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Management of Nuclear Materials and Radioactive Waste

PROGRAMME PROGRESS REPORT

January-June 1977

ABSTRACT

This document is the progress report of the programme Management of Nuclear Materials and Radioactive Waste of the Joint Research Centre for the period January - June 1977.

The programme consists of three projects. The main achievements during the reporting period were the following:

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

Models and methodologies set up during the previous year for waste risk analysis have been reviewed and summarized in two reports. The studies on the long-term stability of vetrified waste have been continued: the activity has also been extended to bituminized waste.

A new activity has been started on the interaction of actinides with environment.

In the field of actinides monitoring a guide is in preparation and experiments have been performed in Pu-fabrication plants.

Project 2: Chemical Separation and Nuclear Transmutation of Actinides

Promising results have been obtained in the separation of the actinides from waste by oxalate precipitation.

Considerable progress has been made concerning nuclear code calculations of actinides formation and transmutation.

Project 3: Decontamination of Reactor Components

This is a new project started in 1977. The activity was mainly directed to identify the areas of major interest by means of an external enquiry and extensive literature survey.



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1. INTRODUCTION

The exploitation of nuclear energy at an industrial level leads to the production of large quantities of radioactive waste. The management and disposal of the radioactive waste is one of the most important issues of public concern, particularly for its long term implications on future generations, due to the presence in waste of α -emitting nuclides of rather long half-life (actinides).

The Joint Research Centre initiated its activity in the field of Radioactive Waste Management in 1973. In the first four-year programme (1973-1976) the efforts were essentially made in three directions:

- 1) to study the feasibility of a management strategy based on the chemical separation of actinides from waste and their recycling into nuclear reactors, in order to minimize the amount sent to waste;
- 2) to study a methodology for the evaluation of long-term hazard of waste disposal in geological formation, in order to see whether there is valid motivation to carry out a sophisticated nuclear-fuel cycle as it is necessary for the management strategy mentioned above;
- 3) to study whether a pyrochemical process treatment of the fuel before reprocessing could simplify the waste disposal problems, by removing in a concentrated form certain gaseous radioisotopes (Kr-85, I-129 and H-3, essentially) and minimizing formation of solid residues during fuel dissolution.

In addition to the JRC programme on Waste Disposal, the Commission of the European Communities started in 1975 an Indirect Action programme on Waste Management. In this programme, 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 among them and with national activities was assured by appointing one Advisory Committee for Programme Management (ACPM) for the two actions.

In 1977 the second multiannual programme of the JRC on Waste Management was initiated and is presently underway. The programme, when compared with the previous one, shows a diminished effort in the technological aspects of waste management and a greater effort on safety studies. Thus, while action 3 of the previous programme (development of pyrochemical processes) was abandoned, action 1 (long-term hazard evaluation) was considerably reinforced.

A particular aspect of the safe operation of nuclear reactors, the decontamination of reactor components, is also being studied within the framework of this programme.

The accumulation of long-lived fission products in reactor circuits is in fact posing to reactor operators severe problems of maintenance and repair, due to dose exposure of personnel in charge of those operations. Substantial quantities of waste are also generated. The action proposed should provide the necessary knowledge in order to optimize, on a case-by-case basis, the decontamination procedure.

The staff allocated to the JRC programme for 1977 consists of 64 research men. The programme is carried out at the Ispra Establishment with a participation of the Karlsruhe Establishment.

The programme Radioactive Waste Management has been organized into three projects, subdivided into several studies:

Project 1: Evaluation of Long Term Hazard of Radioactive Waste Disposal

The objective is to evaluate the long-term hazard of permanent disposal of radioactive waste in appropriate geological formations, by studying the barriers, natural and man-made, which may be interposed between waste and the biosphere for long-term safety assurance.

Project 2: Chemical Separation and Nuclear Transmutation of Actinides

The objective is to obtain, by theoretical assessment studies and experimental actions, a better appreciation of advantages and disadvantages of separating the actinides from radioactive waste and transmuting them into shorter-lived fission products by recycle in nuclear reactors.

Project 3: Decontamination of Reactor Components

The mechanisms of water circuit contamination, the nature of the contaminated layers and the action of decontaminating agents will be studied.



Projects

2. PROJECTS

2.1 EVALUATION OF LONG TERM HAZARD OF RADIOACTIVE WASTE DISPOSAL

This project includes the development and application of waste hazard analytical models and various experimental studies. The studies on the stability of the conditioned waste and on the interaction of actinides with the environment will provide input data for the waste hazard models. The study on actinides monitoring is important both for safety reasons and to optimize the waste management.

2.1.1 Waste Hazard Analysis

OBJECTIVES

The aim of this study is to continue the assessment works started during the past programme [1, 2] in order to get a comprehensive view of radioactive waste hazard, with particular emphasis on quantitative value of the barrier system placed between waste and mankind. This aim will be pursued through the development and application of assessment methodologies, such as Fault Tree Analysis for the probabilistic assessment of the value of geological containment for well defined geological disposal options, and critical pathway analysis for the determination of environmental levels of radioactive pollution and corresponding dose rates to man.

In such a way the weak points of a proposed waste management system can be identified and evaluated.

It is expected that a large number of data of interest for this study will be produced in the framework of the Indirect Action of the Commission.

For the first six months of 1977 the planned activities were the following:

- to describe in two reports the models and methodologies set up for geological disposal risk assessment
- to start developing an aquatic ecosystem model
- to organize, in collaboration with the Nuclear Energy Agency (NEA) of OECD an international workshop on risk analysis and geologic modelling
- to plan future developments of the study.

RESULTS

Models and Methodologies for Geological Disposal Risk Assessment

A hazard evaluation "Model 1" was developed in which the following four barriers which can be interposed between waste and man have been quantified:

- 1) Geological segregation
- 2) Stability of Waste Conditioning
- 3) Retention in Subsoil

4) Mobility of Radioisotopes in the Biosphere.

The model is based on the probabilistic evaluation of barrier 1 (by the "Fault Tree Analysis" methodology - fig. 1 and 2) and on the deterministic evaluation of the other barriers (by a Pathway Analysis).

The FTA showed that the exhumation by future generations could be a likely pathway for the waste to return to the environment during the first thousands of years; on longer term the leaching by ground water becomes the most likely mechanism.

Following this release mode, the resulting pollution of a terrestrial ecosystem (fig. 3) has been analyzed, and the following information has been collected:

- The calculated dose rates to man (shown in fig. 4 and 5) are low; however experimental verification of the assumed mechanisms and transfer coefficients are necessary.
- Management of alpha-bearing waste other than high-activity waste, is the most important parameter for hazard assessment.
- The plutonium inhalation pathway can play a very important role, within the first thousands of years, while Ra-226 becomes the risk governing isotope over very long time periods.

Aquatic Ecosystem Model

Initial considerations of a simple steady-state marine ecosystem have been made using a compartment modelling concept. This approach permits the identification of major components of the system.

Transfer equations can then be applied, often on the basis of classical physical laws, for the movement of radioactivity from one environmental compartment to the next. These equations can be considered to approximately define the outcome of all the processes taking place within each component of the system.

Using the above method, a critical group-critical pathway hazard assessment can be attempted such that dose rates to man from actinides released into a marine environment can be evaluated.

Workshop on Risk Analysis and Geologic Modelling

The workshop on "Risk Analysis and Geologic Modelling in relation to the Disposal of Radioactive Wastes into Geologic Formations" was held at Ispra, on 23-27 May 1977, organized by the OECD Nuclear Energy Agency in cooperation with the CEC Joint Research Centre, Ispra.

43 Experts from 15 countries attended the meeting. The methodologies for the risk analysis under development in the different countries and organizations have been presented and discussed.

In the specialized sessions all the aspects of the problem have been analyzed and a particular attention has been devoted to the problems concerning the quantitative evaluation of the different barriers, natural and arti-

ficial, which are interposed between radioactive waste and biosphere. Problems requiring priority in the research have been identified. The necessity of a continuous confrontation of the methods of evaluation used in the different countries has been recognized.

The participants also agreed on the interest to establish a central bank of geologic data.

CONCLUSIONS

Our present methodology for risk assessment appears to be adequate; however, the model must be considerably enlarged, particularly for what concerns environmental distribution patterns. The objectives listed above have been fulfilled, and guidelines for future developments have appeared clearly.

PLANNED ACTIVITIES AND COLLABORATION WITH EXTERNAL ORGANIZATIONS

A "Model 2" is presently being developed, which comprises a more sophisticated terrestrial environment and a compartmental marine model. Improvements in the data handling techniques are also being studied.

In collaboration with the CEN-Mol (Belgium), site-specific models, involving both Fault Tree Analysis and Pathway Analysis, will be developed, concerning the disposal safety of radioactive waste into some particular Belgian clay formations.

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- [2] BERTOZZI, G., D'ALESSANDRO, M., GIRARDI, F., VANOSSI, F.,
"Safety Assessment of Radioactive Waste Disposal into Geological Formations - A Preliminary Application of Fault Tree Analysis to Salt Deposits". Workshop on Risk Analysis and Geologic Modelling in Relation to the Disposal of Radioactive Wastes into Geologic Formations, Ispra, May 23-27, 1977. To be published as EUR-Report.

2.1.2 Long Term Stability of Conditioned Waste

OBJECTIVES

The aim of this study is to obtain information on long term behaviour of conditioned high and medium-level waste, which is necessary for waste hazard evaluation.

The planned activities for the first half of 1977 were the following:

