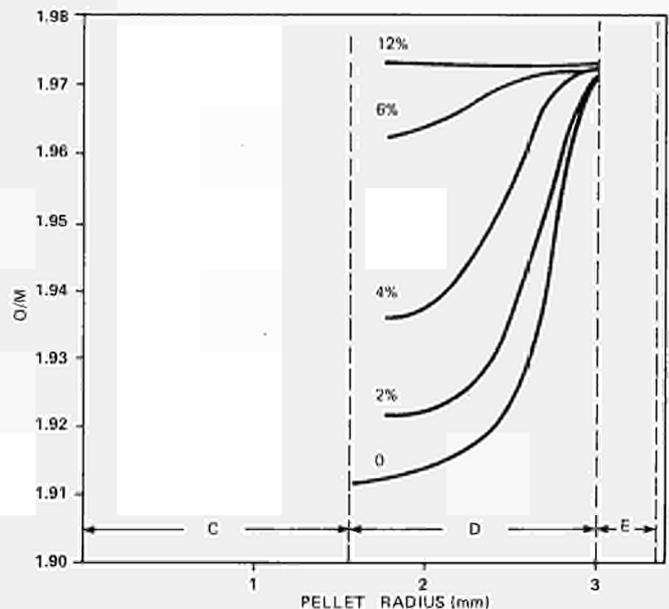


Fig. 6 - Radial Oxygen Profiles Calculated for Annular Pellet Configuration. Initial Composition Np0.727, Am0.216, Cm0.057, O1.927. C = hole radius; D = pellet thickness; E = cladding thickness. Parameter is the burn-up in atom %

Risk Assessment

In the field of risk assessment the releases of radioactive material to the biosphere have been estimated for the reference fuel cycle and the cycle with by-product actinide recycling. A 1 GWe Pu-fuelled LMFBR was considered, linked to a fuel reprocessing and fabrication plant and a high level waste solidification plant. The release contributions of other nuclear fuel cycle supporting plants were neglected. The isotopic composition for the fuel with and without by-product actinide recycling was calculated with the ORIGEN code⁷. These data were used as input in MARYON-II⁸ for the

assessment of operational releases and the accumulated releases for a nuclear power economy lasting about 400 years. The release factors for the isotopes in the different fuel cycle plants, relating the isotope flow to the releases, were taken from literature and are in general values, which are considered as target values to be obtained in future installations⁹.

Table V gives the estimated annual releases from the most important fuel cycle plants to air per GWeY generated. The loss fraction to the HLW was assumed to be 10^{-3} for the fuel actinides and 1 for the by-product actinides and fission products in the reference fuel cycle. In the by-product actinide recycling case all actinides had the loss fraction 10^{-3} .

The activity of the fission product releases dominates the actinide releases by orders of magnitude. The most important release source is the reprocessing plant. The released activity of the actinides is slightly smaller recycling actinides in the equilibrium recycle conditions as compared with the reference fuel cycle. This astonishing result could be reversed in case the release factor of the HLW solidification plant would be reduced from 10^{-10} to 10^{-11} or by assuming a higher loss fraction for actinides from the HLW. The same result is found for the ingestion and inhalation hazard.

Figs. 7 and 8 give the accumulated ingestion hazard of all actinides and fission products released to air as a function of time for the most important plants of the reference fuel cycle and the cycle with by-product actinide recycling, respectively. The time 0 is at the end of a 400 years lasting nuclear power duration in which an energy of 400 GWeY was generated. The decay of the ingestion hazard is dominated in both fuel cycles by the I-129 releases of the reprocessing plant. The hazard of the fabrication plant releases increases as function of time mainly due to daughter products of original U-releases. The release factor for uranium are in general in the literature given as 10^{-6} . In order to bring the hazard of the fuel fabrication plant to the order of magnitude of the other fuel cycle supporting facilities the release factor for U should be reduced to about 10^{-9} . This makes sense only in case that the I-129 and Tc-99 releases of the fuel reprocessing plant can be reduced correspondingly.

