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Multiannual programme of the Joint Research Centre 1980-83

1983 Annual Status Report

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SAFETY OF NUCLEAR MATERIALS

1983

Research Staff: 52
Budget: 6 095 000 ECU
(committment appropriations)

Projects:

— Risk evaluation
— Protective barriers
— Actinide separation
— Actinide monitoring

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1. INTRODUCTION

The management of radioactive wastes produced in the exploitation of nuclear energy at an industrial level, requires the solution of various technical problems related to the treatment and conditioning of a large variety of waste forms and to their storage and disposal. In particular it is necessary to set-up solutions which ensure the safe disposal of long-lived radionuclides for extremely long time periods.

A Community action in the field of radioactive waste management is motivated by the pre-eminence of the public service aspects over the economic aspects, the fact that the radioactive waste management problems involve directly or indirectly all Member States and the fact that a Community approach affords stronger guarantees in terms of best possible protection of the public and of the environment.

The Commission is operating in the area of radioactive waste management R and D by means of a programme carried out at the Joint Research Centre (Programme Safety of Nuclear Materials) and by means of a shared cost action programme.

In addition to the research programmes, the Council approved in 1980 a Community Plan of Action in the field of Radioactive Waste (1980-1992). This plan entrusts the Commission with a wider role in the implementation of the waste management practices, such as:

— continuous analysis of the situation with a view to the adoption of the necessary solutions.
— examination of measures which could assure the long-term or permanent storage of waste under optimum conditions.
— consultation on waste management practices.
— continuity of Community research and development work during the plan.
— providing the public with regular information.

The programme Safety of Nuclear Materials is part of the JRC activity in the field of Nuclear Safety and the Fuel Cycle; the staff allocated to the programme consists of 52 research.men corresponding to about 5% of the total JRC research staff.

The central theme of the JRC activity is the study of the long-term safety of radioactive waste management.

Research is carried out essentially in three directions:

— the evaluation of the long-term risk linked to geological disposal of radioactive waste, carried out both by theoretical assessment studies (risk evaluation) and by experimental activities aimed at providing the necessary data base (protective barriers).
— the study of possible modifications of the fuel cycle to keep hazardous radionuclides at the lowest possible level in waste (actinide separation).
— the study of analytical control procedures for actinides in waste (actinide monitoring).

2. RESULTS

A. RISK EVALUATION

The long-term risk of radioactive waste disposal in geological formations is studied by an approach based on the evaluation of the barriers provided between disposed waste and man.

The JRC activity in this area includes the development of models and codes for the assessment of the long-term safety of disposal in geological formations (continental and marine) and their application to specific sites.

Special attention is being given to the conceptual problem of risk quantification. Waste disposal in geological formations is based upon the geologist's belief that, on the basis of past history and present conditions, it is possible to identify formations which, among the numerous existing, are likely to remain unaltered for very long periods of time and to assure that the enclosed wastes will be adequately isolated.

The JRC approach to assess the long-term risk associated to such an option is based on the separate assessments of release probabilities and consequences for each scenario of loss of geological isolation, followed by quantification of the risk as the product of probabilities and consequences. A special attention is given to the problem of input data uncertainty. Parameters affected by large uncertainties are described by probability distribution functions, and statistical methods are used to calculate the overall uncertainty and the dominant risk parameters.

This approach enables the comparison with preestablished acceptability criteria (for instance, the natural radiation background) both in terms of dose and of risk.

The methodological approach for the risk assessment is now completely defined. The way to combine the probability and consequence terms into an overall figure of risk is clarified.

The code LISA utilyzing the Monte Carlo method with the Latin Hypercube Sampling Technique to handle data distributions is fully operational.

During '83, the code LISA was used for the analysis of the risk linked to a waste repository possibly mined in the clay of Boom (Belgium). Different probabilistic scenarios were considered; among them, the most relevant are the Faulting Scenario and the Glacial Erosion Scenario. For them, a complete risk assessment is being performed. Preliminary results of the analysis show that the Faulting Scenario does not pose major problems, because both probabilities of occurrence are very low and consequences are not severe, so that the overall risk appears quite acceptable.

The situation is less favourable for the glacial erosion scenario; in this case potential exposures caused by such an occurrence could be more relevant. However the overall risk turns out to be acceptable, because of the very low value of the scenario's probability. The situation appears similar to many risks of every day life, that are frequently accepted only on the basis of the low event probability, independently of the level of the event consequences.

The PAGIS action (Performance Assessment of Geological Isolation System), developed in the frame of the Community Plan of Action on Waste Management, was continued. Phase 1, which aimed at providing a complete data and models base for the risk analysis, has been completed with the preparation of a Summary Report.

The application of the code LISA to the sub-seabed option for waste disposal is also underway. An initial choice of models to describe the biosphere (world oceans) and geosphere (30 m of seabed sediments) unique to this option has been made. This has allowed a demonstration probabilistic assessment to be carried out with a sensitivity analysis to identify the relative importance of each sub-model.