- Post-irradiation examination of glasses irradiated in the Petten reactor

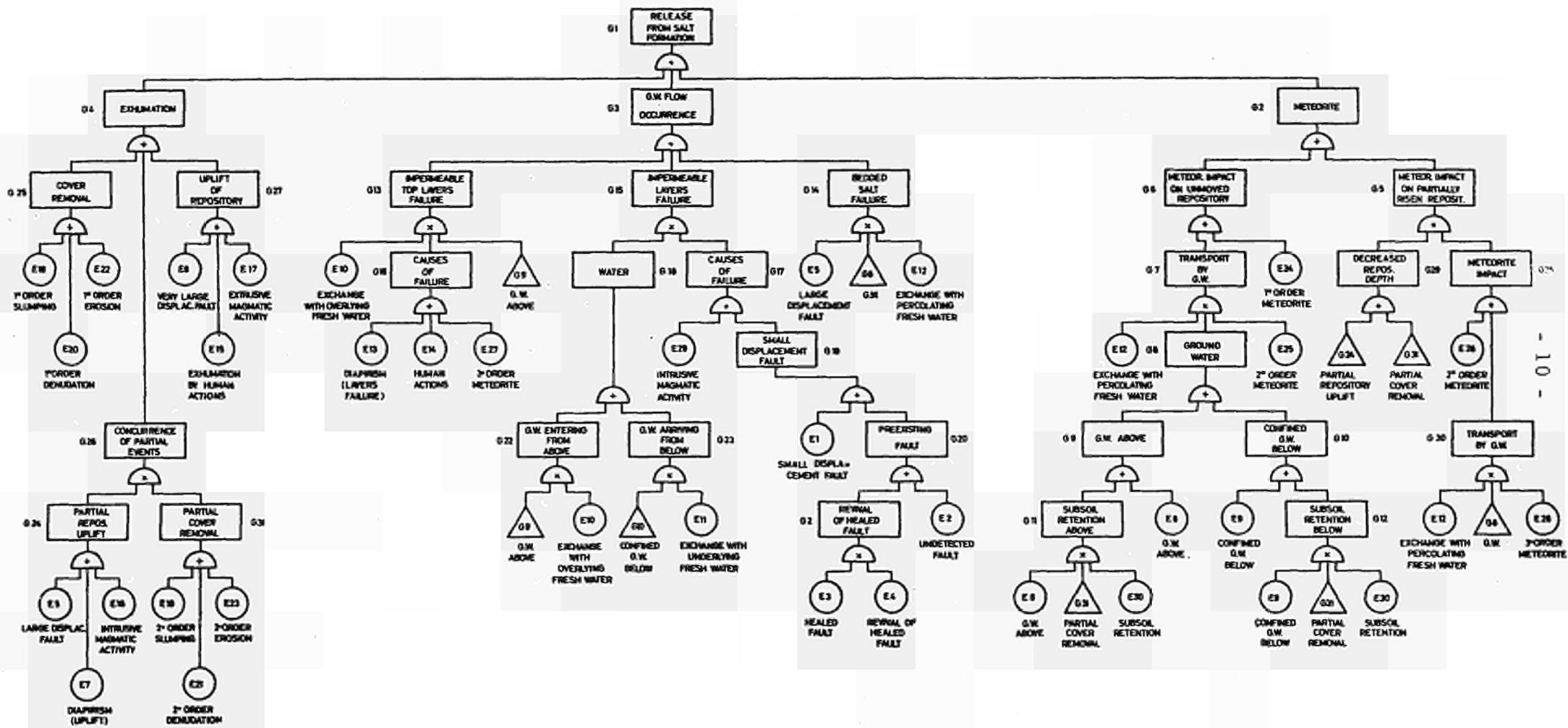


Fig. 1 - Fault Tree for Disposal in Bedded Salt

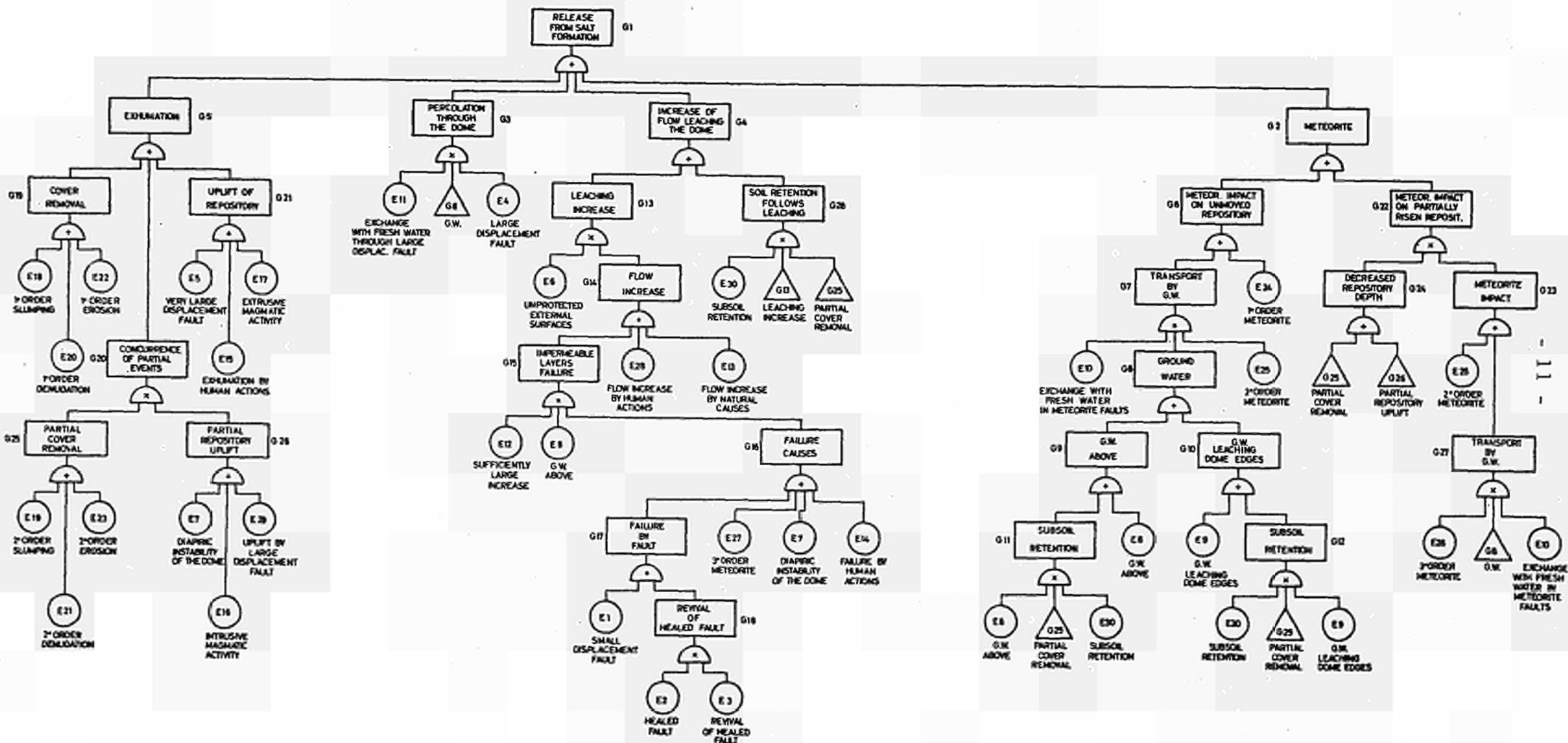


Fig. 2 - Fault Tree for Disposal in Salt Dome

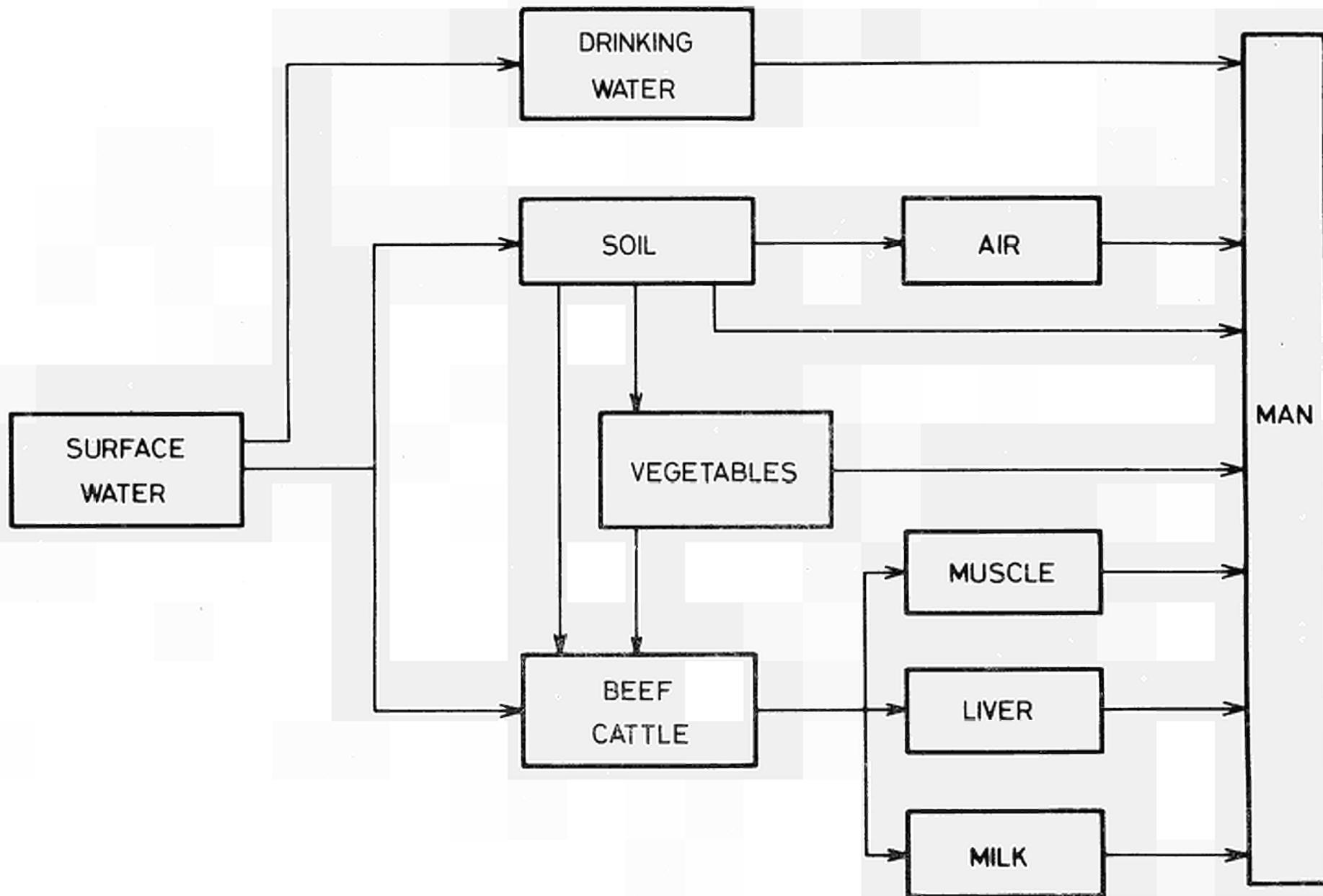


Fig. 3 : TERRESTRIAL SYSTEM OF THE PRESENT MODEL

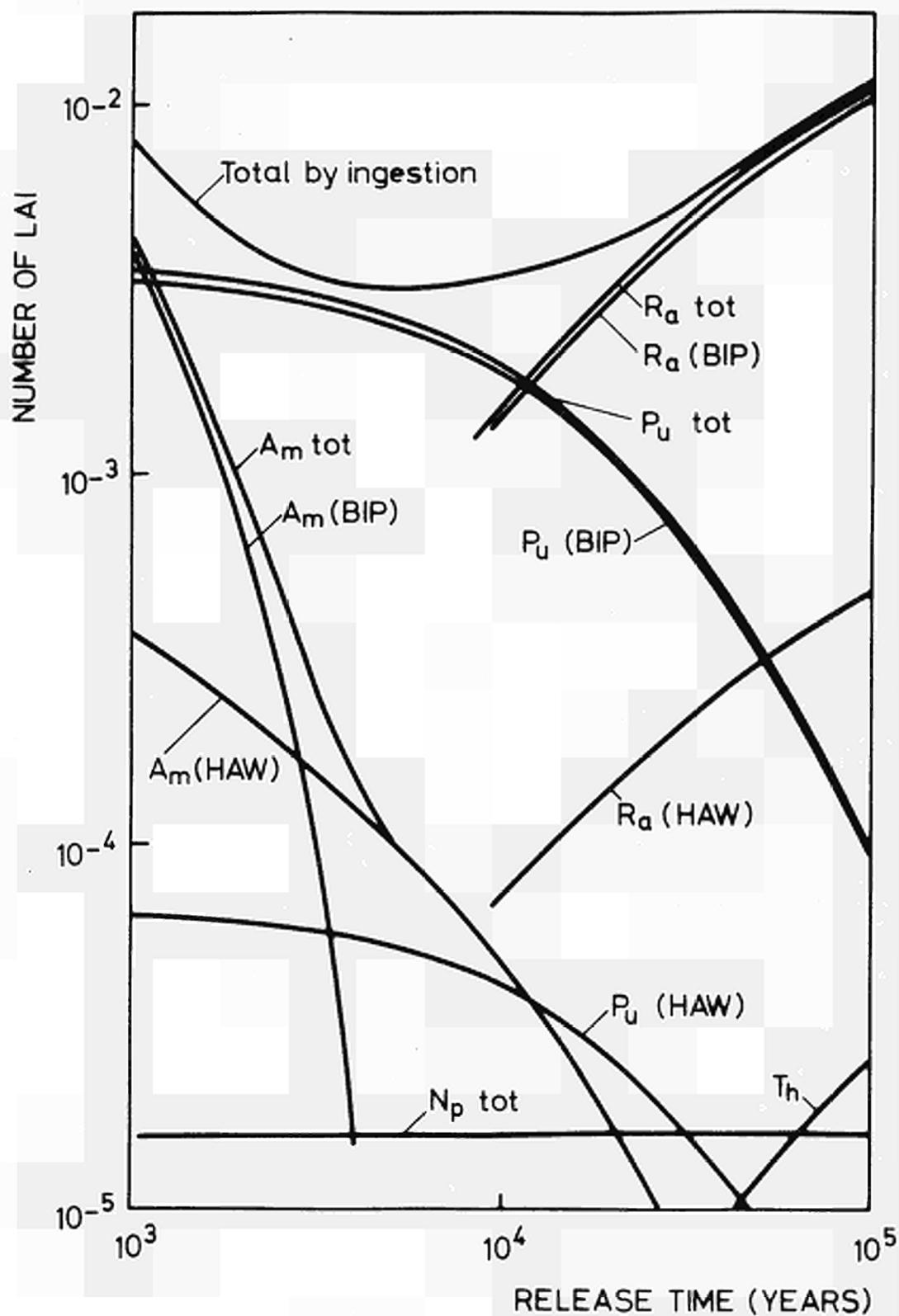


Fig. 4 : NUMBER OF LAI RECEIVED BY INGESTION, AS A FUNCTION OF RELEASE TIME

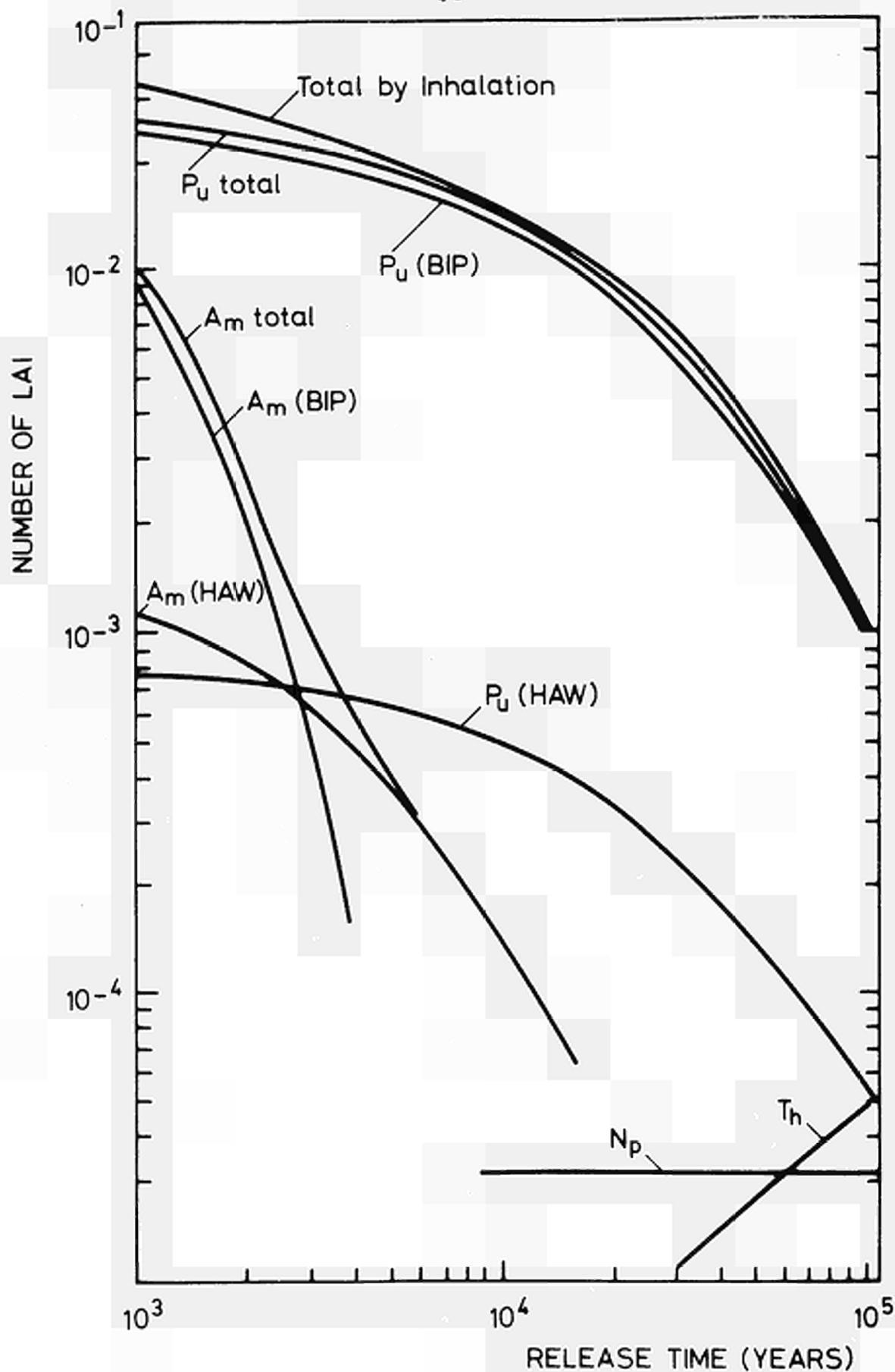


Fig. 5 : NUMBER OF LAI RECEIVED BY INHALATION, AS A FUNCTION OF RELEASE TIME

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

RESULTS

Post-Irradiation Examination of Irradiated Glasses

The capsules irradiated in a fast flux at the Petten reactor under a cadmium screen have been dismantled.

The irradiation was intended to simulate, by the damage caused by fission fragments, the damage produced by the alpha particles [1]. A maximum of fission density of $4 \cdot 10^{17}$ fissions/cm³, which corresponds to $2 \cdot 10^{22}$ displaced atoms/cm³, was calculated for the irradiated samples.

Post-irradiation tests have been initiated. At the dismantling it was noted that the glass pellets were intact.

As the calculation of the number of fissions in an irradiation under cadmium shield is not very reliable, measurements of fission products in the irradiated samples are being carried out in order to verify the calculated fission densities.

Density measurements and leaching tests have been started. The post-irradiation measurements are expected to be completed before the end of November.

Glass Leaching Tests

The results obtained in the first part of the investigation of long term leaching of glasses have been analyzed and a report has been prepared [2]. The surface layer tends to be enriched in the less leachable components at first but, after a certain period of time corresponding to a weight loss of about 2 mg/cm², it evolves towards a composition similar to the composition of the leached glass. Thus the assumption of a coherent dissolution seems to be confirmed.

A new series of leaching tests utilizing samples which are not reintroduced in the corroding medium, in order to avoid thermal cycling and possible damage to the surface gel, has been started.

The first results show that the weight loss is quite homogeneous at the beginning. After a certain time the scattering increases sharply. It is possible that after the film has reached a critical thickness, the release of product is obtained not only by a solution process but also by a colloid release.

Stability Tests on Bitumen

Inglobation in bitumen has been considered for alpha-contaminated waste.

The main advantage is a good long-term stability in abiotic environments. The radiation damage has been mainly studied for its short-term implications (hazard from the formation of H₂ and CH₄ gases, swelling). For long-term implications the formation of open porosity is the main point of interest resulting in large increase in the leaching rate.

Some preliminary tests on bitumen have been initiated in order to verify the dependence of the leaching from the solubility of the bitumen-incorporated salt and to clarify the mechanism of the long-term leaching.

A detailed planning of the research will be done after discussions with national experts which should indicate the possible lines of management of low and medium-level wastes.

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

The necessity of accelerated tests to simulate the long-term damage puts the problem of studying the influence of annealing and self-annealing in the radiation processes.

An investigation of this type requires:

- 1) the evaluation of the various possibilities of irradiation at controlled dose and dose-rate,
- 2) the investigation of the measuring techniques most suitable to monitor the damage,
- 3) the choice of the samples,
- 4) the set-up of experiments.

1) Irradiation Problem

A review of the literature and a discussion with experts in Harwell lead us to choose external irradiation due to the following reasons:

- a) External irradiation acting on very small amounts of material drastically reduces the irradiation time necessary to produce the same number of relative displaced atoms, with respect to an internal source, and it eliminates remote handling of irradiated active samples.
- b) By avoiding the inclusion of active substances in the glass, it allows us to see only the effect of the radiation and not other effects due to the introduction of foreign substances.
- c) This technique allows us to separate the effect of alpha-particles from that of heavy ions.
- d) The external source allows better control of the various radiation parameters.
- e) By means of energy variations of the bombarding particles, the ratio

between bulk and surface damage may be controlled; in particular an increase of surface damage may be interesting in connection with properties such as the leaching parameter and etching rate which are related to the modifications of surface properties.

- f) Irradiation with a source internal to the sample is already done at Ispra; therefore it is convenient, in order to have complementary results, to use an external source.

2) Technique for Post Irradiation Tests

We thought to divide the activity into two phases. In a first phase we have the necessity of techniques that allow us to compare our results with those obtained recently in Harwell as well as in Ispra with internal sources. Therefore density and stored energy measurements are considered.

In a second time we plan to use techniques which allow better understanding of the structural damage at a sub-macroscopic stage (light scattering, ESR, electron microscopy, optical absorption, X-ray diffraction).

3) Sample Choice

Samples investigated will be amorphous silica and borosilicates. The first has a simple structure where it is easier to analyze the damage processes. The second ones are necessary for a comparison of the effects obtained in a simple SiO_2 matrix with those obtained in real glasses (containing also alkalis).

4) Set-up of Experiments

Preliminary irradiation tests will be carried out at Ispra with alpha-particles (2.5 MeV maximum energy) on samples of tetrasil amorphous silica (99.99 purity) of 5 mm of diameter and 1 mm thickness, optically smooth.

Some attention has to be devoted, when preparing irradiation tests, to experimental problems arising from poor conduction of heat and current through the samples. Moreover, a special effort has to be made in order to calibrate the dose rate.

An estimation of particle range and of the DPA (atomic displacements per atom) for an integrated alpha-particle flux of $10^{18}/\text{cm}^2$ has been worked out (Table 1).

We envisage also to perform irradiation with alpha-particles of higher energy and with heavy ions utilizing the facilities available at Harwell.

Stored energy measurements will be carried out on the irradiated samples, by means of a DSC Dupont calorimeter (0.01 mC/inch sensitivity).

Table 1

Energy (MeV)	Range (μm)	DPA
3	14	1.1
2	8	1.6
1	5	2.7
.5	3	3.9
.2	2	5.4
.1	1.7	6.5

Density measurements will be carried out with the Preston method (sensitive to the 4th decimal figure) at the "Stazione Sperimentale del Vetro" Murano (Italy). Preliminary measurements of density will be performed in our laboratories with a torsion balance (sensitive to the 2nd decimal figure).

In parallel to the density measurements, determinations of the refraction index will be carried out (with sensitivity to the 4th decimal figure). Refractometry has the advantage of a high sensitivity for surface damage and correlates directly with surface density.

CONCLUSIONS

As far as radiation effects on glasses are concerned, the first post-irradiation tests do not show any significant effect.

The aim of the leaching tests is to verify that the model assumed in the hazard analysis of a homogeneous dissolution of the glasses is correct and to give the values of the rate of dissolution.

The results obtained up to now seem to confirm this assumption and have given a first set of dissolution rates in pure water.

For what concerns the check of the validity of damage simulation, the initial phase of evaluation has been completed.

In general the planning was respected with the exception of the post-irradiation measurements which have been delayed due to licencing procedures of the LMA laboratory.

PLANNED ACTIVITIES

For the second half of 1977 the following activities are planned:

Radiation effects on glasses:

Completion of the post-irradiation measurements on the glasses irradiated in the Petten reactor; verification of some specific points with additional irradiations.

Leaching tests on glasses:

Completion of the tests in pure water; examination of the influence of some special water compositions related to specific geological repositories.

Stability tests on bitumen:

Evaluation of the leaching of conditioned salts as a function of their solubility in water; preparation of a detailed planning of research in order to define some lines of management of low and medium-level wastes.

Check of the validity of damage simulation on glasses:

Irradiation on silica samples with alphas and heavy ions; measurements of stored energy, density and refraction index on the irradiated samples.

COLLABORATION WITH EXTERNAL ORGANIZATIONS

For the check of the validity of damage simulation on glasses a collaboration has been established with the "Stazione Sperimentale del Vetro", Murano (Italy).

Contacts have also been made with Harwell (UK).

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"Simulation Possibilities of Radiation Effects in Glasses Used for Conditioning High-Activity Waste". EUR 5560e, 1976
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2.1.3 Interaction of Actinides with Environment

OBJECTIVES

The objective of this new study connected with the long-term hazard evaluation, is the quantitative description of the behaviour of the actinides in terrestrial and aquatic environments following loss from a geological repository containing alpha-bearing wastes.

The planned activities for the first 6 months of 1977 were the following:

- A systematic literature survey for a better definition of the experimental work on physico-chemical aspects of the interaction
- Start-up of the experimental effort on the study of the interaction of plutonium with different soil components and of its partitioning in an aquatic system.

A collaboration with the Radiobiology programme of the Commission is

foreseen.

RESULTS

- a) A systematic literature search has been carried out in order to build up an initial information base for the planned study of the migration and distribution of actinides in aquatic and terrestrial systems.

Taking into consideration this survey, the following experimental study areas have been identified:

- Problems related to the detection of different chemical species of actinides in water and sediments, specially in relation to their behaviour in very dilute solutions.
- Characterization of physico-chemical state of actinides in leached solutions from borosilicate glass.
- The development of laboratory systems which adequately simulate natural conditions.

b) Analytical Techniques

Plutonium has been chosen initially for the study on actinide interaction with environment. Two isotopes of high specific activity are considered particularly suitable for the present experimental study. These are Pu-238, an alpha-emitter, for use in long term experiments such as migration in soil columns, and Pu-237, a gamma-emitter of short half-life, which allows very easy determination of rapid phenomena such as initial particle formation in water, pH and redox effects, distribution variation at sediment-water interface etc.

c) Glass Production and Leaching Solution Characterization

Simultaneously with the ordering of equipment (high temperature furnace, grinding and sieving apparatus) required for the production in glove-box of Pu-238 contaminated glass, a laboratory production of a standard borosilicate glass has been carried out. The composition of this glass is as follows: 60% SiO₂, 18.8% B₂O₃, 6.2% Al₂O₃ and 15% Na₂O. In this glass was incorporated 20 wt% simulated waste oxide containing approximately 7% UO₂. The glass was ground and the resulting material (1-2 and 0.5-1 mm size range) leached at different temperatures with water. This developed technique will be used to make the glass containing 0.5 mg Pu-238/g glass, from which a leaching solution for its subsequent chemical characterization will be produced.

d) Laboratory Simulation

- The equipments for the carrying out of sub-soil column experiments, for the determination of actinides interaction with different soil components, have been ordered; the start-up of this activity is foreseen

for the second half of 1977.

- An aquarium has been prepared in order to study the Pu-237 partitioning in an aquatic system consisting of lake water and sediments, including biological components, characteristic of a lake near Ispra (Lago di Monate). The methodology evolved in this work will form the basis for future development on more realistic environmental systems

CONCLUSIONS AND PLANNED ACTIVITIES

The objectives proposed in this initial phase of the programme have been fulfilled. For the second half of 1977 the planned activities are:

- a) Production of a borosilicate glass spiked with Pu-238 and start-up of the study of the chemical characterization of the leaching solution.
- b) Start-up of the experimental activity on plutonium migration through sub-soil columns.
- c) Continuation of the study with Pu-237 in an aquatic system in order to determine its distribution and size range of formed particles as a function of pH and redox potential.
The partitioning between water and sediments, including biological components will also be checked.

2.1.4 Actinides Monitoring

OBJECTIVES

The study aims at the development of a methodology for plutonium waste monitoring in compliance with pertinent regulations.

This goal is going to be pursued in the framework of an Advisory Laboratory*. The organization of this laboratory and its activity since 1975 were rigorously discussed during an Expert Meeting on this subject held on November 23-24, 1976 at Ispra [1].

No major criticisms were forwarded by the participants; hence the programme activity has been continued according to the original plannings.

RESULTS

During the reporting period the major activity was concerned with:

- 1) Development of an interpretational model for spontaneous fission neutron measurements
- 2) Measurements of effective escape probabilities of fission neutrons from selected reference materials

*The original name "Central Laboratory" was objected by participants of the Expert Meeting [1] and replaced by the word "Advisory".

- 3) Measurements of plutonium contents in contaminated solid and liquid wastes from a Pu handling laboratory by passive neutron assay.

The results of these studies will be synthesized in Chapter IV of our Guide /2/.

Interpretational Model for Spontaneous Fission Neutron Measurements

The interpretational model of spontaneous fission neutron measurements describes the average history of neutrons from their birth within the sample, their interaction with the sample materials up to their leakage out of it and their probability of generating detection signals.

From that we deduce the corrections relative to a point reference sample located at a given reference position (r_0, z_0) within the 4π -neutron detector cavity. This model is outlined in the flow diagram of fig. 1.

The total correction of the observed neutron detection signals is expressed by a set of six correction terms. Those terms are grouped together into 3 correction factors:

The first factor (point 2 of fig. 1) takes into account the dependence of the detection probability on the sample geometry for a given neutron detection assembly. This detector is assumed as being Cd-shielded in such a manner that sub Cd cut-off neutrons do not penetrate the sample cavity-detector boundary. The detection probability may depend on the neutron energy, being zero for sub Cd cut-off neutron energies.

The second factor (point 3 of fig. 1) corrects for removal of neutrons with the sample by elastic and/or inelastic scattering below Cd cut-off, radiative capture, for variation of neutron energy spectrum by elastic and inelastic scattering and for $(n, 2n)$ reactions with sample materials. This factor we call the effective mean neutron escape probability. It is the effective self-shielding factor of the sample. The mean square effective escape probability is needed for correction of the autocorrelated signal, as this is proportional to the square of the detection probability.

The third factor corrects for contributions of induced fission reactions to the autocorrelated signal. The four terms of this factor describe the contributions of four types of induced fission sources, namely:

- fast fission by spontaneous fission neutrons,
- thermal fission by spontaneous fission neutrons,
- fast fission by non-spontaneous fission neutrons,
- thermal fission by non-spontaneous fission neutrons.