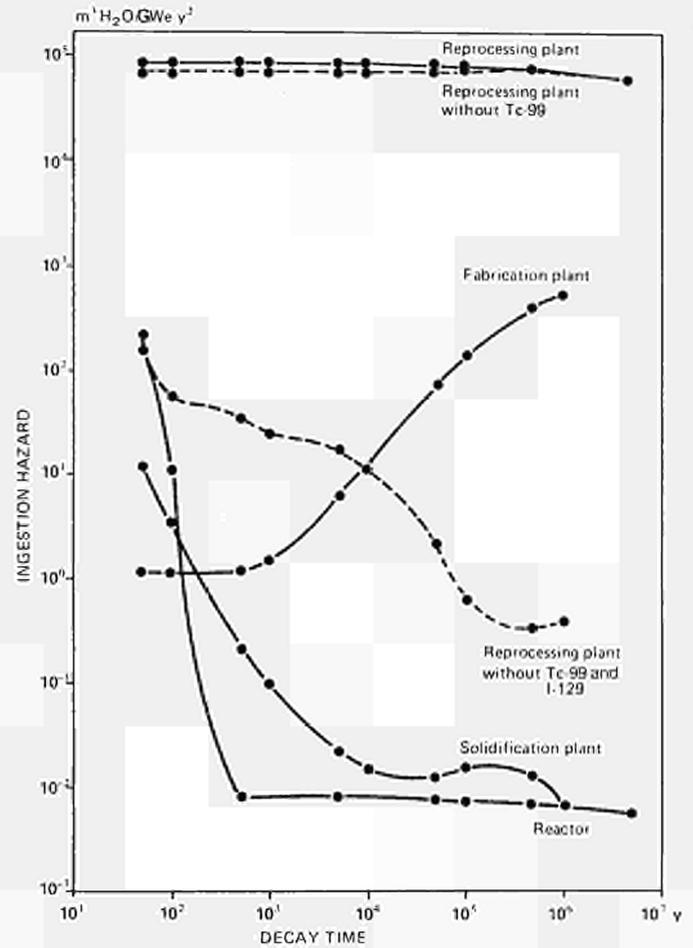


Fig. 7 – Accumulated Ingestion Hazard of Releases from the Most Important Fuel Cycle Plants to Air for the Ref. Fuel Cycle

COLLABORATION WITH EXTERNAL ORGANIZATIONS

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

Table V – Summary of Annual Releases from the Most Important Plants of the Fuel Cycle to Air. Values Refer to End of Reactor Operation Cycle, During Time of Reprocessing, Fabrication and Waste Solidification

	Activity [C/GWe y ²]			Ingestion Hazard [m ³ H ₂ O*/GWe y ²]			Inhalation Hazard [m ³ air*/GWe y ²]		
	Fission Products	Actinides		Fission Products	Actinides		Fission Products	Actinides	
		All Cycles	Ref. Cycle		By-product Recycle 216	All Cycles		Ref. Cycle	By-product Recycle 216
Reactor	4.33+2	—	—	2.33+5	—	—	2.30+9	—	—
Fuel Reprocessing	2.12+3	2.09-5	3.20-5	3.09+8	4.27-1	9.68-1	7.21+9	1.07+7	1.87+7
Fuel Fabrication	—	2.15-5	3.35-5	—	3.87-1	1.10	—	1.03+7	1.89+7
HL Waste Solidification	6.70-3	5.24-5	1.2 -6	5.80+2	2.41	3.39-2	1.53+7	3.27+7	2.19+6
TOTAL	2.55+3	9.48-5	6.67-5	3.09+8	3.22	2.10	9.53+9	5.37+7	3.98+7

* Hazard indices used are based on 0.5 rem/y

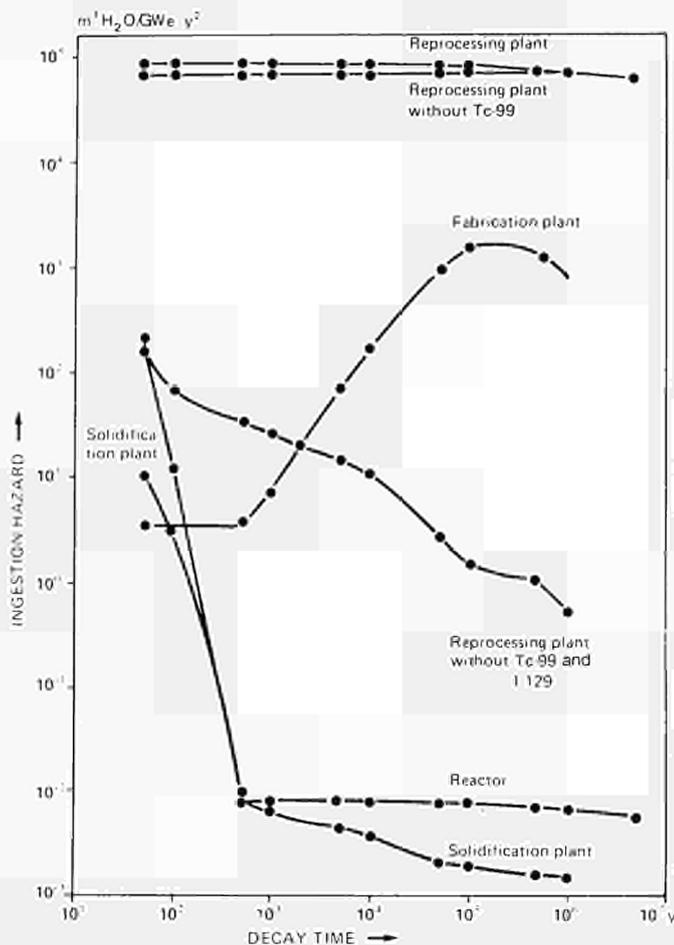


Fig. 8 - Accumulated Ingestion Hazard of Releases from the Most Important Fuel Cycle Plants to Air, Recycling By-Product Actinides

CONCLUSIONS

The work is proceeding in the various areas in a satisfactory agreement with the planning.