Improvements in these models and the input data are now being made to obtain a better description of the system. This work is continuing to be persued in close co-operation with the Radiological Assessment Task Group of the NEA International Seabed Working Group. In this framework various intercomparisons are being set up to test the biosphere and geosphere models and data and to encourage where necessary improvements to be made.

In order to study the emplacement of HLW into deep ocean geological sedimentary formations investigations on the use of free-fall penetrators were undertaken in 1983.
The first tests in this programme were carried out on RRS Discovery Cruise 134 in the Great Meteor East radioactive waste disposal study area (GME) in March 1983 as a collaborative experiment between the Building Research Establishment, the Joint Research Centre and the Institute of Oceanographic Sciences of the U.K.
The GME area lies in the southwestern distal part of the Madeira Abyssal Plain. Extensive geological and geophysical studies have been carried out there over the past few years by the Rijks Geologische Dienst of the Netherlands and by the U.K. Institute of Oceanographic Sciences.
Four similar penetrators were dropped in the GME area, each measuring 3.25 m in length and 0.325 m in diameter. They were constructed of solid steel and weighed 1.8 tonnes in air. The design of the penetrators was based on a detailed hydrodynamic analysis of a number of possible configurations of nose shape length/diameter ratio and stabilising surfaces carried out for the JRC by Aermacchi S.p.A.
The first two penetrators were dropped about a kilometre apart in a location where previous coring and the ship's 3.5 kHz profiler records indicated negligible quantities of sand in the upper part of the sedimentary column. Good acoustic penetration on 3.5 kHz records is obtained in regions with little or no coarse sediment: that achieved at the first site was 80 m. Very similar records of frequency versus time were obtained from the first two impacts, with the acoustic signal being detected to the full penetration depth (zero Doppler shift) in each case so it was decided to move to second site 65 km to the east, where a sandy layer was known to be present in the sediment column from the Dutch coring, for the remaining two drops. The penetrators were dropped but the signal from the last one was lost soon after impact and its penetration history was not recorded.
Depths of tail penetration were between 30-32 m at the two sites.

B. PROTECTIVE BARRIERS

This project deals with experimental studies in support and validation of theoretical modelling studies. The activity is essentially aimed at:

— determining the long-term stability of waste in conditions of geological disposal, thus providing the source term of radionuclides which may escape geological confinement.

— studying the physico-chemical laws which control the migration of radionuclides through the geosphere to the biosphere.

Long-term stability of conditioned waste

For what concerns leaching of borosilicate glasses utilized for solidification of high level waste, the major effort has been spent to study the leaching in normal repository conditions i.e. with clay, or sea sediments.

A mathematical model has been set up. In this model the weight loss of the sample is related to the dissolution and diffusion of SiO_2 in the porous media. Experiments conducted in order to validate such a model have confirmed the general picture. It has been verified that at the beginning of the experiment the weight loss rate can be higher than the equilibrium weight loss due to the fact that SiO_2 can be absorbed by clays. However after more than 100 days the model seems to describe correctly the leaching phenomena.

The mathematical model allows also to evaluate the release of the various radioactive ions.

For modelling purposes ions have been grouped in three categories:

— ions which diffuse rapidly in glass and are readily soluble (ready availability for geochemical transport).

— ions which are readily soluble but do not diffuse in glass (availability controlled by matrix disgregation).

— ions which do not diffuse in glass and which are present as oxides which are less soluble than the glass matrix (availability controlled by oxide solubilisation).

A verification has been performed for the third class comparing release and diffusion of uranium in clay at various contact times. The results are in good agreement with the values calculated by the use of the model.

Further confirmation is required to verify the behaviour in different media and temperatures and to evaluate release and diffusion of some actinides (Np, Am).

Studies were also performed to characterize cement as a matrix for the conditioning of medium level waste. The influence of chemical composition and different manufacturing procedures were investigated, by measuring density, pore size distribution, compressive strength and leachability.
The possibility of collecting relevant available information on the different conditioning matrices for HLW into a data bank is being investigated.

Interaction of radionuclides with geological media and environment

The objective of this experimental investigation can be summarized as follows:

a. to assess the physico-chemical forms of radionuclides leached from vitrified wastes.

b. to describe their migration towards the biosphere following the eventual release from the repository.

c. to study the environmental biogeochemistry of long-lived radionuclides.

The activity involves the development of laboratory simulation techniques for studying natural environmental conditions and interpretative modelling of the process identified.
Terrestrial and marine situations are being considered; the application of physico-chemical methods for investigating the radionuclide speciation constitutes also an important development of the experimental programme.
The output of this activity should provide input data needed for risk assessment models.

On the basis of results obtained by laboratory column experiments, a filtration model has been developed which describes the distribution pattern of actinides, taking into account the size distribution of their colloidal fraction in a freshwater aquifer.