Neutron Escape Probability Measurements

The influence of neutron interactions other than induced fission with sample materials is expressed by the effective neutron escape probability. In the first order approximation, this quantity is defined in analogy to the well-known neutron shielding removal cross section concept. This means that removal of neutrons from being detectable is described

by an exponential function (first collision model). The adjustment of this kind of approximation to a more realistic value of the effective escape probability is accomplished by standards.

Those standards or reference materials must be similar to the sample materials in terms of the above mentioned neutron removal effects. The effective escape probability for given neutron source distributions can be determined experimentally by measuring the distribution of the neutron detection probability over the volume of the reference material by means of a reference point source. We have determined in this way the escape probability spectra for a uniform source density in cylinders from some reference materials such as polyethylene, wood and iron. The spectra of probabilities for polyethylene are shown in fig. 2 and fig. 3. These spectra relate to two different neutron detector types as shown in fig. 4. The effective mean escape probability, for a uniform source density distribution is obtained by averaging over the volume of the reference material.

It has been proved that the escape probability for the first collision model is described rather well by an "equivalent sphere". This effective sphere radius is obtained from substituting a cylinder with the height H and the radius R by a sphere having the same volume to surface ratio. In evaluating the effective escape probability of an unknown sample, we have to know about its material composition, geometry and weight. From this we calculate the removal cross section Σ_R . Then we compute the equivalent sphere radius of the sample R_S .

From the relationship

$$\Sigma_R \cdot R_S = \Sigma_R^{\text{ref}} \cdot R_S^{\text{ref}}$$

we compute the effective radius of the sample if it is replaced by the reference material. With this effective radius we obtain the corresponding effective mean escape probability. For non-uniform source density distribution we have to return to the effective Pe distribution and to evaluate its mean value for the given source distribution.

Pu-Waste Measurements by Passive Neutron Assay

These measurements are part of a so-called "integral experiment" on Pu waste monitoring in collaboration with CNEN Casaccia, Italy. Such experiments are aimed at the verification of the waste monitoring concept as outlined in our Guide [2]. About 300 waste drums were assayed. The interpretation method, following the model outlined in fig. 1, is to be applied. For this purpose a computer program is going to be written. Preliminary results have been elaborated.

CONCLUSIONS

The preliminary results from interpretation of spontaneous fission neutron measurements on plutonium contaminated wastes are encouraging but must still be reviewed and compared with destructive analyses prior

to concluding upon the validity of the developed interpretational model. The complexity of non-destructive assay by radiometric assay in general and of passive neutron assay in particular calls however for a major effort which has been underestimated in the beginning of this programme activity. The acceleration of the progress in this field calls for additional research manpower. Such support can also be achieved by means of collaboration with research institutes. In view of this, contacts with a university have been established, hoping that some work of our interest can be carried out by students. For this purpose the subject matter has been presented in a seminar organized and invited by this university. The discussion on possible contributions to our work is in course.

PLANNED ACTIVITIES

In the following 6 months period are planned:

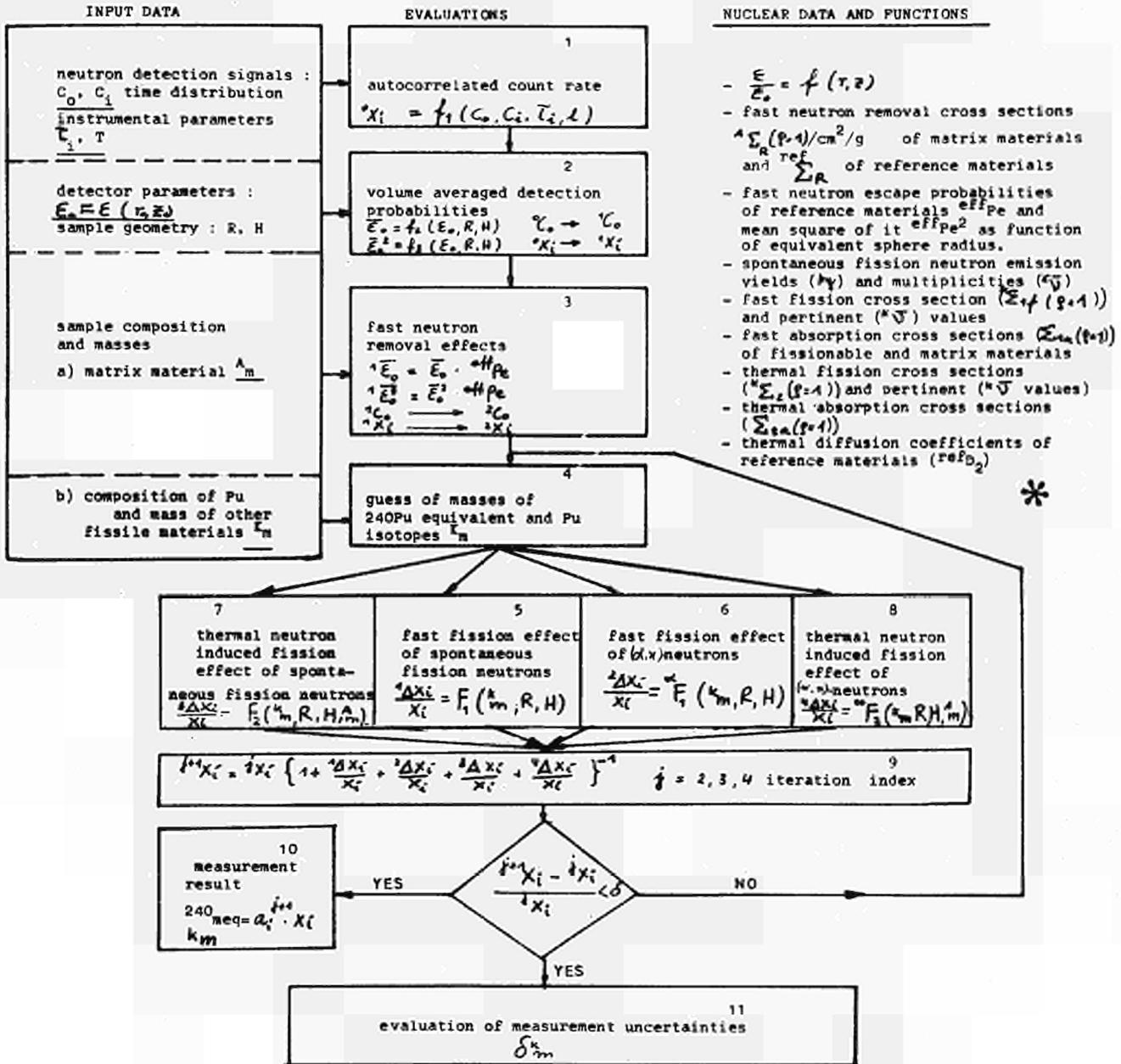
- 1) Experimental verification of the validity of the reference monitor for passive gamma assay as defined conceptually in chapter III of our Guide
- 2) The same as 1), but for passive neutron assay
- 3) Final analysis of the waste measurements on Pu waste from CNEN Casaccia
- 4) Writing chapter IV of our Guide: "Application of passive neutron assay"
- 5) Testing of an on-line liquid alpha-monitor under operational conditions of a fuel reprocessing facility.

COLLABORATION WITH EXTERNAL ORGANIZATIONS

- 1) Contract with EUREX Saluggia, Italy, concerning the testing of an on-line liquid alpha-monitor (stipulated in 1977)
- 2) Contract with CNEN Casaccia on integral experiments on monitoring Pu-contaminated waste streams, to be stipulated in fall 1977.

REFERENCES

- [1] Minutes of the Expert Meeting on Monitoring of Pu-Contaminated Solid Waste Streams, held at Ispra on November 23-24, 1976
- [2] Monitoring of Plutonium-Contaminated Solid Waste Streams - A Guide for Design and Analysis of Monitoring Systems:
BIRKHOFF, G. and NOTEA, A., Chapter I: "Planning of Monitoring Systems". EUR 5635e, 1976
BIRKHOFF, G., BONDAR, L., NOTEA, A., SEGAL, Y., Chapter II: "Principles and Theory of Radiometric Assay". EUR 5636e, 1976
BIRKHOFF, G., NOTEA, A., Chapter III: "Application of Passive Gamma Assay". EUR 5637e, 1976
BIRKHOFF, G., BONDAR, L., Chapter IV: "Application of Passive Neutron Assay",
Chapter V: "Application of Active Neutron Assay". In preparation.



* induced fission effects by spontaneous fission and (α, n) -neutrons

$$\frac{1,2 \Delta x}{X} = F_{1,2}(\rho_f, \bar{\nu})$$

$$\frac{3,4 \Delta x}{X} = \alpha_F F_{1,2}(\rho_f, \bar{\nu})$$

Fig. 1 : Interpretation of spontaneous fission neutron measurements

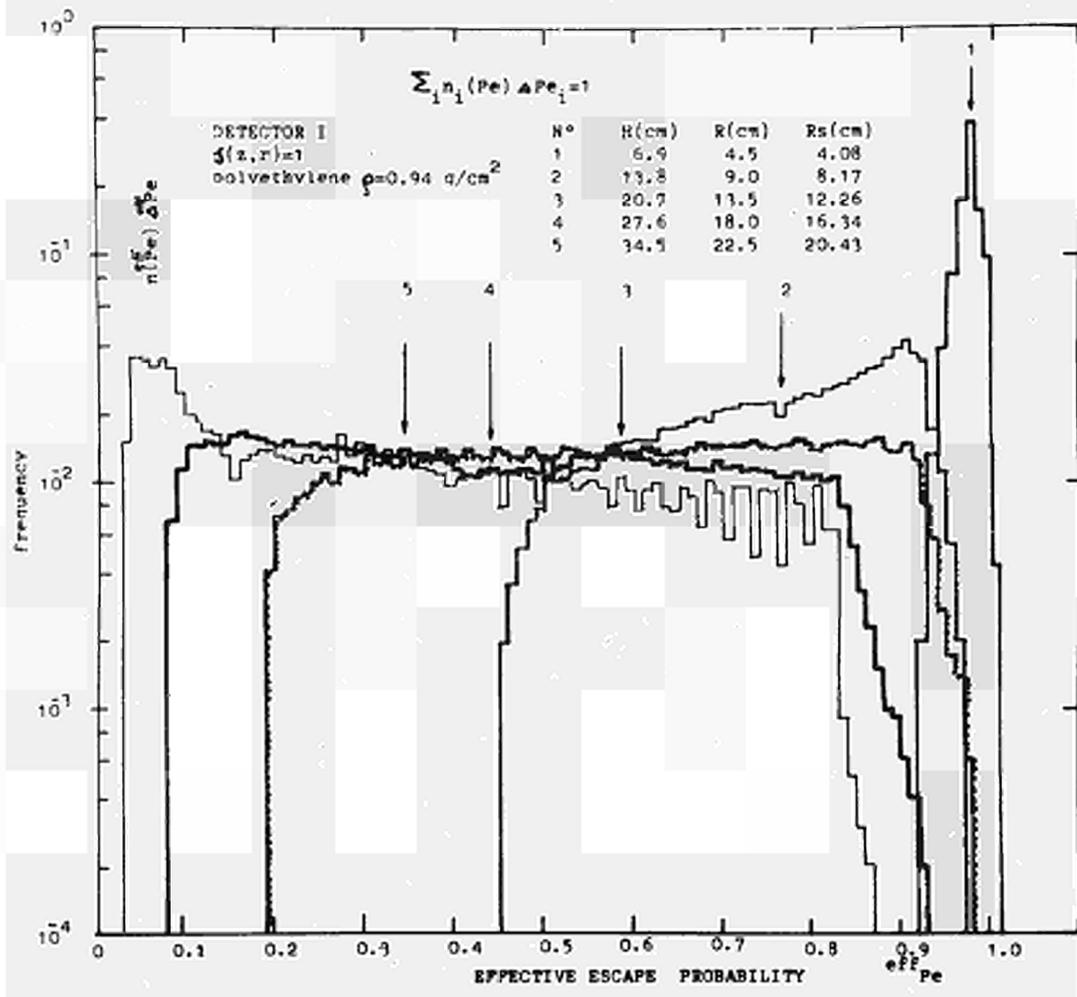


Fig. 2 : Spectra of effective escape probabilities of fission neutrons from polyethylene cylinders of various dimensions, measured with detector type I (see Fig. 4)

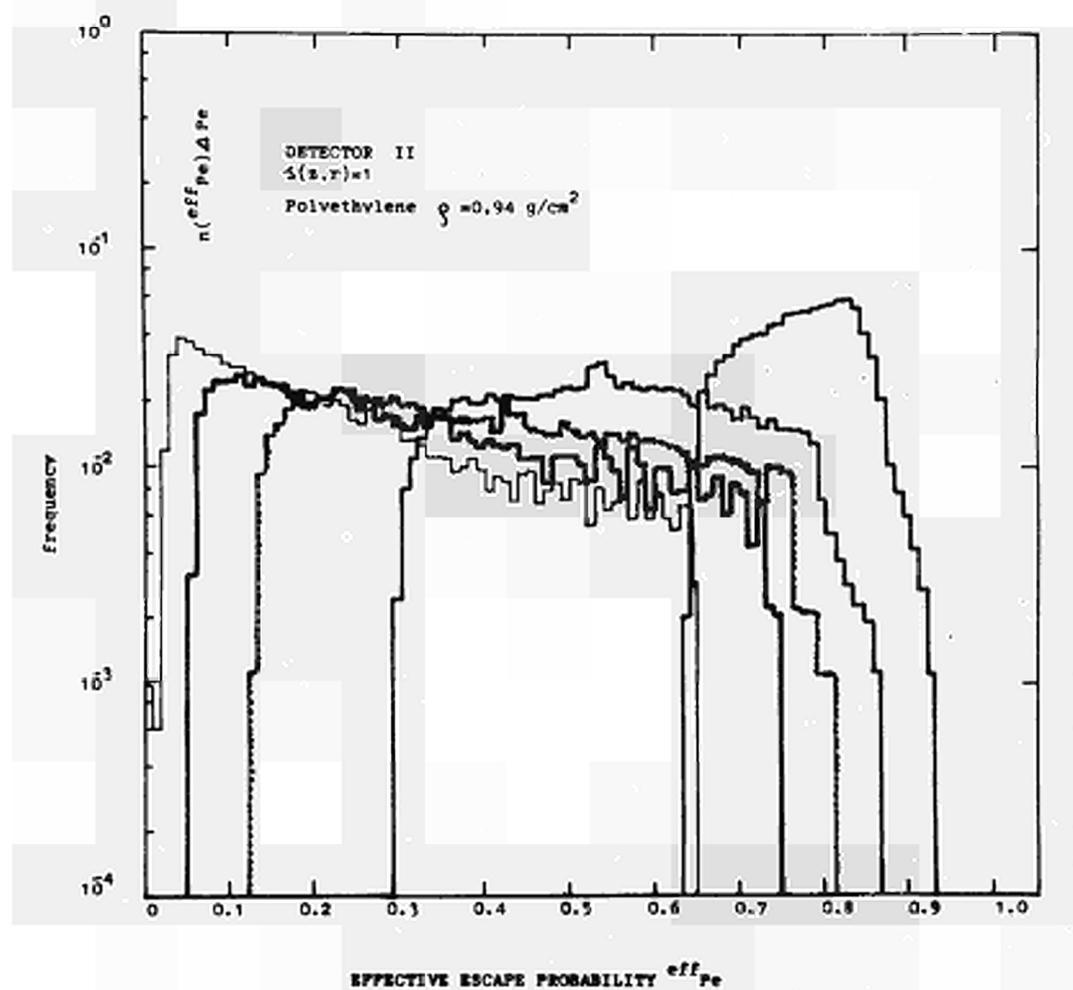


Fig. 3: Spectra of effective escape probabilities of fission neutrons from polyethylene cylinders of various dimensions, measured with detector type II (see Fig. 4)

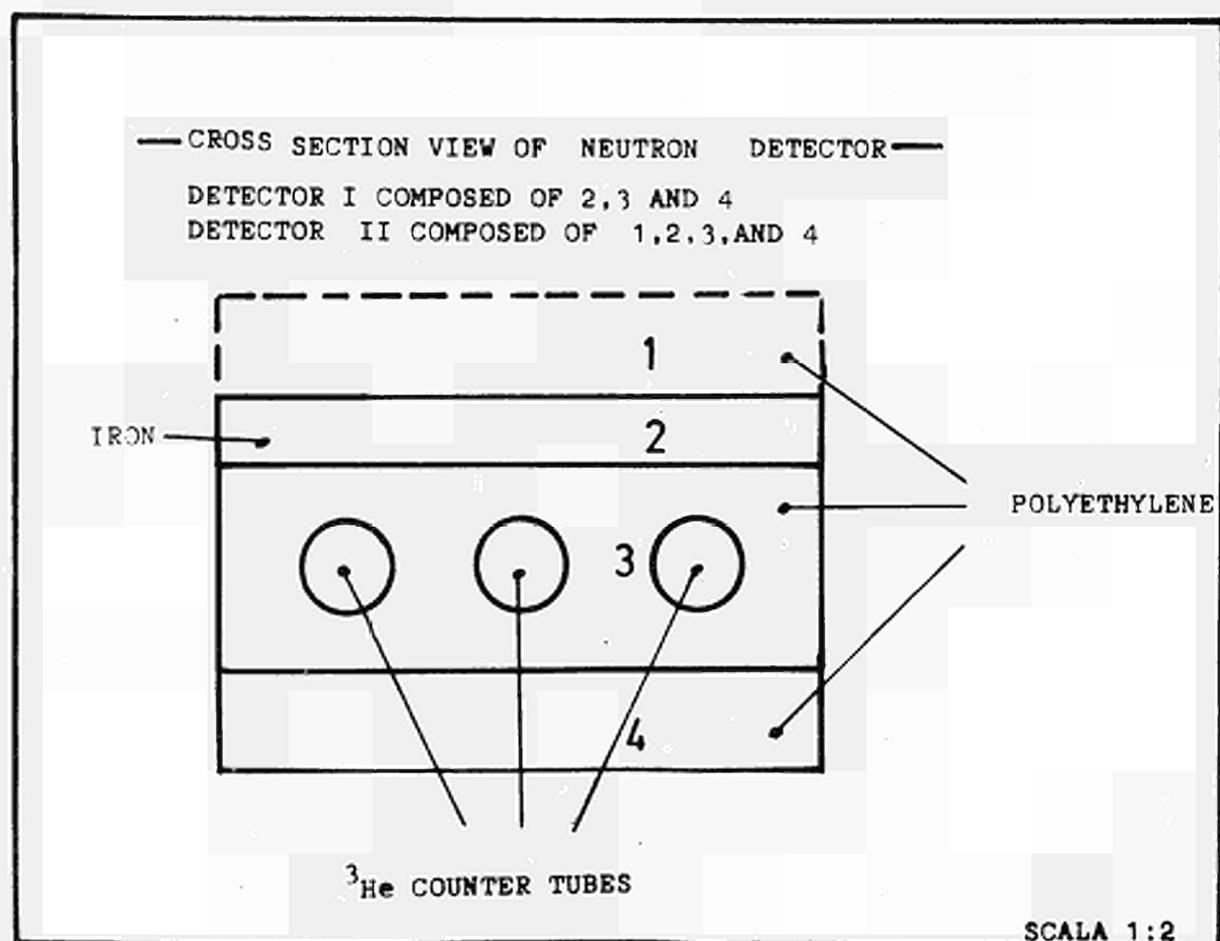
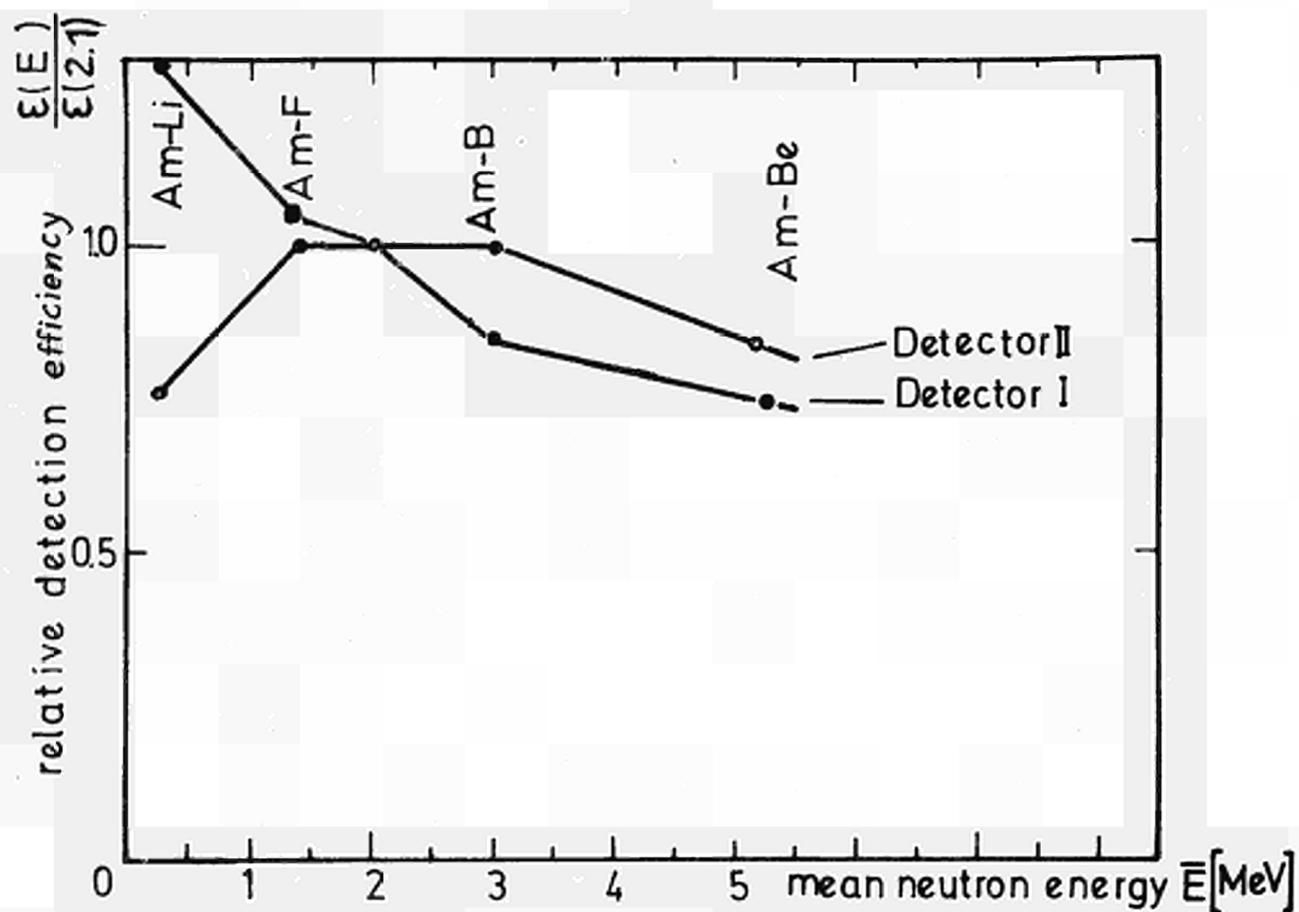


Fig. 4: Cross section views of detector type I and II and their relative detection probabilities as function of neutron energy

2.2 CHEMICAL SEPARATION AND NUCLEAR TRANSMUTATION OF ACTINIDES

This project comprises the following activities:

- Research on chemical separation of actinides from waste
- Assessment studies on the transmutation of actinides in nuclear reactors and on the technological implications of the actinide recycling
- Measurements to improve the knowledge of actinide neutron cross sections.