Considerable progress has been made in the reactor physics calculations for LWRs.

The first progress report on the FBR calculations has been presented by CNEN Casaccia.

The first progress report on the calculations for thorium-fuelled reactors has been presented by KFA Jülich. KFA Jülich has asked for a prolongation of 3 months of the time period agreed for the termination of the work.

In the field of fuel element design for FBR, calculations of temperature profiles have been carried out for various configurations. The results of the calculations indicate that an annular configuration make possible to maintain the temperature at acceptable values. The problem of the stability of the annular configuration during the irradiation time is under discussion.

The activities concerning the fuel element design for LWRs and the cladding/fuel interactions are in progress.

In the field of risk assessment interesting results have been obtained on the contributions of fission products and actinides to the hazard produced by the release from the nuclear plants. The contribution of the fission products appears to be dominant both in the reference fuel cycle and in the cycle with by-product actinide recycling.

PLANNED ACTIVITIES

In the next semester the assessment studies on transmutation will be terminated in a preliminary way. The expected results are :

- Definitive by-product actinide generation rates in typical HTRs, FBRs, U-fed LWRs, and Pu-fed LWRs derived from improved reactor-physical calculational techniques (partly obtained by contracts).
- Influence of by-product actinide target elements on flux and power distribution as well as on reactivity.
- Transmutation rates for these reactors.
- Definitive proposal for target fuel elements in FBRs and LWRs, including temperature and oxygen profiles.
- Parametric evaluation of additional fuel cycle cost due to by-product actinide recycle.
- Establishment of the nuclear power scenario for the EC on which the recycle strategy will be based.
- Elaboration of a definitive recycle strategy for by-product actinides.
- Extension of the risk assessment methodology aiming at quantifying the effect of releases on mankind.
- Preliminary proposal for the content of the final report on transmutation as an alternative high-level waste disposal scheme.

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Actinide Cross-Section Measurements

OBJECTIVES

The activity concerns the preparation and the execution of different experiments for the measurement of the fast-neutron $\sigma(n, \gamma)$ and $\sigma(N, t)$ of ²⁴³Am, which will be performed at the pulsed Van de Graaf at the Institute for Applied Nuclear Physics of the KFK Karlsruhe.

The $\sigma(n, \gamma)$ measurement will be performed with a gram quantity of ²⁴³Am. Due to the very high gamma background from the natural activity, appropriate gamma detectors (Moxon-Rae) with additional shielding will be used. In order to correct for the fission gamma-ray component admixed with the capture gamma-ray spectrum, the experiments will include also a simultaneous determination of the fission rate, by means of a gamma insensitive neutron scintillation counter (PSD).

Measurements of $\sigma(n, f)$, relative to ^{235}U , are foreseen with different methods. The fission neutron detection method, using the same (PSD) technique and a thick target, is efficient and suitable especially in the subthreshold energy range, but there are problems due to the uncertainty in the estimate of ν and of the fission neutron spectrum, relative to ^{235}U . However, measurements based on the detection of coincident fission neutrons could provide the normalization to ^{235}U , at particular points. These measurements require neutron detectors with improved neutron efficiency and gamma ray discrimination. A fission fragment detection method is also considered, for the measurements of $\sigma(n, f)$ near and above the fission threshold. This method is based on the use of gas discharge chambers, intrinsically insensitive to alpha particle background and allows a relative measurement of thin ^{243}Am and ^{235}U targets, using a simple double chamber configurations.

RESULTS

In this period, the activity has been devoted to the design of the experiments and to the preparation of the instrumentation. Different parts of the conventional instrumentation have been realized in our laboratory.

In particular, a considerable effort has been spent to develop an improved scintillation neutron detector with gamma-ray discrimination (PSD). This new device allows to record recoil proton pulses of amplitude greater than 20 keV electrons, with a high rejection ratio of gamma-rays as shown in Fig. 1. Therefore the counter is highly efficient over the fission neutron spectrum. (A standard system, available from the trade, of the kind used for ^{243}Am $\delta(n, f)$ measurement, is inefficient for neutrons below 2 MeV). In addition, our counter operates with a submicrosecond

resolving time and a high stability, which is mostly important for the intercomparison of ^{43}Am and ^{235}U measurements that are taken in different periods.

COLLABORATION WITH EXTERNAL ORGANIZATIONS

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

CONCLUSIONS

The basic instrumentation which has been set up allows to start the measurements of $\sigma(n, \gamma)$ and successively of $\sigma(n, f)$ (single neutron detection method), as soon as the ^{243}Am target and the V.d.G. machine time will be at disposal. The improved neutron detection technique allows to improve and to extend the experimental possibilities in the neutron cross-section measurements and could be useful also in actinide waste measurements of different kind (discrimination of alpha and beta activity, in the internal counting technique, e.g.).

PLANNED ACTIVITIES

The preparation of the different experiments will continue. The $\sigma(n, \gamma)$ measurements and the data treatment should be completed by the end of September.