The Indirect Action of the Commission for Radioactive Waste Management includes an activity in the field of the chemical separation and nuclear transmutation of actinides.

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.

First Technical Meeting on the Nuclear Transmutation of Actinides

held at the JRC-Ispra, 16-18 March, 1977 (sponsored by the Nuclear Energy Agency of the OECD).

As a consequence of the leading role assigned to the Commission, a meeting on the Nuclear Transmutation of Actinides in Fission Reactors was organized at Ispra on 16-18 March 1977, in order to establish contacts between workers in this field and to provide a basis for exchange of information.

The meeting was attended by 38 people of 12 countries and international organizations and by 20 staff members of the Commission.

Several papers were presented and discussed in 5 technical sessions.

The first session concerned the presentation of topic papers dealing with the Chemical Separation of Actinides, the Effects of Recycled Actinides in a Nuclear Reactor and the Technological Impacts on the Nuclear Fuel Cycle.

In a second session international and national research programmes were presented and some delegations gave orally the point of view of relevant authorities concerning the nuclear transmutation concept.

The third session consisting of 4 papers was dedicated to the chemical separation of actinides and here mainly to different conceptual methods.

11 papers were presented in the fourth session dealing with the build-up and transmutation of actinides.

The actinide build-up was investigated for LWRs, LMFBRs and for HTGRs. Actinide recycling calculations were presented for LWRs and LMFBRs.

In session 5 sensitivity studies for the influence of nuclear data on the actinide build-up, the long term hazard of waste, the actinide transmutation rate etc. were presented. Sensitivity coefficients for these different aims were given for LWRs and LMFBRs. Experimental techniques and new cross section measurements closed this session.

The conclusions of the meeting were summarized in 3 different workshop sessions:

Session 1 : Chemical Separation of Actinides

Session 2 : Neutron Physics Feasibility Studies for Actinide Recycling

Session 3 : Implications of the Fuel Cycle Burning Actinides.

In a final plenary session the conclusions arrived at in the workshop sessions were discussed.

The meeting has been considered by the participants as most successful and the subjects treated stimulated the audience to fruitful discussions. The workers in the different fields found personal contacts where these did not yet exist and contacts were taken for future closer exchange of information.

It was decided to publish the presented papers, the declarations presented in written form and the results of the workshop sessions approved by the participants in Proceedings, which will appear late 1977.

2.2.1 Chemical Separation of Actinides

OBJECTIVES

The objectives of this study are to obtain by laboratory experiments fundamental data and all other information needed to develop on a laboratory scale, a complete process flow-sheet for actinide removal from high activity waste (HAW). Solvent extraction and oxalate precipitation are the methods presently investigated to remove actinides. The experimental work planned for the reporting period was directed to demonstrate the chemical feasibility of various process steps of some flow-sheets proposed for HAW partitioning and based both on solvent extraction [1, 2] and oxalate precipitation [9].

Simulated HAW solutions and batch extraction techniques [1, 2] have been employed for solvent extraction tests, simulated and real HAW solutions for oxalate precipitation tests.

RESULTS

1) Actinide Separation by Solvent Extraction

The solvent extraction processes now being developed for partitioning purposes involve the use of organic extractants such as an acidic (HDEHP) and a neutral alkyl derivative of phosphoric acid (TBP).

a) Solvent extraction by HDEHP

Two options have been envisaged for the partitioning flow-sheet (fig. 1) based on HDEHP. The first option involves the selective extraction of higher valent actinides before the denitration step, the second one the direct denitration of untreated HAW raffinates.

The results of laboratory experiments carried out according to the various process steps and operating conditions envisaged by these options are briefly reported.

- Selective Extraction of Pu and Np from Acidic HAW

As expected, the results of the laboratory tests show that nearly quantitative extraction of Pu(IV), U(VI), Mo(VI) and Zr(IV) and rapid extraction kinetics can be obtained when a simulated acidic HAW solution is contacted with a mesitylene solution of suitably purified HDEHP*. Iron is also extracted but less rapid extraction kinetics have been measured. Neptunium can be extracted only when it is stabilized at tetra- and hexavalent state (Table 1).

Table 1 - Extraction of Pu, Mo, Zr, U, Np and Fe from Acidic HAW Solution

Org. Phase: 0.25 HDEHP in Mesitylene, Aq. Phase: Simulated HAW solution, 4 M HNO ₃ + 0.1 M NaNO ₃ O/A = 1, Stirring time = 4 min.						
Extracted Element	Pu	Mo	Zr	U	Np	Fe
% Extraction	98	96	~ 100	~100	87	22

The addition of nitrite ions was performed in order to obtain hexavalent neptunium and to prevent the formation of small amounts of interface precipitates, probably due to palladium and rhodium.

Pu(IV), Mo(VI), Zr(IV), U(VI), Np(VI) and Fe(III) can be stripped from loaded solvent by a low-acidity solution (pH=2.5) of ammonium oxalate (Table 2).

To improve to an acceptable level (<4 min.) the back-extraction kinetics, it is necessary to operate at 45°C.

* Experimental tests already performed under such conditions were not representative due to the fact that the HDEHP was not sufficiently purified. These tests were therefore repeated using a 99.9% pure HDEHP supplied by the firm Bayer.

Table 2 - Back-extraction of Pu, Mo, Zr, U, Np and Fe from loaded HDEHP by oxalate ions

Org. Phase: loaded 0.25 M HDEHP in mesitylene						
Aq. Phase: 0.35 M $C_2O_4^{2-}$, pH=2.5						
O/A = 1, Stirring time = 4 min., t = 45°C						
Stripped Element	Pu	Mo	Zr	U	Np	Fe
% Stripping*	~100	96	92	82	95	100

* expressed for each element as percentages of amounts previously extracted.

- Denitration of previously extracted HAW

As a low-acidity feed (pH ~ 2) is required to separate trivalent actinides from HAW solution by HDEHP, the excess of nitric acid present in the HAW solution, is generally eliminated by chemical reaction with concentrated formic acid. The acidic HAW solution is usually added to hot formic acid according to a properly selected HCOOH/HNO₃ molar ratio.

The removal of Pu, Np, Mo and Zr from the acidic HAW solution allows to simplify the control of the denitration level, to prevent the formation of Pu-polymers and to minimize the production of precipitates.

Palladium, rhodium (deposited mainly as metals on the walls of the reaction vessel) along with barium and a minor fraction of ruthenium, have been determined as the main constituents of the precipitates produced during the denitration of previously extracted HAW.

The recovery of noble metals would be possible by a selective nitric acid dissolution. Practical possibilities to achieve this recovery will be experimentally investigated.

- Extraction of trivalent actinides from low-acidity HAW

As the experimental data concerning this step have been already reported [3, 4], only few extraction tests have been carried out in order to confirm these data. 95% of americium has been in fact extracted from a denitrated (pH ~ 2) HAW solution in a single, volume-to-volume, extraction stage. The solvent was a 0.25 M solution of HDEHP and NaDEHP (13%) in mesitylene. The optimization of the operating conditions needed to improve the Am/R, Es separation factors and to facilitate the solvent clean-up, is in progress.

The second option of the proposed HDEHP flow-sheet (fig. 1) does not involve the preliminary extraction of acidic HAW raffinates which are directly denitrated by addition of concentrated formic acid. It is practically the "Reverse Talspeak Process" already proposed by G. Koch

and coworkers [4] for actinide recovery from HAW. A considerable amount of experimental data on trivalent actinide extraction and separation from R. Es are thus available. Consequently, the aim of our laboratory tests was to investigate the behaviour of higher valent actinides (Pu, Np) during the direct denitration process of unconcentrated HAW as well as the co-extraction of Pu and Np still present as extractable ion at low-acidity conditions (pH=2).

- Direct denitration of HAW raffinates: chemical behaviour of Pu and Np

According to option 2 (fig. 1) the denitration is directly carried out on HAW solution (5000 litres/ton U). As mentioned above the denitration is performed by adding the HAW solution to hot concentrated formic acid according to a HCOOH/HNO₃ molar ratio of 2.

A voluminous precipitate, mainly composed by zirconium and molybdenum is formed during the denitration. It has been determined that 20-30% of plutonium, 80-90% of neptunium and 5-10% of americium are scavenged by this precipitate after a reaction time of 7 hours (Table 3).

Table 3 - Adsorption and leaching of actinides from precipitates produced during HAW denitration

Untreated HAW solution		% Adsorbed on Precipitates*		
Isotope	Concentr. (mg/l)	after denitration	after 1st leach. (0.5 M HNO ₃)	after 2nd leach. (hot 4M HNO ₃)
Pu-239	10	20-30	20-30	7-15
Np-237	150	80-90	not measured	0-3
Am-241	30.6	5-10	not measured	< 0.1

* Percent values adsorbed on precipitates were indirectly determined by measuring the concentration of each actinide present in solution after denitration and after various leaching steps.

A nearly complete leaching may be obtained for neptunium and americium, but only 50% of the adsorbed plutonium may be leached out by nitric acid. Leached plutonium and neptunium fractions were in extractable forms; they have in fact been easily extracted by a 0.3 M HDEHP in dodecane.

An explanation of the uncomplete leaching of plutonium could be given by the formation, under low-acidity conditions, of plutonium polymers and by their irreversible adsorption on precipitates (Table 3).

The plutonium fraction remained on the precipitate after the leaching step could be redissolved only using hydrofluoric acid or concentrated and hot hydrochloric acid.

The behaviour of the plutonium during the denitration of a simulated HAW solution is shown in fig. 2, where the variation of the plutonium fraction in solution is reported as a function of the reaction time. The variations of HNO₃ and HCOOH concentrations as well as of pH under the same operating conditions are also indicated.

From fig. 2 it appears that after an initial decreasing of the plutonium fraction in solution up to a minimum value (35%, pH = 1.5), a progressive increasing of this value occurs up to a maximum of about 85% (pH ~ 2).

- Extraction of residual Pu and Np from low-acidity HAW

According to the operating conditions already employed at Karlsruhe laboratories for the "Reverse Talspeck Process" [4], the denitrated HAW solution containing the plutonium fraction remained in solution after denitration, has been extracted by an organic solution of 0.3 M HDEHP + 0.2 M TBP in dodecane.

A rapid and nearly quantitative extraction of plutonium has been obtained using an organic to aqueous volume ratio of about 1.5 (Table 4). The neptunium extraction was nearly quantitative after three extraction stages.

The loaded solvent has been then back-extracted according to the process conditions required for stripping americium and curium (Table 4 Aq. Phase I). Under these conditions, only 12% of plutonium and 8% of neptunium originally extracted have been back-extracted. On the contrary stripping yields of 88% for plutonium and 92% for neptunium have been reached using a 0.35 M oxalate solution at pH 2.5 and 45°C (Table 4).

Table 4 - Back-extraction of Pu and Np from loaded HDEHP + TBP by different stripping solutions

Org. Phase: loaded 0.3 M HDEHP + 0.2 M TBP in dodecane				
Aq. Phase I: 1 M lactic acid + 0.05 M DTPA et pH = 3, O/A = 1.5				
" " II: HNO ₃ 4 M, O/A = 2.5				
" " III: 0.35 M C ₂ O ₄ ²⁻ , at pH = 2.5 and 45°C, O/A = 1				
Stirring time: 4 min.				
Extracted Element	% Extraction	Stripping Solutions		
		I	II	III
Pu	~ 100	12 ⁺	0	88 ⁺
Np	~ 100 [*]	8 ⁺	0	92 ⁺

* Cumulative yield of three successive extractions
+ percent values of stripped element referred to the amounts previously extracted.

b) Solvent Extraction by TBP

The extraction of trivalent actinide by TBP may be successfully carried out at low acidity conditions provided the nitrate salt content of the feed solution is sufficiently high [5]. This latter requirement represents a serious problem, especially when HAW have to be solidified. In fact the presence in the final HAW solution of large amounts of nitrate salts (tons of nitrate salts per ton of spent fuel), added to HAW raffinates to allow the actinide separation, is incompatible with the subsequent HAW vitrification.

A removal of nitrate salts by precipitation appears to be a rather unpractical operation because of the large amount of salts to be handled.

The alternative being investigated in our laboratory is the partitioning of concentrated HAW solutions. In this case lower amounts of nitrate salts have to be added per ton of spent fuel, so that the resulting composition of wastes to be vitrified could remain within the limits required by the glass composition.

- Concentration of HAW raffinates

Evaporation and denitration steps are normally performed on the untreated HAW solution to attain the desired concentration factor and acidity value. If the formation of precipitates as well as the actinide adsorption on them have necessarily to be minimized during this step, the acidity of the solution has not to be lowered below certain limits during the whole concentration process.

Evaporation tests so far performed under not specifically selected conditions, have normally produced considerable amounts of alpha-contaminated precipitates.

In order to set up the best operating conditions required for minimizing during this step the formation of such precipitates, laboratory tests are in progress.

- Denitration of concentrated HAW

This process step and its chemical implications (polymerization of Pu still present in extractable form and production of alpha-bearing precipitates) are strictly connected with the operating conditions of the previous HAW concentration step. Denitration tests have been carried out on a simulated HAW solution which was previously concentrated by a factor 12 and separated from the obtained precipitates. Pu-239 along with Am-241 were previously added to obtain typical actinide waste concentrations, Ce-144, Ru-106 and Fe-59 to trace some inactive constituents of simulated HAW solution.

No visible precipitates have been produced during the denitration tests probably because the greater part of hydrolyzable ions was previously precipitated during the HAW concentration step. The attained acidity concentration was about 0.17 M/l. Laboratory tests are being continued.

The process flow-sheet based on the use of TBP (Fig. 3) as actinide extractant involves two options.

The first one (option 1) is based on the possibility of carrying out a selective extraction of higher valent actinides after concentration and before denitration of the acidic HAW solution.

The second option involves a direct denitration of concentrated and stored HAW solution without any selective extraction of higher actinides under acidic conditions.

The laboratory tests so far performed concern mainly the process steps envisaged by this latter option (fig. 3). The obtained results are summarized here.

- E x t r a c t i o n o f t r i v a l e n t a c t i n i d e s f r o m c o n c e n t r a t e d l o w - a c i d i t y H A W

Aluminium and sodium nitrate salts have been added to the concentrated and denitrated HAW solution in order to obtain a concentration of 0.65 M/l and 1.3 M/l respectively. This solution has been extracted by an organic solution of 30% TBP in dodecane.

The obtained results (Table 5) show that after three successive extraction stages more than 99% of americium and cerium, 93% of plutonium and 68% of ruthenium originally present in solution, were extracted. No further extraction of plutonium was observed during subsequent extraction stages, probably due to the formation of plutonium polymers during the denitration step.

Preliminary tests on the vitrification of such decontaminated HAW solutions have been carried out without any process difficulty; the leaching rate of such glasses was comparable to the value currently measured for glasses having a typical composition [6].

The loaded solvent has been subsequently back-extracted by a 0.05 M DTPA and 1 M glycolic acid solution at a pH-value of 3. With the exception of ruthenium, all the previously extracted elements have been almost quantitatively stripped from loaded solvent (Table 6).

Table 5 - Extraction of Am, Pu, Ce, Ru and Fe by TBP from a 12-fold concentrated HAW after addition of nitrate salts

Org. Phase: 30% TBP in dodecane (pre-equilibrated) Aq. Phase: simulated HAW solution, ~ 400 litres/ton U 1.3 M NaNO ₃ , 0.65 M Al(NO ₃) ₃ .9 H ₂ O, total acidity = 0.17 M/l Stirring time = 20 min, O/A = 3 [Pu] ≈ 0.09 g/l* [Am] ≈ 0.07 g/l ⁺						
Isotope		Am-241	Pu-239	Ce-144	Ru-106	Fe-59
% Extraction yields	1st Extr.	77	88	72	41	0
	2nd Extr.	96	91	94	55	0
	3rd Extr.	99.4	93	99.1	68	0
	4th Extr.	~ 99.94	93	99.6	72	0
	5th Extr.	~ 99.94	93	~ 99.8	74	0
	6th Extr.	~ 99.94	93	~ 100	77	0

* Plutonium was added to the concentrated HAW solution before the denitration step

+ A factor 4 lower than typical concentration values.

Table 6 - Back-extraction of loaded TBP

Org. Phase: loaded 30% TBP from the previous extraction test Aq. Phase: 0.05 M DTPA + 1 M glycolic acid, pH = 3 O/A = 3, Stirring time = 30 min.						
Isotope		Am-241	Pu-239	Ce-144	Ru-106	Fe-59
% Back-extr. yields		96	~99	96	28	-

- Trivalent actinide separation from R. Es

The use of the above mentioned type of solution to strip actinides from TBP is adopted in order to facilitate the subsequent removal of the R. Es. The stripping solution containing actinides and R. Es may be in fact directly used to feed the first step of the Talspeak Process [7] by which a selective removal of R. Es can be attained.

Laboratory tests to demonstrate this possibility, have been carried out. The stripping solution containing actinides (Am-241, Pu-239), R. Es (Ce-144 tracer + other inactive lanthanides) and Ru-106 (see section 2.3) has been in fact extracted by a HDEHP solution 0.5 M in mesitylene.

The obtained results (Table 7) show that 95% of Ce-144 and 12% of Am-241 originally present in the stripping solution were removed by this selective extraction step. After a successive scrubbing of the loaded HDEHP by means of a 0.025 M DTPA solution (pH ~ 4), the extracted fractions of americium and cerium were lowered to 0.5% and 90% respectively, so that a Ce/Am separation factor of about 1800 can be achieved.

Table 7 - Am/Ce partitioning by HDEHP

<u>Extraction</u> Org. Phase: 0.5 M HDEHP in mesitylene Aq. Phase: from the previous TBP stripping step O/A = 3, stirring time = 30 min						
<u>Scrubbing</u> 0.025 M DTPA, pH = 4, O/A = 3						
Isotope		Am-241	Pu-239	Ce-144	Ru-106	Ce/Am sep. factor
% Isotope in Org. Phase:	Extraction step	12	not detected	95	13	136
	Scrubbing step	0.5	not detected	90	13	1.8x10 ³

The organic component DTPA of this solution containing practically all the removed actinides can be easily destroyed by concentrated and hot nitric acid [7].

Laboratory tests concerning this process are being continued.

2) Actinide Separation by Oxalate Precipitation (OXAL Process)

The experimental studies to optimize the proposed [9] oxalate precipitation process (Oxal Process) have been continued. To confirm the results obtained on simulated HAW solutions, some precipitation tests have been carried out on a real HAW solution supplied by the Windscale Plant.

- Experimental tests on Windscale waste solution

The Windscale solution (200 ml), arrived at Ispra laboratories at the end of November 1976, was a HAW raffinate generated by reprocessing Magnox fuel elements with a burn-up of 3500 MWD/ton; 5000 litres of waste solution were discharged per ton of dissolved fuel [10].

The overall decay time at the end of November 1976 was about 5 months. The composition of this solution was not determined; relative activity measurements have in fact been carried out to investigate actinide and F. Ps behaviour during denitration and oxalate precipitation processes.

The results of active test runs carried out in lead cell on 20 ml samples of the Windscale solution are reported in figs. 4 and 5 along with the results obtained under the same process conditions on simulated HAW samples.

Two experimental results obtained from a real HAW solution prepared at the Ispra laboratories from UO_2 fuel with a burn-up of 26,000 MWD/ton are also reported for comparison in fig. 5.

The behaviour of plutonium during the denitration step is shown in fig. 4 where the fractions of plutonium remaining in solution at different denitration times are plotted versus the corresponding pH values.

It can be seen that important fractions of the plutonium originally present in both waste solutions precipitate.

A leaching step by nitric acid was unable to redissolve the precipitated plutonium.

The fraction of americium, curium and plutonium remaining in solution after denitration and oxalate precipitation are reported in fig. 5 as a function of the pH for both types of waste solutions (Windscale and simulated).

As expected, the fractions of precipitated actinides increase according to the pH. Except for plutonium (at pH values lower than 1.5), the experimental results obtained from both waste solutions can be considered in fairly good agreement (fig. 5). The best waste decontamination factors for americium and curium have been, however, obtained on Windscale solution operating at pH values higher than 1.5.

The behaviour of some gamma-emitter fission products (R. Es, Ru, Zr) during the above mentioned process steps has also been investigated. Some experimental results obtained from the Windscale solution are reported in Table 8.

These results show that zirconium, niobium and antimony are almost quantitatively precipitated during the denitration and that only few percent amounts may be redissolved by nitric acid. The ruthenium and cesium fractions scavenged by oxalates appear to be higher than expected and further investigations are needed to confirm and explain such results.

Table 8 - Behaviour of some F. Ps during denitration and oxalate precipitation of Windscale HAW solution

Process steps	% Element remaining in solution							
	Denitration				Oxalate Precipitation			
Run	1	2	3	4	1	2	3	4
Element pH	0.37	1.66	1.41	1.22	0.89	1.22	1.99	1.78
Eu	-	-	100	87	=	=	=	=
Ce	-	-	100	97	2	0.8	0.26	=
Sb	-	-	=	=	53	=	=	=
Ru	-	-	100	99	99.7	72	78	59
Cs	-	-	85	89	100	68	57	52
Zr	-	-	21	=	56	=	=	=
Nb	-	-	=	=	44	=	=	=

- not measured = not detected

- Optimization of Oxal Process

Experimental results, so far obtained by testing the Oxal Process, show that major process difficulties arise because of the plutonium behaviour. It has been experimentally observed in fact that a rather important fraction of plutonium, originally present in HAW solutions, is irreversibly adsorbed (fig. 4) on precipitates (essentially Zr and Mo compounds), produced during the HAW denitration. Special treatments would be thus needed to remove plutonium from them. The addition of a complexing agent to prevent, during the denitration, the formation of alpha-bearing precipitates without interfering with the subsequent oxalate precipitation, would certainly solve the problem.

The most appropriate complexing reagent is obviously the oxalic acid. The latter, unfortunately, can be effectively used only when diluted HAW raffinates are processed: after concentration followed by an interim storage period, the oxalate ions are in fact completely destroyed. In fig. 6 the modified flow-sheet is reported in which the addition of oxalic acid is foreseen before the denitration step. An additional advantage of this flow-sheet is that in this way only one filtration step is needed.

Experimental tests concerning this modified process flow-sheet have been so far carried out, using only simulated HAW solutions. The behaviour of actinides (Pu, Np, Am) and some F.Ps (Zr, Nb, Ru) during the simulated HAW denitration in presence of oxalic acid is illustrated in figs. 7, 8 and 9. Percent fractions of each element still remaining in solution are plotted versus the denitration time. The variation of acidity during the same time is also reported. From figs. 7 and 8 the effectiveness of actinide separation by oxalate precipitation appears to be rather satisfactory.

After a denitration time of 2 hours, americium, plutonium and neptunium fractions still detected in solution were less than 1% of the respective amounts originally present.

The obtained precipitates (R. Es., Am and Pu oxalates) were entirely dissolved by nitric acid so that any special treatment on precipitates (in order to convert actinides in soluble form) can be avoided.

The simultaneous denitration-oxalate precipitation step has to be necessarily tested on real HAW solutions; experimental tests are planned for next months.

3) Hot Cells for Fully Active Laboratory Scale Experiments

The modification of the existing LMA lead cells and the construction of new cells designed for chemical tests at fully active laboratory scale is in progress.

Unfortunately, due to the work performed at LMA laboratories to improve the ventilation and to install two new lead cells, other hot facilities have been forced to remain inoperative from February 1977 on.

At the beginning of September 1977 the lead cell designed to simulate the Purex Process for the production of a radioactive waste solution approaching as near as possible a real HAW raffinate, will start to operate. UO₂ samples from the Gundremmingen BWR, irradiated at 26,000 MWD/ton and 4 years cooled, will be dissolved. The multistage TBP extraction cycle will be replaced by an extraction chromatography (reversed phase partition chromatography) cycle [11]; a chromatographic column, filled with Celite, selected as support material for the stationary phase (TBP extractant), will be fed with the nitric acid fuel solution. Instead of a multistage liquid-liquid extraction, a more rapid uranium and plutonium separation process and more simple apparatus can be thus employed.

The new lead cells designed to carry out continuous counter-current extraction tests using laboratory scale mixer-settlers will be ready to operate at the beginning of 1978.

CONCLUSIONS

The most important planned objectives were fulfilled during this reporting period.

Batch-experiments on simulated HAW solutions have been performed as planned by testing different types of organic extractants under the operating conditions envisaged for the respective process flow-sheets.

Experimental tests on oxalate precipitation from simulated and real HAW (Ispra and Windscale wastes), as well as optimization of the Oxal process flow-sheet have also been carried out as foreseen for this reporting period.

PLANNED ACTIVITIES

During the next six months, it is planned to ultimate the batch-extraction tests on simulated HAW and to initiate those on real HAW.

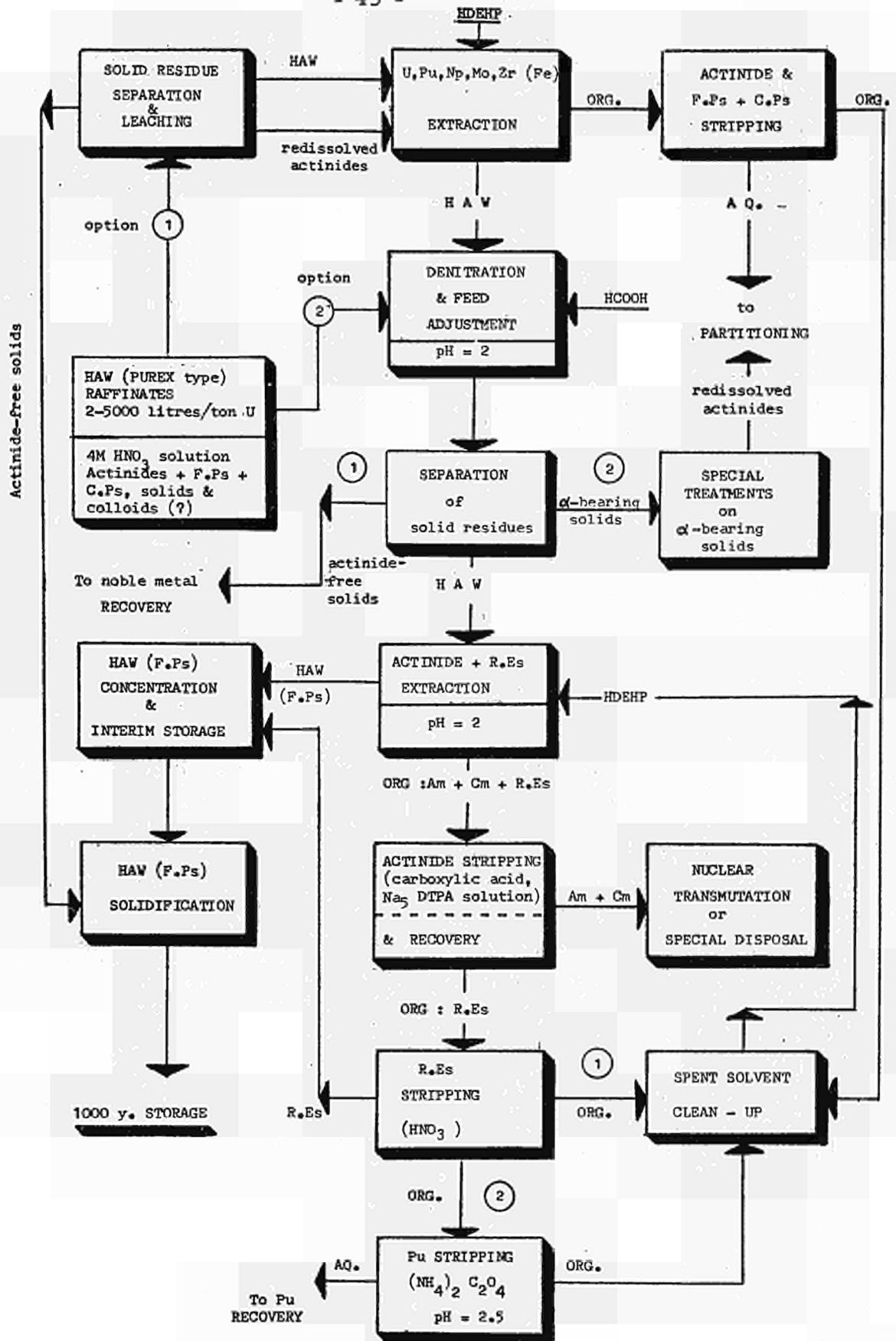
New chemical lead cells will be equipped to this purpose and also for future counter-current extraction tests at fully active laboratory scale planned for the beginning of 1979.

The optimization of the Oxal Process and its verification at fully active laboratory scale, will be continued during the same time period.

The routine production of a synthetic HAW from high burn-up fuels will start around September 1977.

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CCR - EURATOM ISPRA	TENTATIVE FLOW-SHEET FOR HAW PARTITIONING BASED ON ACTINIDE EXTRACTION BY HDEHP	Fig. 1
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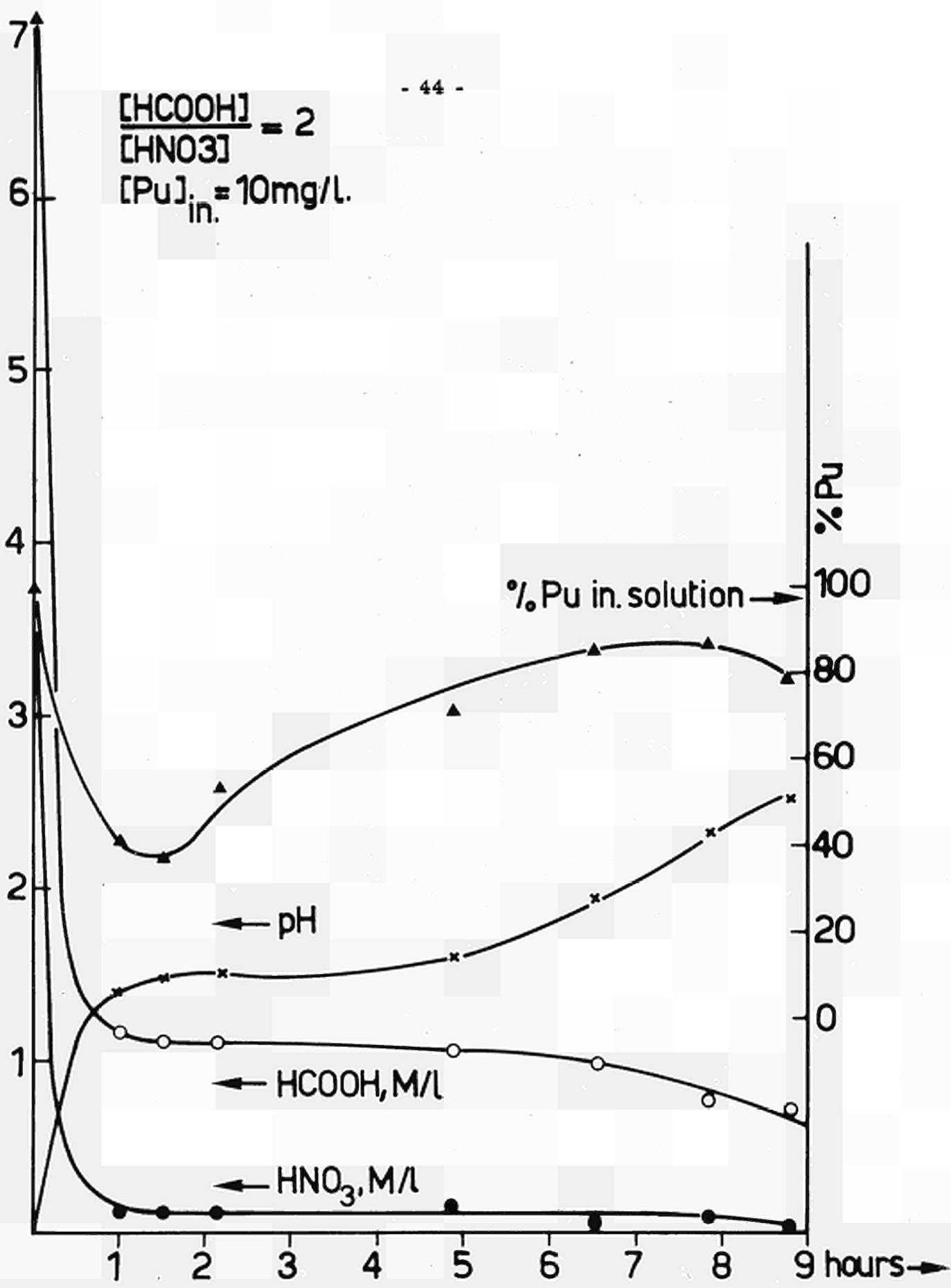
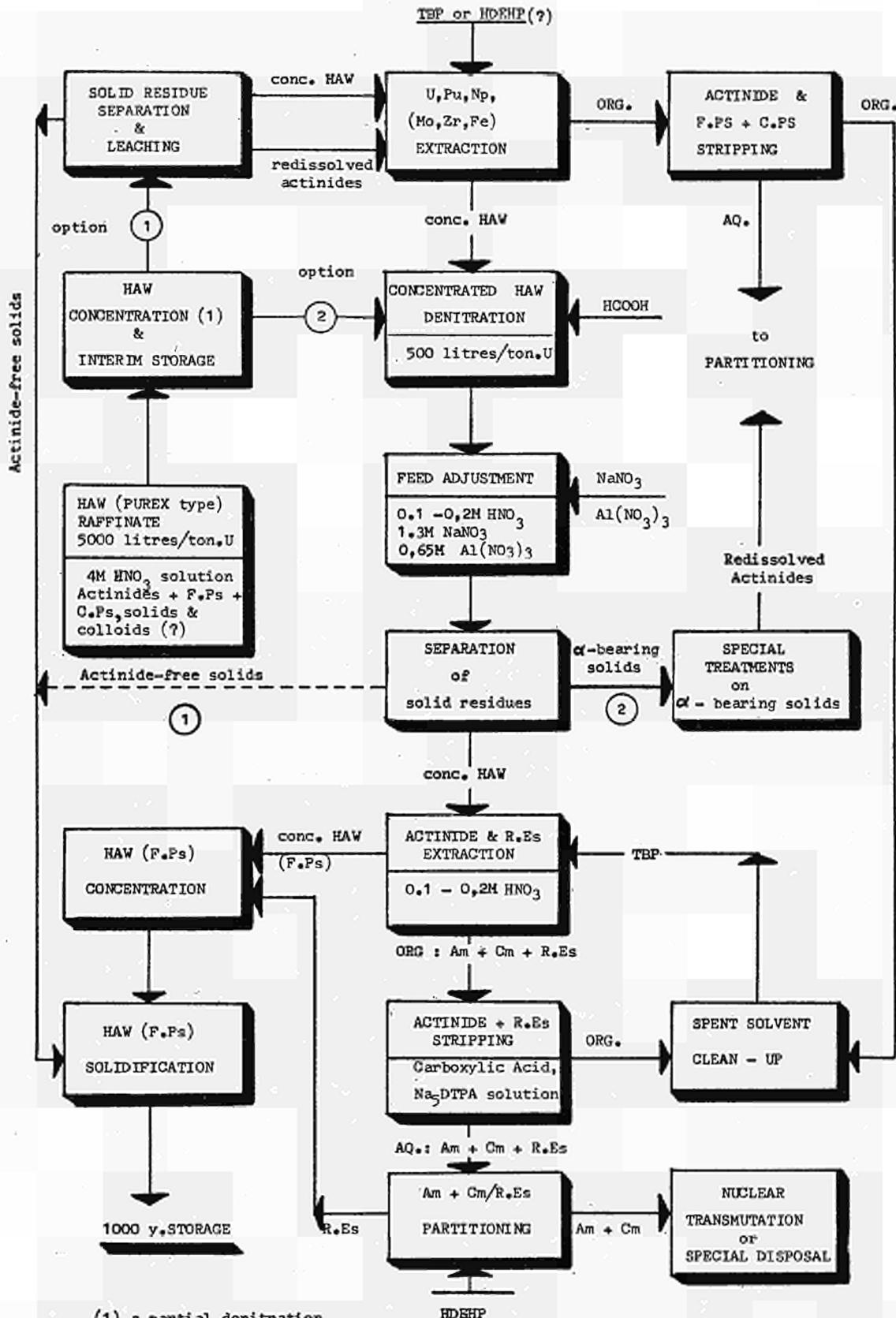


Fig.2 - DIRECT DENITRATION OF THE SIMULATED HAW SOLUTION: VARIATION OF pH, HCOOH & HNO₃ CONCENTRATION AND OF % Pu FRACTION IN SOLUTION AS A FUNCTION OF REACTION TIME