The $\sigma(n, f)$ measurements should then start and measuring cycles with different configurations of the neutron detection method will extend till the end of the year. In the meantime, the fission fragment detection method will be developed and tested in preliminary measurements.

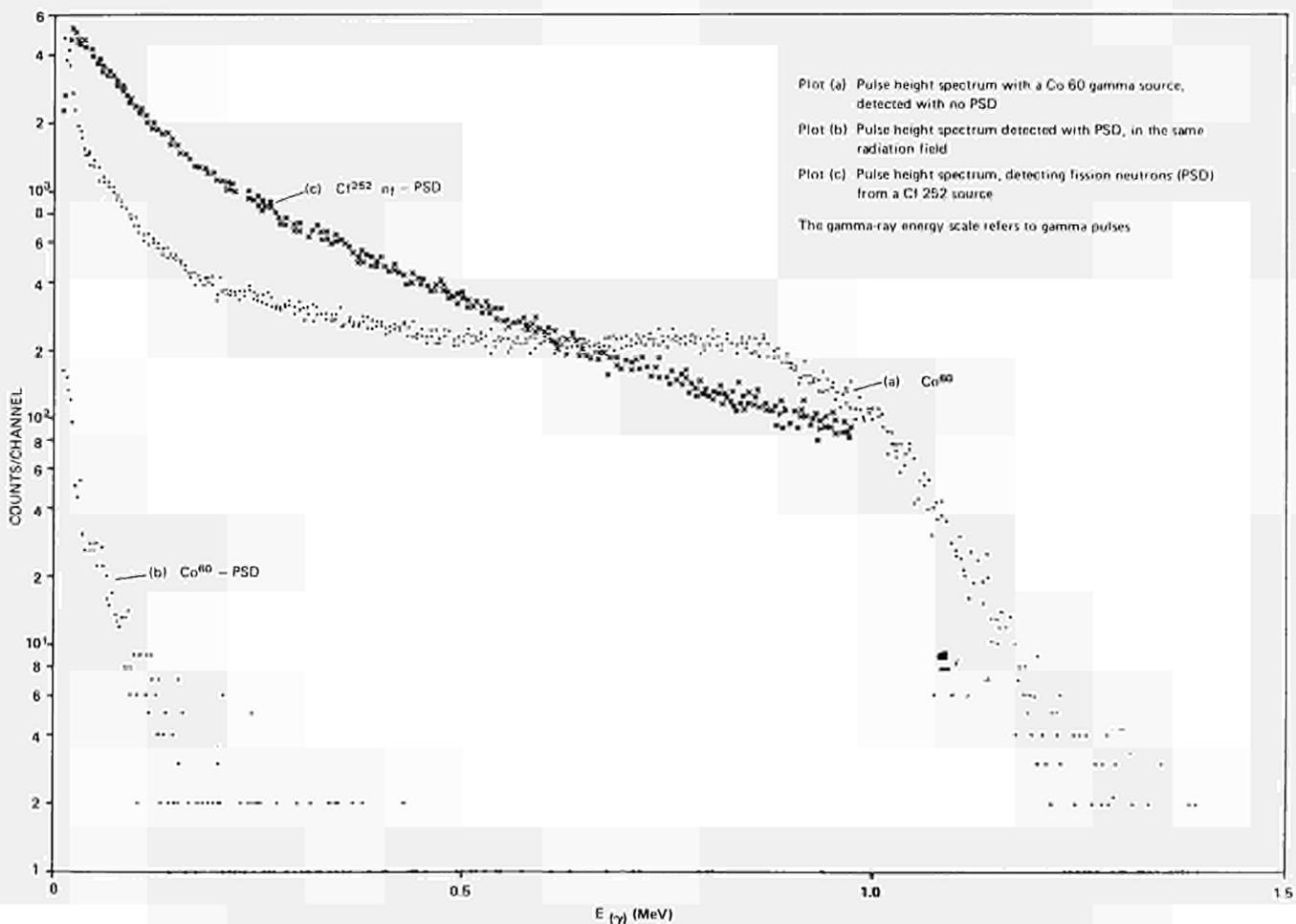


Fig. 1 – Results of the Improved Pulse Shape Discrimination Spectrometer, Using NE 213 Liquid Scintillator

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 :

- Chemical decontamination
- Physico-chemical structure of oxide layers
- Decontamination by redox cycling

Also an evaluation on the interest of the decommissioning of the Ispra I reactor, as a test case, is part of this activity.

RESULTS

Chemical Decontamination

The APAC process is extensively investigated at the JRC Ispra, with the aim to minimize the amount of final sludges generated by the chemical decontamination. The process consists of three stages :

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

a) Systematic Studies on Chemical Decontamination

The efficiency of the decontamination as a function of the concentration of the chemical products utilized in the APAC process, has been investigated on two types of samples :

- Samples obtained by oxidation of AISI 304 L surfaces, at 600°C in overheated steam. These samples are considered representative of surfaces with a long time of exposure in the reactor.
- Contaminated pieces from power stations, used essentially to verify the results obtained on inactive samples.

In the routine application of the APAC process, concentrations of 20, 30 and 20 wt% respectively, are normally utilized in the three stages at a temperature of 80°C.

A series of experiments were performed on the first type of samples, using different concentrations but the same ratio between concentrations as in the normal process.

It has been observed that satisfactory decontamination can be achieved with concentrations as low as 8, 12 and 8 wt%. Below these concentrations the decontamination factors were not acceptable.

A verification performed on active samples have given a decontamination factor of about 30, in agreement with the results obtained on the artificial samples.

Due to the fact that the permanganate gives the highest contribution to the sludge formation a series of tests was carried out to minimize its concentration in the second stage of the APAC process, maintaining at 8 wt% the concentration of the Citrox solution.

The tests on the artificial samples have shown that it is possible to decrease the permanganate concentration to 6 wt%, still maintaining a satisfactory decontamination factor. These results were confirmed by verification on active samples.

Some tests have been performed at 95°C in order to evaluate a possible influence of the temperature. No significant differences with respect to 80°C were observed.

b) Mechanisms of Decontamination Processes

During the reporting period the mechanism of the decontamination process has been particularly investigated by SEM (scanning electron microscope) analysis of specimens, exposed to 5000 h of oxidation in water at 300°C, in a test loop of the Thermal Exchange Service of the JRC Ispra.

These specimens are representative of surfaces oxidized in primary circuits of LWR.

From the SEM analyses, carried out before and after application of the various steps of the APAC process, following the procedures optimised in the systematic studies, the following conclusions were drawn :

- Before treatment the specimens are covered by an oxide film composed of two layers.
- After the first acid treatment the external layer is almost totally removed.
- After the treatment with the alkaline permanganate, the morphology of the internal layer is only slightly changed.
- After the second acid treatment the oxide is totally removed and a bright metal surface appears.

Physico-Chemical Structure of Oxide Films Formed in High Temperature Water on Austenitic Alloys

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 the last Programme Progress Report (July-December 1978).

Because of certain inconsistencies between results obtained with SIMS and those obtained with AES and SEM microprobe analysis, the SEM microprobe studies were intensified during the reporting period in order to get statistically confirmed data, about elemental concentrations in the oxide film at different locations.

The results obtained confirm the concentration values obtained with AES. On the basis of these results some conclusions, previously drawn on the possibility of application of SIMS, have to be revised.

The results of the analytical work on the surfaces oxidized in hydrogenized water will be published in a paper which will report :

- 1) Confrontation of the data obtained with the various analytical methods (AES, SIMS, SEM)

- 2) Local variations of elemental concentration ratios, enrichment and depletion of the various elements, concentration of cation vacancies in the oxide
- 3) Change of elemental ratios in the metal phase in vicinity of the metal oxide interface
- 4) Confirmation of SIMS as the only analytical tool for the measurement of cobalt concentration profiles in the oxides formed on austenitic steel in water at 300°C.

The second part of the study dealing with the oxides formed on austenitic steel at 300°C in oxygenized water is in progress. A part of the SEM morphological studies has been already concluded. The AES and SIMS experiments have been already carried out under contract by Dornier System GmbH and the evaluation of the data will soon be concluded.

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. As known, these effects were used in CANDU reactors for decontaminations with low decontamination factors. Cycling experiments in a test loop were prepared in order to verify these indications.

Decommissioning of the Ispra I Reactor

The Ispra I reactor is a 5MW research reactor which had been in operation more than 10 years. It is a reactor cooled and moderated by heavy water with a graphite reflector. The possible interest of the decommissioning of the Ispra I reactor as a source of information of general interest for reactor decommissioning has been evaluated starting from 1978, in view of the preparation of a programme proposal to be included in the JRC plan 1980-1983.

On January 1979 an expert meeting has been convened at Ispra on this matter, with the participation of 10 experts of 7 Member Countries.

Due to the decision of the General Directorate of the JRC not to include this activity in the research programme proposal 1980-1983 the activity in this field will be concluded soon. A few additional tests will be performed to determine the residual activities which are important for the evaluation of the safety aspects of the reactor which is now in the stage of mothballing.

COLLABORATION WITH EXTERNAL ORGANIZATIONS

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

CONCLUSIONS AND PLANNED ACTIVITIES

Chemical Decontamination

The studies performed have already generated information on the mechanism and efficiency of the APAC process which can be useful for an improvement of decontamination operations. Tests have been performed on contaminated pieces obtained from power stations and artificial surfaces prepared with different procedures. SEM have been applied for the analysis of the surfaces at the different decontamination stages.

In the next reporting period the investigation will be enlarged by including tests on the surfaces oxidized at 300°C for 1800 h in hydrogenized and oxygenized water

(oxidations carried out in the small test loop) and by applying SIMS and AES for the study of the process mechanism. SEM microprobe analysis will be carried out on the contaminated pieces obtained from power stations.

In order to reduce the amount of active waste it would be desirable to use chemicals which can be burned or recycled. In the APAC process the organic acid of the first and third stages does not give problems in this respect. Tests will be performed for the substitution of the permanganate, in the second stage of the process, with chemicals which can be recycled.