(1) a partial denitration is carried out

CCR - EURATOM ISPRA	TENTATIVE FLOW-SHEET FOR HAW PARTITIONING BASED ON ACTINIDE EXTRACTION BY TBP	Fig. 3
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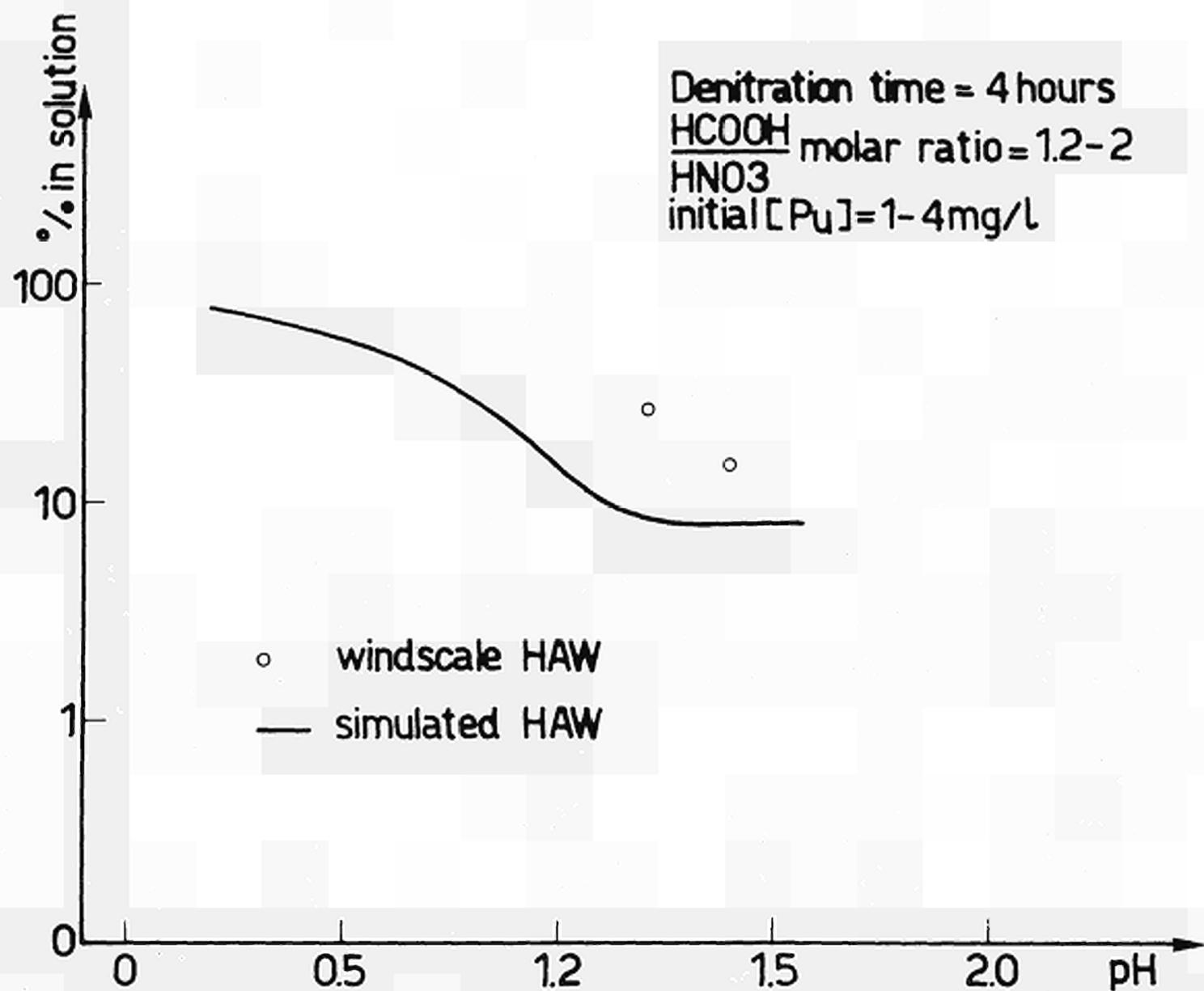


Fig.4- DILUTED HAW DENITRATION PROCESS: Pu % FRACTION REMAINING IN SOLUTION AS A FUNCTION OF pH

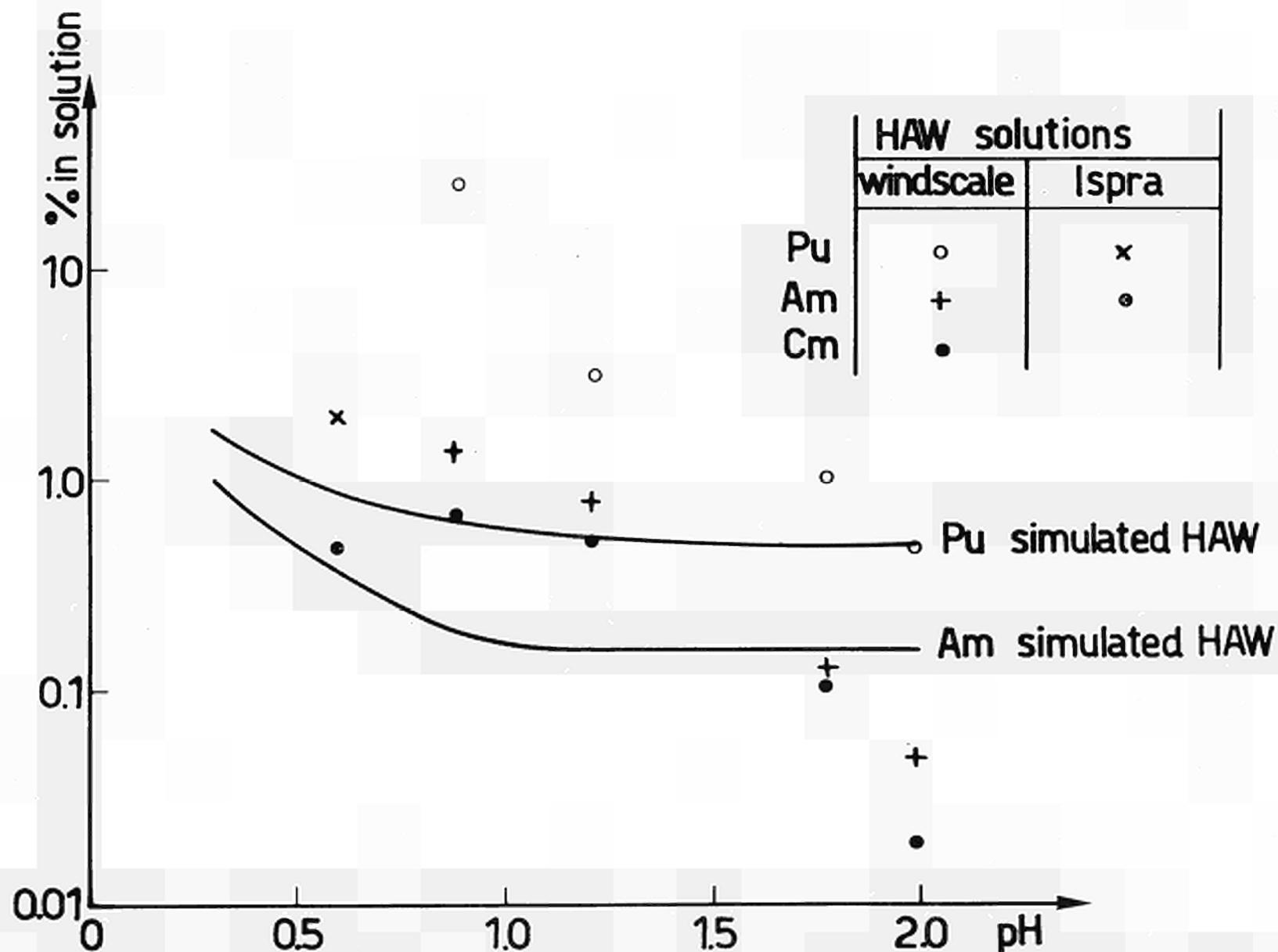


Fig.5 - OXALATE PRECIPITATION FROM DILUTED HAW (OXAL PROCESS): Pu,Am AND Cm % FRACTIONS REMAINING IN SOLUTION AS A FUNCTION OF pH

The given quantities (12) correspond to the HAW from 5 t of LWR fuel, burn-up 33,000 MWd/t, decay 150 d. It is assumed that only 10 percent Np is left in HAW

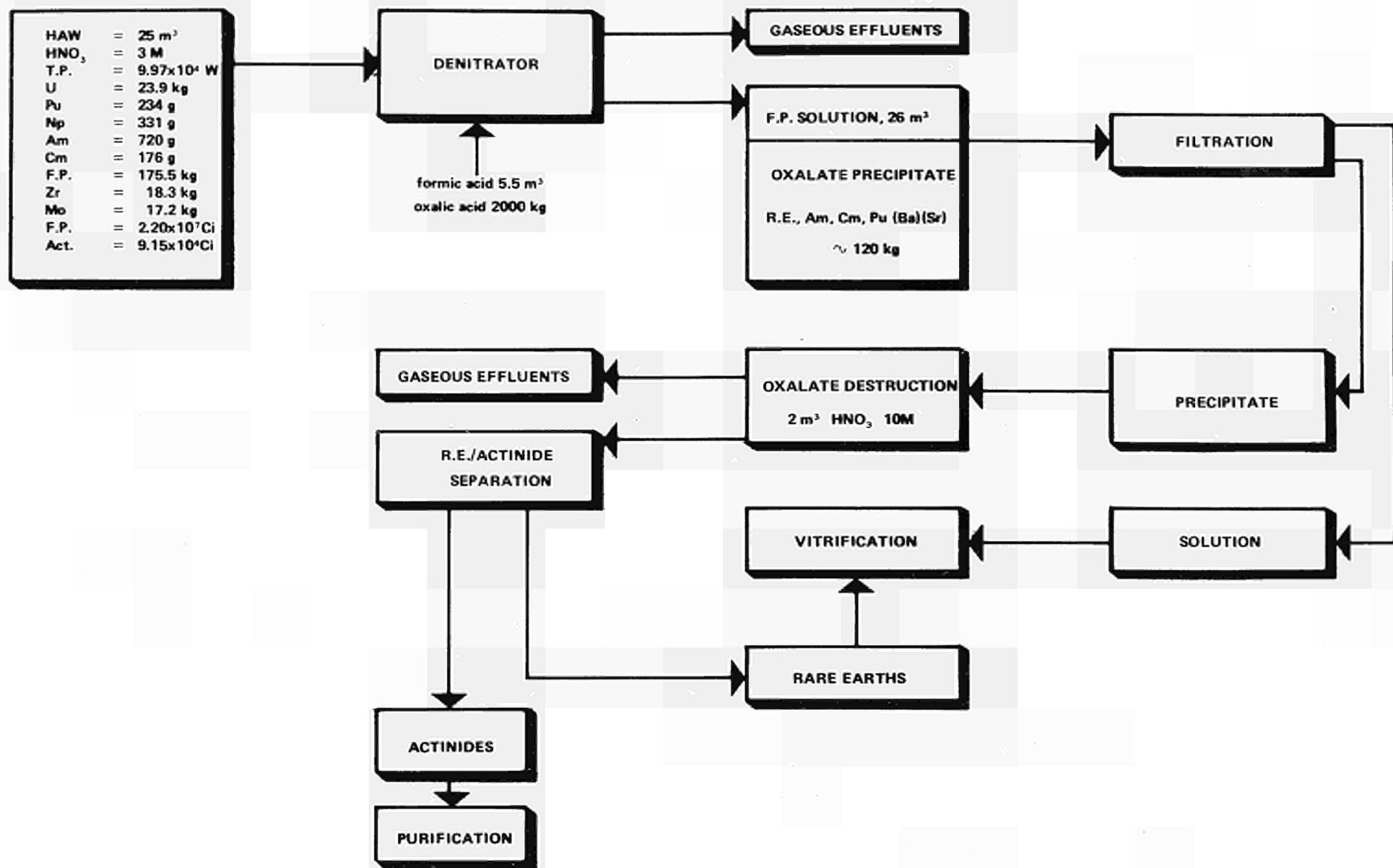


Fig. 6 – Flow-sheet of modified OXAL Process for diluted HAW solutions

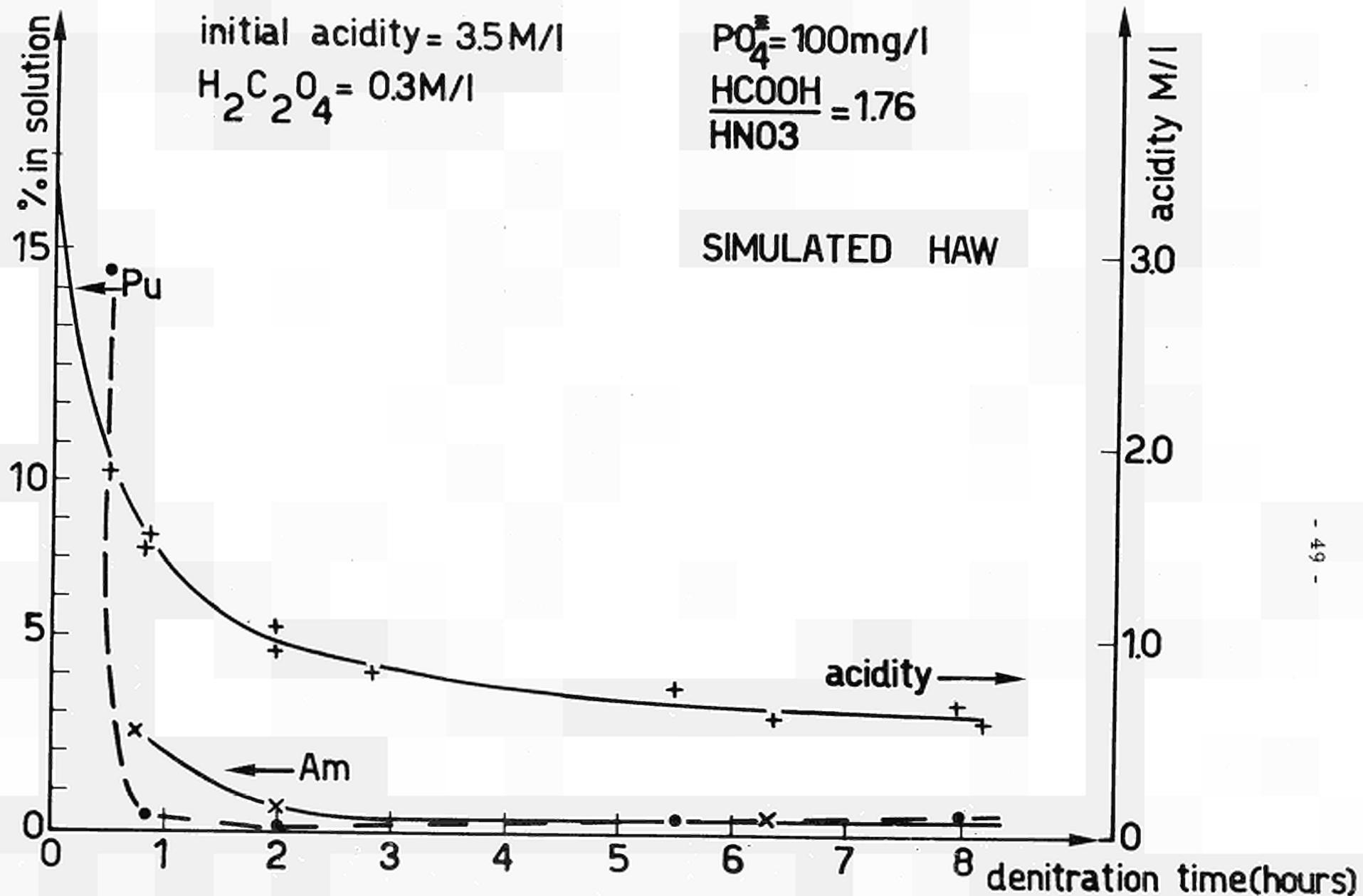


Fig. 7-DILUTED HAW DENITRATION IN PRESENCE OF $H_2C_2O_4$
(MODIFIED OXAL PROCESS): Pu AND Am %
FRACTIONS AND ACIDITY VARIATION AS A FUNCTION OF
DENITRATION TIME

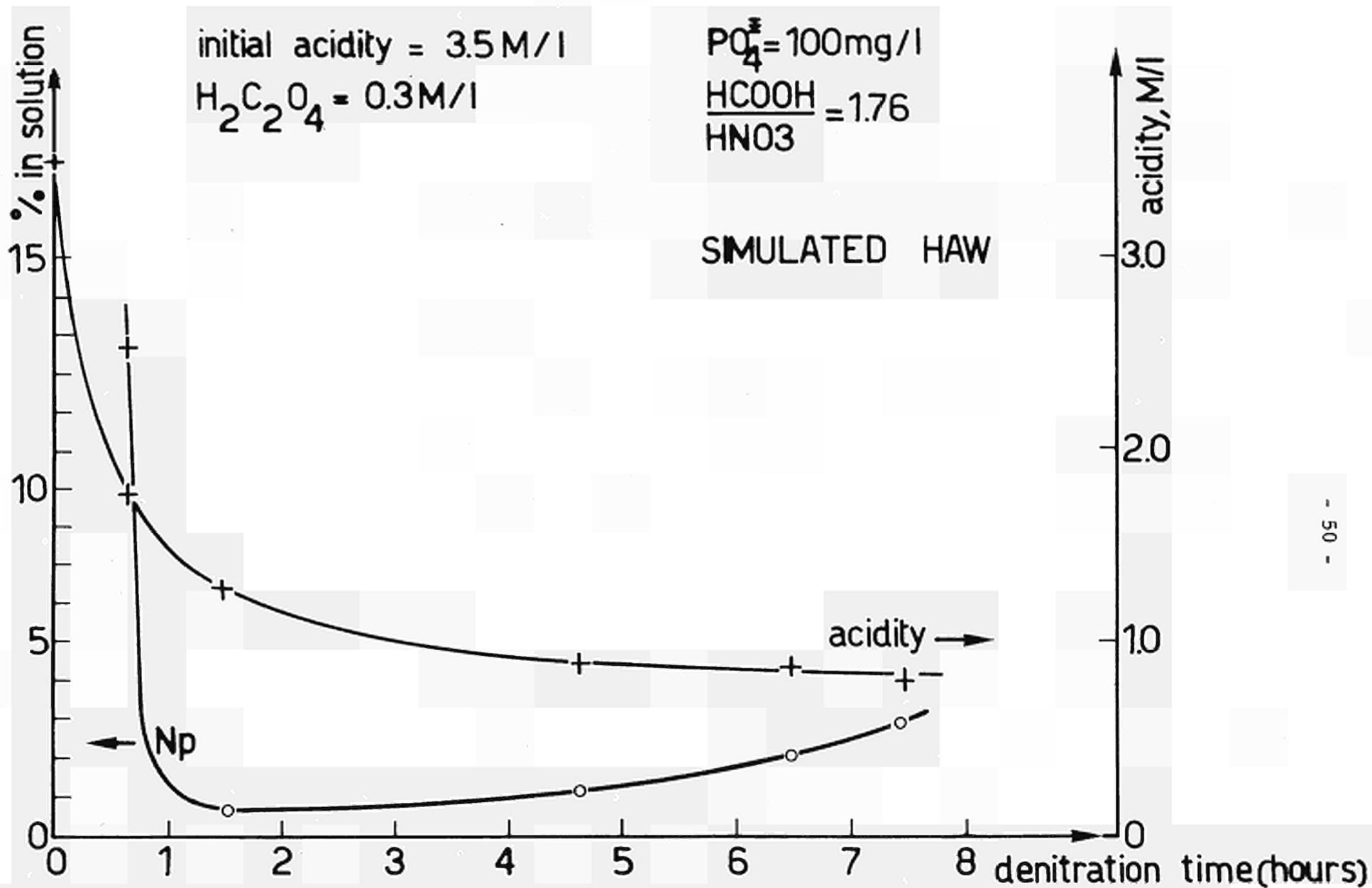


Fig.8-DILUTED HAW DENITRATION IN PRESENCE OF $H_2C_2O_4$:
 Np % FRACTION AND ACIDITY VARIATION AS A
 FUNCTION OF DENITRATION TIME

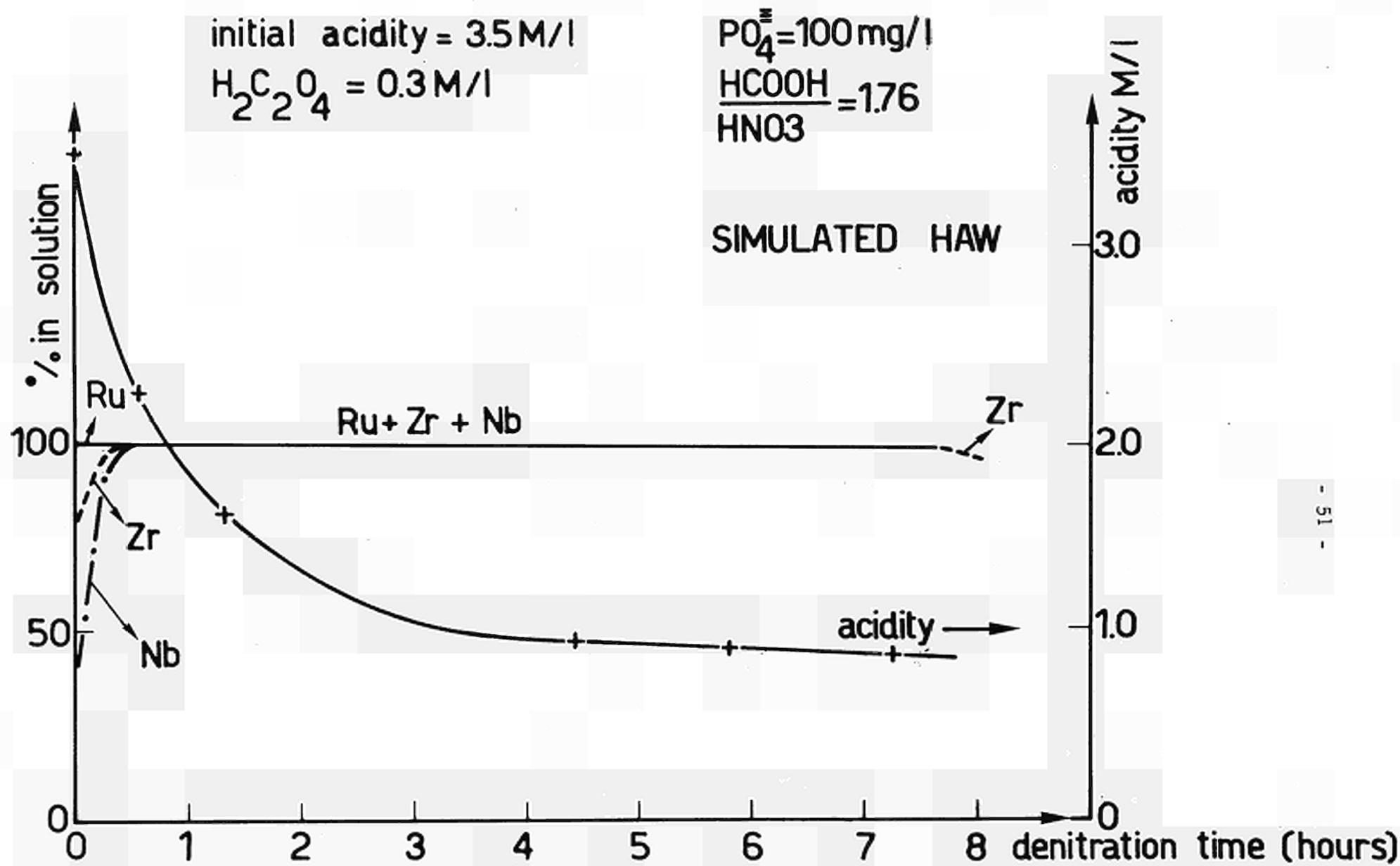


Fig.9-DILUTED HAW DENITRATION IN PRESENCE OF $\text{H}_2\text{C}_2\text{O}_4$:
 BEHAVIOUR OF Zr, Nb AND Ru

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2.2.2 Assessment Studies on Nuclear Transmutation of Actinides

OBJECTIVES

Taking account of the physical, technological, cost/risk implications, to demonstrate the feasibility of actinide transmutation in fission reactors, and to propose an overall strategy for transmuted the actinides produced by the European Community power generating system.

The time period covered by this progress report has been spent in organizing and preparing papers for the First Technical Meeting on the Nuclear Transmutation of Actinides, held at the JRC-Ispra on March 16-18, preparing an EUR-report and a survey paper for the IAEA Conference on Nuclear Power and its Fuel Cycle [8], performing sensitivity studies, studying the neutron physical feasibility of transmutation and collecting physical data.

RESULTS

Sensitivity Studies and Actinide Nuclear Data Requirements

The quality of nuclear and physical data plays an important role in the calculation of actinide concentrations during reactor operation (either operating under ordinary conditions or as actinide burner with or without recycling), or for health hazard studies.

An analytical evaluation of the influence on actinide build-up of various parameters such as initial concentrations, decay constants, branching ratios, nuclear cross sections, flux and time, has been performed and the corresponding report sent for publication [6].

This evaluation consists of establishing the transmutation schemes, writing up and solving the differential equations which describe the physical phenomena represented by the previous schemes, evaluating the sensitivities and identifying the most important nuclear data.

This approach has been applied to U-235 to 239, Np-237 to 239, Pu-238 to 242, Am-241 to 242, these actinides being produced in LWRs, HWRs, GCRs, AGRs and FBRs.

The agreement between analytical values of the sensitivities and the values obtained with a computer code is between 1 and 2%. It has been possible to recalculate all the data found in the literature. Some fundamental results which have appeared during this study are [4]:

- It is usually not possible to define a sensitivity coefficient which is a constant even for a relatively small domain of variation of a parameter. All the information which has been used for the evaluation of a sensi-

vity should be provided.

- The sensitivities strongly differ from one type of reactor to another; they strongly depend on the flux level in the reactor and on the reactor operation time.
- When a reactor is run at constant power, the sensitivity to a parameter is in fact a compound sensitivity since its value depends not only on the variation of the parameter but also on the variation of the flux in the reactor.
- A numerical evaluation of the influence of nuclear data uncertainties on recycle calculations and other results published in ref. [7] has also been applied to a LWR and a FBR [5]. A priority list to establish the most important nuclear data for neutron physical calculations has been given [4, 5, 6].

Neutron-physical Feasibility of Transmutation

A quantitative evaluation concerning the neutron-physical feasibility of transmutation has been made for a LWR and a FBR [2]. The inventory of actinides other than fuel amounts only to 0.13% (LWR) and 0.66% (FBR) of the total fuel loaded to the reactor, if a self-generated higher actinide recycling scheme is being applied. Then, the variation of rate of transmutation into fission products of the fast breeder lies within a range between 12.8 and 16.5% whilst the transmutation rate to fuel plus fission products lies between 21.8 and 24.7%. The corresponding values for homogeneous recycling in the LWR lie at about 20 and 50%, respectively.

The η -values of actinides other than fuel divided by those of normal fuel elements are a measure for the reactivity influence of higher actinides. They lie for the LWR in a range between 0.08 and 0.28 and for the FBR between 0.75 and 0.92. For the case of a self-generated actinide recycling scheme, these reactivity losses could be compensated by increasing the enrichment of the fuel of the LWR from 3.2 w/o to 3.32 w/o and of the fissile Pu in the core of the FBR from 12.3 w/o to about 12.5 w/o.

The additional reactivity losses due to lanthanide impurities have also been evaluated. For instance, if 1% of all lanthanides present in one ton of spent fuel (about 90 g in an LWR and 200 g in an FBR) will go with the higher actinides, their negative reactivity contribution could be compensated by an enhancement of the enrichment of 0.01% and 0.02%, respectively.

In the case of a separate reprocessing of the higher actinide stream, the rare earths fission products could have a considerably different composition than those generated in fuel. Therefore, an evaluation of the fission product yields for higher actinides could become necessary.

The maximum permitted specific power in Watts per length unit of a fuel pin is a limiting factor for the design of the fuel elements. Calculations for actinide target elements without using shielding constants for cross sections show that the specific power is about twice that of the fuel ele-

ment in the FBR, and five times in the LWR. Hence target elements must either be diluted by non-fissionable material or they must be designed in such a way as to reduce their specific power to the value of normal fuel elements.

All these results may be considered as a basis for investigating the technological feasibility of the recycle concept.

Technological Aspects of Actinide Transmutation

Gamma and neutron radiations complicate the actinide fuel fabrication operations, the most limiting factor being the spontaneous neutron emission from Cm-244 and Cm-246. It has been evaluated that the neutron activity of one kg of U-20% Pu-1% actinides ($8.05 \cdot 10^6$ n/sec) is about 40 times the activity of the same fuel without actinides ($2.14 \cdot 10^5$ n/sec). Fabrication of fuel elements using recycled Pu for FBRs requires shielding, anyhow, and it does not appear that the amount of actinides present in fuel elements to be used for homogeneous recycling will require a drastic change in the fuel fabrication operations. A review of the physical and chemical data necessary to evaluate the strategies associated with homogeneous and heterogeneous recycling of actinides has begun.

CONCLUSIONS

The results presented in this progress report were essential to the fulfillment of the objective. It should be noted, however, that a considerable amount of time, during the period covered by this report, has been devoted to the preparation of the First Technical Meeting on the Nuclear Transmutation of Actinides, to the preparation of several papers for this meeting and for the International Conference on Nuclear Energy held at Salzburg on May, and to the finishing up and editing of a EUR-report.

Because of these obligations it was not always possible to spend as much time as desired on the study of new questions.

Another point, which may be worth mentioning, is that the persons involved in the Assessment Studies of Actinides Transmutation appear to be too few to make sure that all the various questions entering in this complex study will be treated with the necessary attention. It has already been recognized as very desirable to increase the number of agents working in this study. During the, hopefully short, time when this manpower is not available, it is thought probable, without changing the objective of this study, to limit somewhat the efforts on technological study, on the comparative study of various types of reactors and possibly on risk analysis.

PLANNED ACTIVITIES

- Assessment of technological implications of keeping actinide in the fuel cycle
- Obtaining neutron physical data
- Calculations of actinide transmutation applied to different types of reac-

tors (thermal, fast)

- Preparation of a comparative method for risk analysis of the fuel cycle
- Review of performance of fuel elements of existing power reactors
- Collection of physical and chemical data for higher actinide important for fuel element design.

In the next reporting period progresses are expected in these activities: a detailed planning for the following period is given in Table 2 of Section 3.

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2.2.3 Actinide Cross Section Measurements

OBJECTIVES

The objective of the work is to improve the neutron cross section data for the physical assessment studies of the nuclear transmutation of actinides other than fuel. Sensitivity studies of the actinide transmutation rate [1, 2, 3] indicated that the fission cross section of Am-241, Np-237, Cm-244, Am-243 and Cm-245 are of greatest importance for the assessment of the actinide transmutation in fast reactors.

The availability of Am-241 targets and its importance for the actinide

transmutation suggested to develop the measurement technique and to start the first differential cross section measurement with this isotope. Two different techniques are applied. One relates the detected prompt fission neutrons, the other the fission fragments, emerging from actinide samples after bombardment with a pulsed neutron beam, to the fission neutron cross section.

In both methods it is intended to use the 3 MeV VDG of the Institute for Applied Nuclear Physics of the GfK as neutron source. For the generation of neutrons the Li-7 (p, n) Be-7 reaction is used. A first series of measurements were performed from July to October 1976 with the neutron detection method. The analysis of these experimental data and the elaboration of data processing programs are in progress.

Measurements of integral cross sections of actinides in reactor spectra are carried out at the Karlsruhe Establishment of the JRC, essentially within the framework of the Plutonium Fuels and Actinide Research programme. The results of these measurements will be utilized for the assessment studies on nuclear transmutation.

RESULTS

The Neutron Detection Method

In this method the fission cross section of Am-241 is measured relative to the one of U-235 by detection of the emitted neutrons per fission. The Van de Graaff accelerator is operated in pulsed mode. It irradiated 3 samples one after the other mounted in a 4-position sample-changer. Each sample position is exposed to the same time average neutron flux, controlled by a monitor, during several operation cycles of the sample-changer in order to eliminate drift effects.

The following samples were used:

- | | |
|------------|--|
| position 1 | Am-241 with 3.89 g AmO ₂ cladde with Al |
| " 2 | Dummy sample of 1 with identical structure but 3.99 g UO ₂ nat. |
| " 3 | U-235 disc with 7.107 g U-235 |
| " 4 | free |

The measurements were performed with a short flight path (distance actinide target - Li target) of about 6 cm with thick Li targets for the energy range below 100 keV and with one of 4 cm and thin Li targets for $100 \text{ keV} \leq E \leq 1 \text{ MeV}$. The prompt fission neutrons were detected in a proton recoil scintillation detector situated at a distance of about 28 cm from the actinide targets. For each adjusted proton energy of the VDG, 4 different time-of-flight spectra were obtained [4] which permit corrections for the isotopic composition and background effects.

These corrections lead to time-of-flight spectra for pure Am-241 and U-235 with a time-independent background superimposed by a prompt fission neutron peak and a well separate reduced capture gamma-ray peak.

The time-of-flight spectra were obtained with 3 different operation modes of the Li target.

- mode 1 The use of a thick Li target producing a white neutron spectrum permitted cross section measurements below 100 keV
- mode 2 Above 2.273 MeV the thin Li targets produce a second neutron group for which all data have to be corrected
- mode 3 In the neutron energy range $100 \text{ keV} \leq E \leq 800 \text{ keV}$ thin Li-targets generating monoenergetic neutrons could be used.

The detailed analysis of the measurements according to mode 3 has been started.

The signal-to-background ratio of the time-of-flight spectra was, below 140 keV, smaller than 1 due to the small fission cross section in this neutron energy region and the prominent time-independent neutron background caused by α -n reactions with the O_2 and Al present in the target. With higher neutron energies the signal-to-background ratio increased rapidly to reach values of better than 40 above 500 keV neutron energy. The signal-to-background ratio was for the corrected U-235 time-of-flight spectra in all cases better than 35.

The short flight paths between the Li target and the actinide samples and between Li target and the prompt fission neutron detector (proton recoil scintillation detector) demanded the use of a pulse shape discrimination of neutron pulses against gamma-pulses and a rather high cut-off energy for the detected prompt fission neutrons. The cut-off energy was estimated as $2.3 \text{ MeV} \pm 0.5 \text{ MeV}$. With the assumption of a Maxwellian prompt neutron energy spectrum [5] and a correlation of its mean neutron energy to the value of ν [6], about 7% U-235 prompt fission neutrons are less detected than the one of Am-241 at this threshold energy.

The average neutron energy spectrum causing reactions in the actinide target is as well affected by the short flight path. Neutron energy and intensity are a function of the angle existing between the monoenergetic protons and the emitted neutrons. From a measurement of the emitted neutrons at $\alpha = 0$ degree, the kinematics of the Li-7 (p, n) Be-7 reaction [7] and its neutron production cross sections as function of proton energy and angle, the average neutron energy spectrum and its resolution have to be calculated in order to define the correct energy and its resolution at which the cross section measurement has been executed.

Fig. 1 gives the neutron energy spectrum of a typical measurement at the position of the actinide target for an incident neutron beam of $\alpha = 0^\circ$ and the average incident neutron beam spectrum for the 30 mm diam. actinide targets in a 4 cm distance from the Li target. In this case the energy defined by the half height spectrum value has been reduced by about 10 keV and the mean spectrum by about 8 keV comparing the measured with the average incident neutron spectrum. The energy resolution remains practically unchanged but the low energy tail of the average neutron spectrum is increased as compared to the $\alpha = 0$ degree case.

The cross section values measured with the thin target have been analyzed in detail in the energy range of $140 \text{ keV} \leq E \leq 800 \text{ keV}$ (mode 3). All derived results depend strongly on the ν -values of U-235 and Am-241 as function of energy. For the cross section determination of Am-241 the $\sigma_f(\text{U-235})$ values reported by Sowerby /8/ and the α -values of Am-241, correlated by Hinkelmann /9/, were used (fig. 2).

Fission Fragment Detection

The Fission Fragment Detection Method requires less corrections than the neutron detection method and will therefore be used to control the normalization of the cross section data obtained. It has the advantage that only a small amount of fissile material is required for the cross section measurements, but its applicability is bound to an energy range in which the cross section to be measured reaches a sufficiently high value.

For the cross section measurements the same 3 MeV VDG will be used with thin Li targets, a collimated neutron beam and the time-of-flight technique, for background suppression. The fission fragments are detected in a gas scintillation chamber with 0.1% N_2 and 99.9% He as scintillator. The two targets have a thin backing of $250 \mu\text{g}/\text{cm}^2$ Al and subdivide the chamber into 3 optically separated thirds, such that fissions are identified by coincidence signals of fission products in two neighbouring chambers.

The test target will be a mixed target of 1 mg U-235 and 1 mg Am-241, the reference target will have 1 mg U-235. Exposing the chamber once to a neutron energy above the fission threshold of Am-241, once to a neutron energy below its fission threshold, the cross section of Am-241 is obtained without any intercalibration of the counting chains or corrections taking into account the different flight paths of the two targets. The pure U-235 target serves only as a monitor and its physical data do not enter into the analysis. The dominant advantage of this method is that only the cut-off energies of the chains with the mixed target enter into the analysis and that the fission fragment pulses of U-235 are statistically superimposed to the same α -pile up distribution as the Am-241 pulses.

The difficulties of the past were:

1. rupture of thin target backings,
2. excessive α -pile up due to gas scintillation
3. drift of photomultiplier signals as function of time
4. timing of the pulse height spectrum.

These have been overcome now and measurements to establish the most suitable operation conditions of the measurement chains are in progress.

CONCLUSIONS

The first analysis of the experimental results indicates, that in the region below 200 keV the obtained Am-241 fission cross section deviates considerably from the ENDFB-IV data file used at present for reactor calculations.

The final analysis of the experimental data did not advance as fast as originally planned, firstly due to some difficulties in the determination of the neutron energy from the recorded data and secondly due to the organization of an NEA-CEC meeting during March 1977 at the JRC-Ispra which deviated efforts.

PLANNED ACTIVITIES

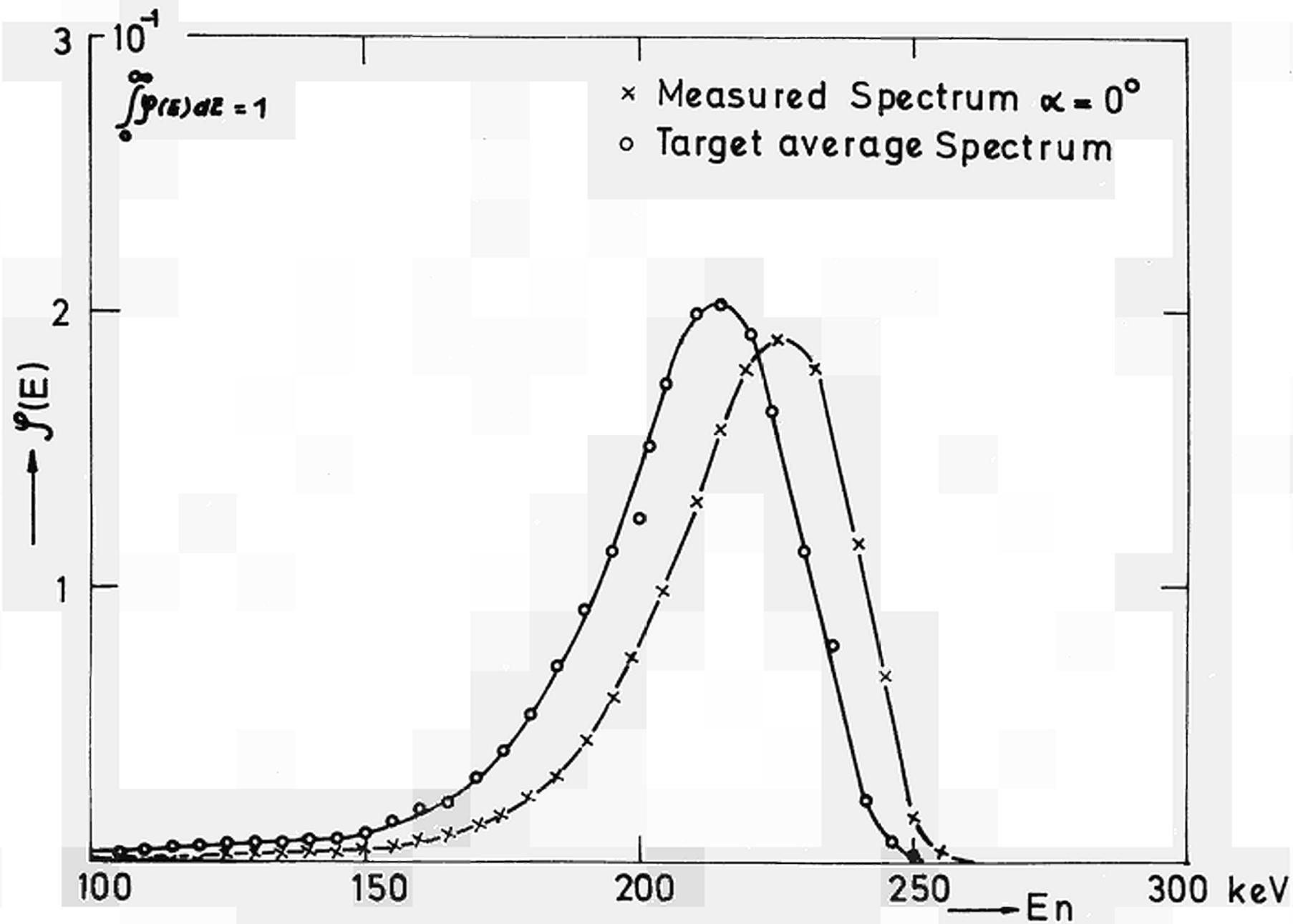
The final analysis of the experimental results obtained with the neutron detection method should be terminated by September/October 1977. During June 1977 the instrumentation for the fission product detection method will be set up at the GfK Karlsruhe for cross section measurements to be started during September 1977.

COLLABORATION WITH EXTERNAL ORGANIZATIONS

The objective makes use of the 3 MeV Van de Graaf, its operation, instrumentation and computer soft- and hardware existing at the Institute for Applied Nuclear Physics of the GfK, Karlsruhe. The experiments are performed in a common effort and the data analysis will be in charge of the JRC-staff.