Study of the Physico-Chemical Structure of the Oxide Layers

The second part of the study concerning oxide layers formed in oxygenized water will be concluded.

The results of the investigations on the physico-chemical structure of the oxide layers will be presented at the meeting on Water Chemistry organized by VGB at Essen on October 1979.

Decontamination by Redox Cycling

Cycling experiments will be carried out, in the small test loop, controlling the release of oxide and the morphological and chemical changes of the surfaces.

Decommissioning of Ispra I Reactor

In order to get information on the safety aspects of the installation the following operations are planned :

- Dose mapping in the central region of the reactor
- Analysis of radioactive nuclide content in materials sampled in the reactor shielding
- Evaluation of the state of conservation of the structure and services of the installation.



Conclusions

3. CONCLUSIONS

In the first 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 the study for the application of the methodology developed at Ispra, to quantify the probabilistic value of the geological barrier in the Boom (Belgium) clay formation, has been completed. The overall failure probability appears to be low enough to assure a large margin of safety.

The application to the same geological formation of the deterministic section of our model, used to calculate pathways and dose rates to man, will be started in the next few months in collaboration with CEN/SCK, Mol.

During the reporting period also the work for the comparison, on the basis of the long-term risk, of the once-through strategy and of the uranium-plutonium recycle strategy in LWRs, has been completed. The evaluation of the long-term risk has been carried out by means of our deterministic model, assuming an hypothetical waste repository.

The results of this study indicate that the long-term risk is higher in the case of the once-through strategy; however, also in the worst cases considered, the dose rates to individuals appear to be very small.

The above reported studies show that the methodology developed at Ispra can be applied both for the evaluation of specific sites and for the comparison of alternative fuel cycles. All the results obtained in our studies indicate that the geological disposal can assure a large margin of safety.

In the field of the long-term stability of the conditioned wastes, further contributions have been produced, by the JRC studies, to the knowledge of radiation damage and leaching phenomena in vitrified wastes.

In particular it is worth mentioning that experiments conducted on borosilicate glasses in different conditions, show that the dose-rate has not a considerable influence on the damage. This is a proof in favour of the validity of the accelerated tests on vitrified high activity waste, which are carried out in several laboratories to simulate the long-term radiation damage.

In the field of the interaction of the actinides with geological media, interesting results have been obtained in experiments on the migration of plutonium and americium in columns of the soil which overlays typical clay formations. The analysis of the results indicates that the migration of the actinides in geological media is a complex phenomenon of difficult interpretation. The physico-chemical nature of the migrating species seems to play a major role. This confirms the validity of our orientation to put more effort on the study of the physico-chemical forms of the actinides in the conditions of the geological disposal. During the reporting period considerable progresses have been made in this area by the setting-up of techniques for the characterization of the actinide physico-chemical forms.

In the field of actinides monitoring, the collaborative programme for an Integral Experiment on the plutonium waste measurement system of the Dounreay Nuclear Power Development Establishment (DNPDE) is being developed following the planning. In this area it is worth mentioning the organization of an International Meeting on Monitoring of Pu-Contaminated Waste, to be held at Ispra on 25-28 September 1979.

This meeting will be 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.

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 on 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.

In the field of the chemical separation of actinides the experiments on the OXAL process have been essentially completed. The experiments on the solvent extraction processes (TBP and HDEHP) are directed to provide the information needed to complete the flow-sheet optimization. The proposed flow-sheet will be verified in the laboratories of CEA (CEN, Fontenay-aux-Roses, France) by continuous counter-current experiments, based on the use of mixer-settler batteries.

This activity will be carried out in the framework of a collaboration contract between CEA and JRC.

Engineering evaluations of the three proposed flow-sheets are in progress.

The considerable laboratory activities, complemented by engineering studies, are expected to produce for the end of 1979 a valid assessment on the chemical separation of actinides from HAW.

Also in the field of the assessment studies on the nuclear transmutation of actinides, the various lines of activities are organized in such a way to produce for the end of 1979 an overall analysis of the most important aspects of this advanced strategy.

During the reporting period considerable progresses have been made in the field of reactor physics calculations for LWRs, fuel element design and risk analysis.

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

Decontamination of Reactor Components

The studies in progress in this area using advanced analytical techniques, are producing high quality information on the physico-chemical structure of the oxide layers. These data, complemented by investigations on the mechanism and efficiency of the chemical decontamination, should give an important contribution to the improvement of the decontamination procedures. The planning of future activities is shown in Table III.

For further information please contact the Programme Manager.