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TARGET NEUTRON SPECTRA

Fig.1

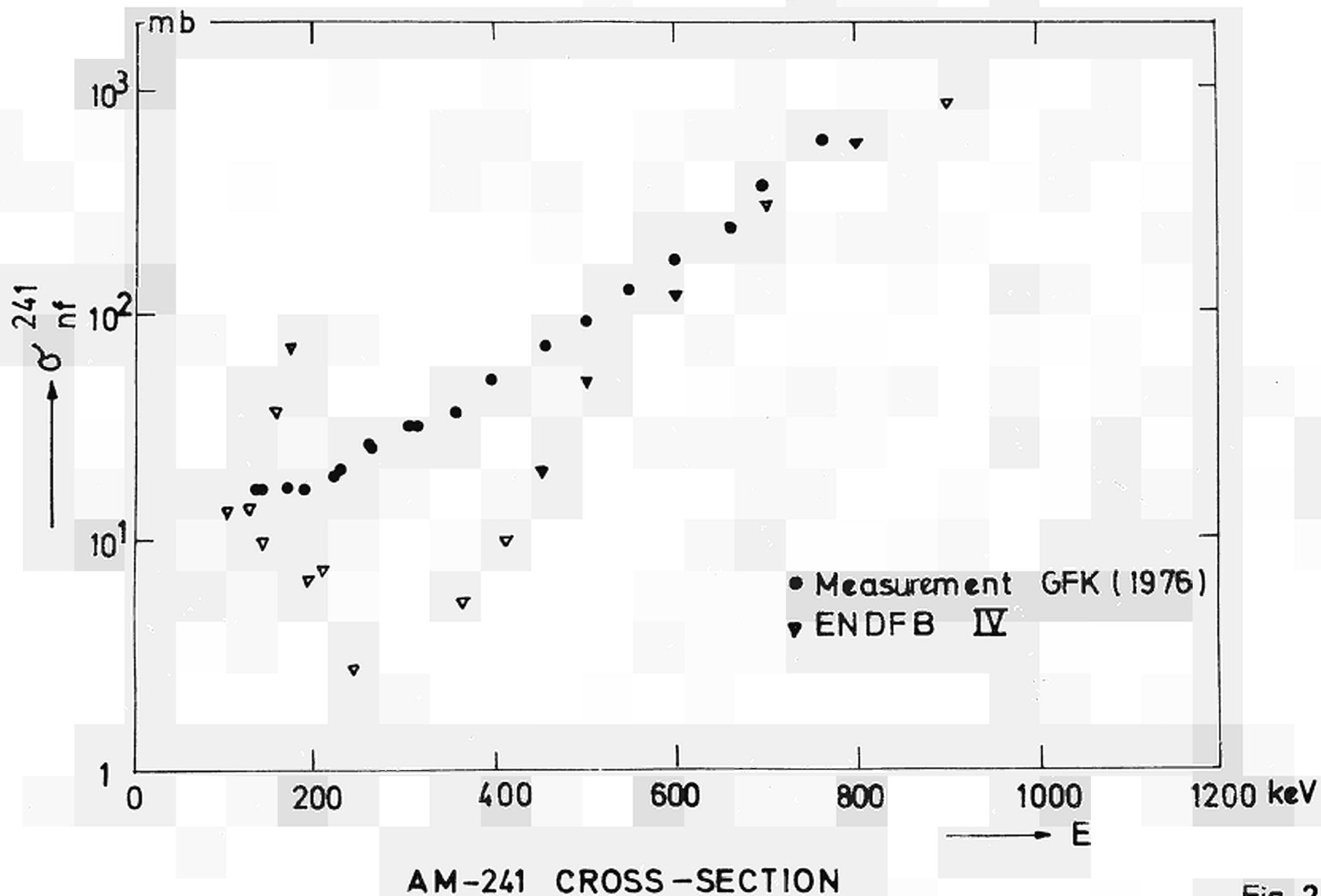


Fig. 2

2.3 DECONTAMINATION OF REACTOR COMPONENTS

OBJECTIVES

During the last pluriannual plan, in the framework of the programme "Support to Nuclear Power Stations", a small activity on the decontamination of reactor components was started. This activity was limited practically to the examination of some contaminated pieces and of some residues filtered from the water circuits of nuclear power stations.

At the time of the preparation of the present pluriannual programme, it was asked by the ACPM of the programme "Support to Nuclear Power Stations" that the study on the decontamination be continued and reinforced.

Decontamination can be intended in different ways. Firstly it can be intended as an operation which has the scope of sending the decontaminated equipment to a normal workshop or to a final repository (for this specific point decontamination is a synonymous of decommissioning). Secondly, it can be intended as an operation which has the scope of sending the decontaminated equipment to a controlled workshop. Finally, it can mean an operation performed in order to reduce the radioactivity of the piece or of the plant in order to facilitate the maintenance operation.

The activity of the first 6 months of 1977 was mainly directed to define the lines of work in this new field.

RESULTS

In order to be able to define better where lies the main interest of utilities, a contract has been passed to the firm Laborelec (Belgium). The scope of the contract is to perform an inquiry in the different power stations in order to define the need in Europe of decontamination and the scope of the decontamination operations.

In the meantime we are performing an extended bibliographic study which is centered mainly on the following subjects:

- evolution of the radioactive nuclides as a function of the operating time
- possible dependence of the intensity of radioactive contamination from reactor materials
- evolution of the composition of the contaminated layer with time with particular reference to the presence of cobalt
- decontaminating procedure with particular reference to chemical decontamination as far as partial decontamination is concerned and electrochemical method as far as total decontamination is concerned.

Some preliminary laboratory work has been initiated with the aim to have some techniques ready and to test some preliminary ideas. In particular, an existing high pressure laboratory loop which has been set up to study the influence of water chemistry on the corrosion and microsuspension production is in modification in order to allow the study

on particle deposition and on the effect of changing B and O₂ concentrations. Some tests on accelerated production of inactive crud has been conducted. Some preliminary tests on the use of an electrobrush for decontamination have been initiated.

PLANNED ACTIVITIES

Our aim is to be able to prepare a report on the state of the art for December 1977 which collects the bibliographic information and the results of the inquiry. As a consequence the research programme is not to be considered fixed; at least until the bibliographic review is completed and the inquiry is available some modifications are always possible. However, some principal lines of research can be drawn already now.

The radioactivity transport depends on the type of material and corrosion processes, on the purification system of the reactor, the time of life of the deposited layer. Therefore it does not seem possible to define completely decontamination methods. For every action of decontamination, a procedure adapted to the specific conditions must be defined by the operational staff. The best help that research can offer regards the basic knowledge of the mechanism of film formation and of the action of the decontaminating methods.

The main lines of research therefore will be:

- mechanism of deposition of contaminated particles,
- evolution of film composition and morphology as a function of time,
- analysis of the mechanism of chemical decontamination,
- analysis of the mechanism of electrochemical decontamination.

In particular we plan to carry out some preliminary tests on the mechanism of chemical decontamination using scanning electron microscope techniques and radiochemical techniques.

A test on the influence on the film dissolution of the oxygen concentration will be performed in the existing loop.



Conclusions

3. CONCLUSIONS

The programme 1977-1980 of the Joint Research Centre in the field of Nuclear Materials and Radioactive Waste Management includes the continuation of some activities of the previous programme 1973-1976 and the beginning of some new activities.

The increase of the staff allocated to the programme and the conclusion of some activities of the previous programme made possible the concentration of a larger effort on the projects "Evaluation of Long-Term Hazard of Radioactive Waste Disposal" and "Chemical Separation and Nuclear Transmutation of Actinides".

In particular the first project was reinforced by:

- Introduction of a new competence in oceanography by external recruitment
- Beginning of the new activity on the interaction of actinides with the environment
- An increase of staff in the activity Long-Term Behaviour of Conditioned Waste, which makes possible studies on the validity of simulated experiments of radiation damage on glasses.

The second project was reinforced by:

- An increase of staff in the chemical separation of actinides mainly to accelerate the work in hot cells
- An increase of staff in the assessment studies on nuclear transmutation of actinides mainly to make possible the evaluation of technological implications of the actinide recycling. A further increase in this field is planned.

The third project "Decontamination of Reactor Components" is the development of a small action started during the previous years.

The competences for this type of study are essentially available.

Not all the staff problems are actually solved. There is an important need of competences in chemical engineering for various points of the programme. This need can be partly covered by the recruitment of a chemical engineer planned for the second half of 1977. We also plan to solve some chemical engineering problems by contracts with external organizations.

Competences in nuclear physics are also needed for the activity on actinide monitoring. Due to the very limited possibilities of external recruitment we plan to obtain external contributions by an expert contract.

The delay in the distribution of the 1977 budget has not heavily affected the work in the first months of the year. The effect of this delay will be much more important in the second half of 1977 mainly for the new experimental actions which require initial investments.

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 first half of 1977 the work performed in the previous year in the field of waste hazard analysis has been reviewed and summarized in two reports.

The organization at Ispra, in cooperation with the OECD Nuclear Energy Agency, of the workshop on "Risk Analysis and Geological Modelling", has made possible a valuable confrontation of our results with those of various external organizations.

The planning for future activities in the field of waste hazard analysis includes the improvement and extension of the models previously developed and their application to specific sites in the framework of collaborations with national organizations (see Table 1). In this way the results of the JRC activities are of direct use for the member countries.

The work on the long-term behaviour of conditioned waste, directed to provide input data for the waste hazard models, has been continued by means of experiments on radiation damage and long-term leaching of glasses. The development of the experimental work is considered satisfactory in spite of some delays in the analysis of irradiated samples in hot cells.

The work has been extended to the long-term behaviour of bituminized waste. In fact, the waste hazard analysis has shown that a large part of the long-term hazard can derive from alpha-contaminated waste conditioned with bitumen.

The planning for future activities is shown in Table 1.

The knowledge of the behaviour of the actinides in terrestrial and aquatic environments following loss of segregation of the geological repository, is of greatest importance for the waste hazard analysis.

A literature survey has been carried out to identify the areas of major interest. Due to the preparative work, done during 1976, it was possible to start the laboratory activities already in the first months of 1977.

A planning of the future activities is given in Table 1.

In the field of actinide monitoring the use of spontaneous fission neutron measurements has been investigated during this period in view of the preparation of a new chapter of the guide on monitoring of plutonium contaminated waste. The experience gained in the past years has been used to help operators of plutonium fabrication plants (CNEN-Casaccia) in solving problems of fuel management.

A planning for the future activities is reported in Table 1.

Chemical Separation and Nuclear Transmutation of Actinides

The leading role assigned to the Commission, by the Nuclear Energy Agency, in the field of the studies for the nuclear transmutation of actinides, is a recognition of the considerable effort spent on that subject by the Commission in the Joint Research Centre and in the framework of

the Indirect Action.

The "Technical Meeting on the Nuclear Transmutation of Actinides", organized at Ispra in March 1977 made possible a valuable confrontation of our activities with those of other organizations.

In the field of chemical separation of actinides promising results have been obtained in the experiments with the OXAL process. For what concerns solvent extraction the activity has been concentrated on the use of HDEHP and TBP.

The planning of the future activities (Table 2) is directed to produce for the end of 1979 a critical evaluation of the perspectives of the chemical separation of the actinides. In addition to the laboratory work, engineering evaluations of the various processes under development are required.

Our planning could be modified in the future in order to make possible a repartition of tasks with other organizations interested in a collaboration in this field.

In the field of the assessment studies for nuclear transmutation of actinides considerable progress has been made concerning nuclear code calculations of actinides formation and transmutation.

Studies have also been started concerning the technological implications on the fuel cycle of the actinide recycling.

The planning of the future activities (Table 2) is directed, as for the chemical separation, to the preparation of a report before the end of 1979. Also this planning can be influenced by possible external contributions. We plan to reinforce the activity in this field in 1978 by a reduction of the effort spent on the actinide cross section measurements.

The activity in the field of the actinide cross section measurements has produced new data for the fission cross section of Am-241. The results of the measurements are particularly interesting due to the large difference in respect of the previously accepted values. The planning of the future activities is reported in Table 2.

Decontamination of Reactor Components

The activity in the first half of 1977 was mainly directed to identify the areas of major interest, by means of an enquiry under contract with an external firm and by means of an extended literature survey.

This activity will result, in the second half of 1977, in the preparation of a report on the state of the art in the field of decontamination.

The experimental activities planned for the second part of 1977 concern the mechanisms of the chemical decontamination and the influence of the oxygen concentration on the film dissolution.

At present it is not possible to draw a planning for a longer period.

For further information, please contact the Programme Manager.

Table 1 : Planned activities and important milestones for the project 1 : Evaluation of the long-term hazard of radioactive waste disposal

Activities	Next period for 1977 (month)						1978	1979	1980
	7	8	9	10	11	12			
Waste hazard analysis							①		
A									
B							②	③	
Long-term stability of conditioned waste									
C								⑤	
D					④			⑥	
E						⑦		⑧	⑨
Interaction of actinides with the environment								⑩	
F									
G									
Actinides monitoring							⑪	⑫	⑬
H									

- | | | | |
|---|---|---|--|
| A | Modelling of failure of geological barrier | ① | Completion of the model for clay formations (site specific) and start-up of the preparation of models for other geological formations |
| B | Actinides distribution in environment following failure of geological barrier | ② | 1977 Revision (more detailed waste inventory, more detailed terrestrial environment, introduction of marine environment) |
| | | ③ | 1978 Revision |
| C | Leaching tests on vetrified waste | ④ | Completion of the experiments in pure water |
| | | ⑤ | Completion of the experiments in conditions of geological disposal |
| D | Leaching tests on bituminized waste | ⑥ | Completion of the experiments on bituminized waste and possible start-up of experiments on other matrices |
| E | Experiments of radiation damage in glasses | ⑦ | Completion of the post-irradiation analyses on the glasses irradiated in HFR Petten |
| | | ⑧ | Completion of the experiments on the radiation damage simulation by fission fragments |
| | | ⑨ | Completion of the experiments on the validity of accelerated tests |
| F | Interaction with abiotic environment | ⑩ | Conclusion of initial column (soil and sediments) experiments with leached actinides and development of further experimental programme |
| G | Interaction with biosphere | | |
| H | Plutonium waste monitoring | ⑪ | Completion of the chapter IV of the Guide "Application of passive neutron assay" |
| | | ⑫ | Completion of the chapter V of the Guide "Application of active neutron assay" |
| | | ⑬ | Revision of the Guide |

Table 2 : Planned activities and important milestones for the project 2 : Chemical separation and nuclear transmutation of actinides*

Activities	Next period for 1977 (month)						1978	1979	1980
	7	8	9	10	11	12			
International coordination	A								
Chemical separation of actinides	B						①	② ③	⑦
	C					④		⑤	⑥
	E						⑧	⑨	⑩
Assessment studies on nuclear transmutation of actinides	F							⑪	⑫
	G						⑬	⑭	⑮
	H							⑯	⑰
	I						⑱	⑲	⑲
Actinides cross section measurements	J								
	L								

* The edition of a summary report and the preparation of a new planning is foreseen for the end of 1979

A In the framework of the activity of the OECD Nuclear Energy Agency

B OXAL process

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

C Solvent extraction

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

E Reactor physics calculations

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

F Collection of chemical and physical data for fuel element design

- 11 Proposal for fuel element designs containing actinides other than fuel
- 12 Elaboration of an overall recycle strategy

G Cost and risk analysis

- 13 Establishment of cost calculation procedure
- 14 Establishment of the methodology for risk assessment
- 15 Results of cost and risk assessment

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

- 16 Proposal of changes in a fabrication plant
- 17 Preparation of a report on the feasibility of the actinides transmutation

I Differential cross section measurements

- 18 Completion of the measurements on Am-241 and Start-up of measurements on Np-237 and Am-243

L Integral cross section measurements

4. JRC PUBLICATIONS

- 1) F. GIRARDI, G. BERTOZZI, M. D'ALESSANDRO, "Long-Term Risk Assessment of Radioactive Waste Disposal in Geological Formations", paper presented at the 1977 Annual Meeting of the American Nuclear Society, New York, 12-17 June 1977; to be printed as EUR-report
- 2) G. BERTOZZI, M. D'ALESSANDRO, F. GIRARDI, M. VANOSI, "Safety Assessment of Radioactive Waste Disposal into Geological Formations: A Preliminary Application of Fault-Tree Analysis to Salt Deposits", paper presented at the "Workshop on Risk Analysis and Geologic Modelling in Relation to the Disposal of Radioactive Wastes in Geologic Formation", Ispra, 23-27 May, 1977; to be printed as EUR-report
- 3) F. LANZA, "Long-Term Leaching of Borosilicate Glasses", to be printed as EUR-report
- 4) G. BIRKHOFF, "Euratom Activity in Monitoring Plutonium-Contaminated Waste Streams", paper presented at the Annual INMM Meeting, Washington, June 1977
- 5) F. MANNONE, "Removal of Actinides from High Activity Wastes by Solvent Extraction: Outline of the Research Work at Ispra JRC Laboratories", EUR 5527-e (1976)
- 6) F. MANNONE, L. CECILLE and D. LANDAT, "Removal of Long-Lived Actinides from Purex Type HAW Raffinates by Solvent Extraction: The Use of Neo-Tridecano Hydroxamic (HX-70) as Extractant", EUR 5537-e (1976)
- 7) F. MANNONE, L. CECILLE, "Removal of Long-Lived Actinides from Purex Type HAW Raffinates: Development of Conceptual and Experimental Studies on Solvent Extraction"; to be printed as EUR-report
- 8) L. CECILLE, M. LE STANG, F. MANNONE, "Séparation des actinides des solutions de déchets nucléaires à haute activité (HAW) au moyen de l'acide di (éthyl-2-hexyl)phosphorique (HDEHP). 1ère partie: Résultats expérimentaux obtenus en batch sur une solution "HAW" simulée"; to be printed as EUR-report
- 9) L. CECILLE, D. LANDAT, F. MANNONE, "Séparation des actinides des solutions de déchets nucléaires à haute activité (HAW) par extraction par solvant. I: Description d'un schéma de séparation faisant appel au TBP"; to be published in "Radiochemical and Radioanalytical Letters"
- 10) L. CECILLE, H. LE STANG, F. MANNONE, "Séparation des actinides des solutions de déchets nucléaires à haute activité (HAW) par extraction par solvant. II: Description d'un schéma de séparation faisant appel à l'HDEHP"; to be published in "Radiochemical and Radioanalytical Letters"
- 11) F. GIRARDI, J. GODFRIN, F. MOUSTY, J. TOUSSAINT, "Séparation des actinides des déchets liquides de haute activité - Le procédé OXAL"; to be published in "Radiochemical and Radioanalytical Letters"

- 12) F. MANNONE, L. CECILLE, D. LANDAT, "The Neo-Tridecano-Hydroxamic Acid as an Extractant of Long-lived Actinides from PUREX Type HAW Raffinates"; paper to be presented at the International Solvent Extraction Conference 1977 (ISEC 77) - Toronto, Canada, 10-17 September 1977
- 13) L. CECILLE, D. LANDAT, M. LE STANG, F. MANNONE, "Séparation des Actinides des Solutions de Déchets Nucléaires à haute Activité par Extraction par Solvent - Etat des travaux expérimentaux réalisés au CCR-Ispra", Proceedings of the First Technical Meeting on Nuclear Transmutation of Actinides, JRC-Ispra, 16-18 March, 1977; to be published
- 14) F. MOUSTY, J. TOUSSAINT, J. GODFRIN, F. GIRARDI, "Séparation des Actinides des Déchets Liquides de Haute Activité - Le Procédé Oxal", ibidem
- 15) W. HAGE, E. SCHMIDT, "Reactor Physics Aspects of Burning Actinides in a Nuclear Reactor", ibidem
- 16) E. SCHMIDT, J. CAMETTI, "On the Neutron Physical Feasibility of Transmutation of Actinides other than Fuel in Nuclear Power Reactors", ibidem
- 17) A. SOLA, "Some Preliminary Results on Actinide Incineration and Transmutation in a Thermal and in a Fast Reactor", ibidem
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- 22) A. SOLA, "Analytical Evaluation of Actinide Sensitivities", EUR-5763e (1977)
- 23) E. SCHMIDT, "Predictions for the High-Level Active Waste to be Generated by Nuclear Power Stations in the Member States of the European Community", EUR-5690e (1976)
- 24) L. CECILLE et al., "Nuclear Transmutation of Actinides other than Fuel as a Radioactive Waste Management Scheme", IAEA-CN-36/366 (1977).

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CDNB06476ENC