PLANNED ACTIVITIES AND IMPORTANT MILESTONES (JULY - DECEMBER 1979)

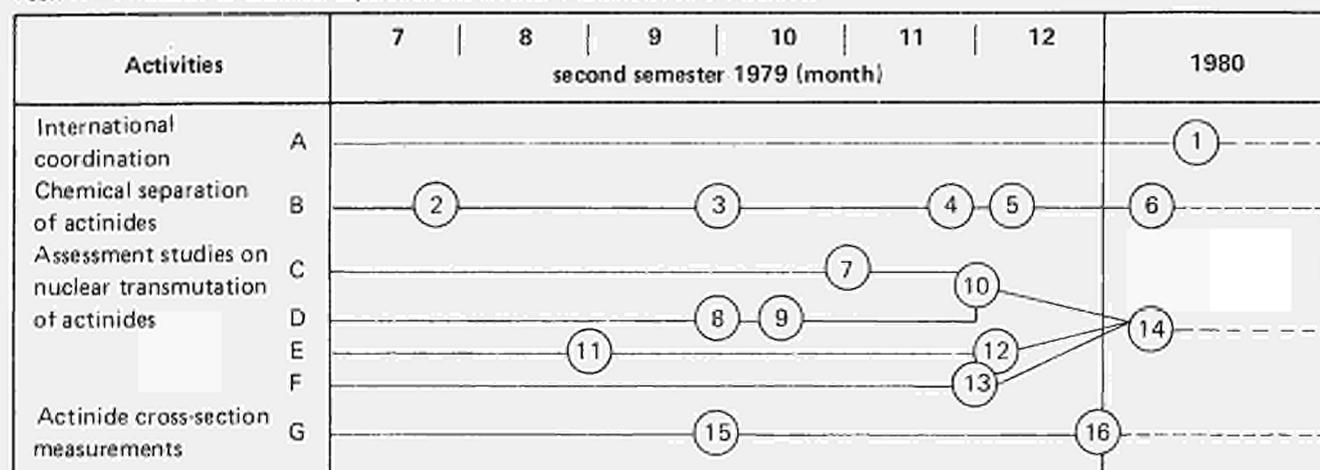
Table 1 - PROJECT 1: Evaluation of the Long-Term Hazard of Radioactive Waste Disposal

Activities	second semester 1979 (month)						1980
	7	8	9	10	11	12	
Waste hazard analysis	A						
	B						1
Long-term stability of conditioned waste	C		2			3	
	D	4					
	E						5
	F				6	7	8
Actinide monitoring	G						
	H	9		10		11	12 13

A	Modelling of failure of geological disposal						
B	Actinide distribution in 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 vetrified waste			2			Completion of the experiments with water in conditions of geological disposal
				3			Completion of the analysis of the leached surfaces
D	Leaching tests on bituminized waste			4			Start-up of experiments on bitumen charged with dispersed oxide powders
E	Experiments of radiation damage in glasses			5			Completion of the glass sample irradiation in the HFR reactor and start-up of the post-irradiation tests
F	Interaction with abiotic environment			6			Completion of column experiments on glauconite sand with Pu and Am
				7			Completion of column experiment on glauconite sand with Np
				8			Completion of a set of experiments on Pu chemical forms using Pu-237
G	Interaction with biosphere						Collection of data and promotion of activities in a strict link with the indirect programme Radiation Protection
H	Plutonium waste monitoring			9			Upgraded signal processor for passive neutron assay operative
				10			Organization at Ispra of the Intern. Meeting on Monitoring of Pu Contaminated Waste
				11			Completion of the experimental verification of the interpretational model for passive gamma assay
				12			Revision of the guide
				13			Completion of the integral experiment in a reprocessing plant

PLANNED ACTIVITIES AND IMPORTANT MILESTONES (JULY - DECEMBER 1979)

Table II - PROJECT 2: Chemical Separation and Nuclear Transmutation of Actinides



A International coordination in the framework of the OECD Nuclear Energy Agency

B Chemical separation of actinides by solvent extraction and OXAL process

C Reactor physics calculations

D Fuel element design

E Cost and risk analysis

F Study of the implications of actinide recycling on the fuel cycle

G Measurement of cross sections of Am-243

1 Organization of the 2nd Technical Meeting on the Nuclear Transmutation of Actinides, JRC-Ispra, April 1980

2 Completion of fully active experiments on HDEHP batch extraction

3 Completion of tracer and fully active experiments on HDEHP batch countercurrent extraction and of HAW denitration tests

4 Completion of tracer experiments on TBP batch-countercurrent extraction

5 Completion of engineering evaluation

6 Report on the feasibility of actinide separation

7 Completion of reactor physics calculations

8 Proposal of by-product actinide fuel element for LWR's

9 Assessment on pellet-cladding interaction

10 Elaboration of an overall cycle strategy

11 Parametric studies of fuel cycle costs

12 Definitive results on cost and risk of actinide recycling

13 Assessment on alternative fabrication routes

14 Report on the feasibility of actinide transmutation

15 Completion of the (n, γ) measurements

16 Completion of the (n, f) measurements

PLANNED ACTIVITIES AND IMPORTANT MILESTONES (JULY - DECEMBER 1979)

Table III – PROJECT 3: Decontamination of Reactor Components

Activities	second semester 1979 (month)						1980	
	7	8	9	10	11	12		
Decontamination of reactor components	A	①					②	
	B						③	
	C	④						
	D						⑤	

A	Systematic study on chemical decontamination	1	Completion of the study on the APAC system
B	Study on the physico-chemical structure of oxide layers	2	Completion of tests on total decontamination
C	Partial decontamination using thermal-redox cycling	3	Decision point
D	Evaluation of the decommissioning of the Ispra 1 reactor	4	Decision point
		5	Completion of the measurements of the residual activity

4. JRC PUBLICATIONS

Published or Presented

- 1.G. BIGNOLI, G. BERTOZZI: «Modeling of Artificial Radioactivity Migration in Environment. : a Survey» - EUR 6179 EN, 1978.
- 2.H. SCHNEIDER, A. CARETTA: «Functional Relation of Risk to Input Data Uncertainties» - EUR 6196 EN, 1979.
- 3.A. SALTPELLI, A. AVOGADRO, G. BERTOZZI: «An Assessment of Plutonium Chemical Forms in Groundwater and their Migration through Porous Media» - Paper presented at the Workshop on the Migration of Long-Lived Radionuclides in the Geosphere, CEC-OECD, Brussels, January 1979.
- 4.A. AVOGADRO, C.N. MURRAY, A. DE PLANO: «Laboratory Experimental Development for the Study of Transuranic Migration in Porous Media» - Paper presented at the Workshop on the Migration of Long-Lived Radionuclides in the Geosphere, CEC-OECD, Brussels, January 1979.
- 5.F. LANZA, E. PARNISARI: «Evaluation of Long-Term Leaching of Borosilicate Glass in Pure Water» - Paper presented at Intern. Symp. on Ceramics in Nuclear Waste Management, Cincinnati (USA), April 30-May 2, 1979.
- 6.M. ANTONINI, F. LANZA, A. MANARA: «Simulation of Radiation Damage in Glasses» - Paper presented at Intern. Symp. on Ceramics in Nuclear Waste Management, Cincinnati (USA), April 30-May 2, 1979.
- 7.L. CECILLE, H. DWORSCHAK, F. GIRARDI, B.A. HUNT, F. MANNONE, F. MOUSTY: «Separation of Actinides from Purex Type HAW Raffinates: Development of Experimental Studies at JRC Ispra Establishment» - Paper presented at the Actinide Separation Symp. - Pacific Chemical Conf., Honolulu, April 1979.

Submitted for Publication or Presentation

- 1.F. GIRARDI, A. AVOGADRO, G. BERTOZZI, M. D'ALESSANDRO, F. LANZA, N.C. MURRAY: «A Risk Analysis Methodology for Deep Underground Radioactive Waste Repositories and Related Experimental Research» - To be presented at the Colloque Intern. sur l'Evacuation des Déchets Radioactifs dans le Sol, IAEA/NEA, Otaniemi (Finland), July 1979; IAEA-SM-243/161
- 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» - To be published as EUR Report
- 3.L. ANSELMI, K. CARUSO, W. HAGE, E. SCHMIDT: «On the Hazard Accumulation of Actinide Waste in a Pu-Fuelled LMFBR Power Economy with and without By-Product Actinide Recycling» - To be published as EUR Report.
- 4.O. GAUTSCH, F. LANZA, F. VAN RUTTEN, P. WEISGERBER: «Decontamination of Water Cooled Reactors Primary Circuits and Components: A Bibliographic Review.» - To be published as EUR Report.
- 5.G. BIRKHOFF: «Monitoring of Plutonium Contaminated Solid Waste Streams - A Guide for Design and Analysis of Monitoring Systems. Chapter V: Application of Active Neutron Assay» - To be published as EUR Report.
- 6.F. GIRARDI, M. BRESESTI, S. ORLOWSKI, P. VENET: «Radioactive Waste Disposal in Geological Formations. Research Activities and Programmes of the Commission of European Communities» - Paper submitted for presentation at the International Symposium «Scientific Basis for Nuclear Waste Management», Boston, Massachusetts, November 26-29, 1979.
- 7.A. AVOGADRO, C.N.MURRAY, A. DE PLANO: «Transport through Deep Aquifers of Transuranic Nuclides Leached from Vitrified High Level Waste» - ibidem
- 8.M. ANTONINI, P. CAMAGNI, F. LANZA, A. MANARA: «Atomic Displacements and Radiation Damage in Glasses Incorporating High Level Waste» - ibidem

LIST OF AUTHORS

Programme Manager: **M. Bresesti**

Assistant to the Programme Manager: **J. Ley**

Scientific Coordinator: **F. Girardi**

CONTRIBUTORS

Project 1:

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

Project 2:

L. Anselmi, L. Cecille, H. Dworschak, M. Forte, F. Girardi,
S. Guardini, W. Hage, B. Hunt, F. Mannone, F. Mousty,
E. Schmidt, G.R.B. Smith, E. Zamorani

L. Koch (JRC - Karlsruhe), F. Kaepfeler, K. Wisshak
(Institute for Applied Nuclear Physics, KFK, Karlsruhe)

Project 3:

F. Lanza, P. Weisgerber.

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