A parallel activity has been started for the assessment of the potential migration of transuranic nuclides in brine saturated aquifers. First results obtained with Am leachate indicate a saturation effect of the retention capacity in a typical soil overlaying a salt dome. The equilibrium distribution of soluble Am species around a salt rock repository has been theoretically evaluated from the saturated zone to the typical concentration of surface fresh-waters passing through a dilution zone.

In this zone there is a continuous change in the equilibrium distribution of americium chloro- and carbonate-complexes governed by the lowering of chloride concentration and the increasing of pH and total CO_2 content. In the saturated zone the $AmCl^{2+}$ and $AmCl_2^+$ species are predominant while in the superficial aquifer the prevailing species are negative americium carbonate complexes.

During the year the coordinated action MIRAGE (MIgration of RAdionuclides in the GEosphere) has been started. This project conducted in the framework of the Community R&D actions on Radioactive Waste Management in jointly managed by the CEC-DG XII-Bruxelles and the JRC-Ispra.

Experimental activities concerning the sub-seabed disposal option have also been started; this work can be divided into two main elements; (i) integral experiments to simulate leaching and convection - diffusion transport processes in deep ocean sediments and (ii) studies of physico-chemical forms of leached radionuclides and corrosion products present in interstitial waters of sediments. In order to simulate as closely as possible real conditions three main parameters have been focused upon, temperature, redox conditions and pressure.

C. CHEMICAL SEPARATION OF ACTINIDES

The objective of this project is to assess the possibility of optimizing the back end of the fuel cycle from a waste management point of view. This may be done either by slight modifications of conventional practices or by addition of special waste treatment methods.

This objective is pursued both by theoretical evaluations and experimental tests, with special reference to radionuclides which give raise to long-term risks, such as the alpha-emitting nuclides (actinides).

Assessment studies on Alpha Waste Management

The study concerns alpha-waste streams produced both in reprocessing and fabrication facilities. Research effort has focussed on alternative treatment methods for alpha-contaminated waste which minimise the alpha waste arisings rendering the bulk of the waste «alpha-free». The conventionally applied treatment modes have been confirmed with the following optional processing modes:

— combination of the medium-low level waste (MLLW) fraction with the highly active waste (HAW) fraction.

— treatment of the MLLW fraction using the OXAL process scheme.

In the former processing mode the merging of alpha waste streams is seen to provide for a potential coherent approach in long term waste management. Provided progressive implementation of «salt-free» techniques and minimisation of off-standard (management) waste arising is applied such an option could become feasible even without increasing the final vitrified waste volume, taking advantage also of the fact that the vitrification process is highly flexible and adaptable for what concerns the range of acceptable chemical constituent compositions which can be incorporated. In applying the OXAL processing mode five options have been identified depending on whether plutonium is required to be recovered or not and whether special actinide conditioning is considered a waste management option or not. The cost of introducing an add-on OXAL process unit within a reprocessing facility has been evaluated to be less than 1% of the cost of a reprocessing facility.

In addition to the assessment studies consideration has been given to the approach of optimising waste management strategies through the possibility of modelling of waste management systems.

The possibility of setting up a data bank on waste management systems is also being investigated.

Experimental studies on actinide separation

Laboratory studies have been carried out at the JRC on actinide separation since 1973. Several chemical schemes for separating actinides from high-activity waste were developed and tested at a few-kilogram spent-fuel scale in collaboration with CEA, taking advantage of the Fontenay-aux-Roses chemical hot cells.

The extension of the developed chemical techniques to the isolation of alpha-emitters from several types of medium level waste has been studied in 1982, but the verification at a semi-industrial scale has been hindered by the lack of suitable hot cell equipment in Ispra.

The reconversion of the ADECO hot cell system approved in March 1983 by the Council of Ministers has made possible to reach a positive decision on the implementation of a multipurpose chemical facility (named PETRA), designed to provide representative quantities of the various waste streams of reprocessing plants, with possibilities of several chemical treatments on the waste itself, so that integrated strategies of alpha-waste reduction can be developed and verified at a semipilot scale. A conceptual design of the facility has been completed. The facility will be built by an italian-german consortium and is scheduled to be operational in 1987.

D. ACTINIDE MONITORING

Aim of this study is to set up methodologies and instrumentation for the non-destructive assay of plutonium content in different waste streams.

During '83, the revision of the guide «Monitoring of Pu-contaminated solid waste streams» was completed, by revising the chapters 4 and 5 (Passive and Active Neutron Assay).

In the area of passive neutron assay, the Time Correlation Analyser connected to a high- efficiency detector head was tested, in collaboration with AERE (Harwell, UK) and ENEA (Casaccia, Italy). A new version of the detector head was designed; two firms were selected for the construction of the TC analyser.

The computer software, developed for the interpretation of the data in terms of plutonium grams in the waste, was implemented at the DNPDE, Donnreay. The results of the non-destructive analysis were checked by comparison with destructive analyses on various waste types.

The JRC has also contributed to organize an interlaboratory comparison of non-destructive plutonium assay monitors, in the frame of the shared-cost action programme.

3. CONCLUSIONS

The activity of JRC in the frame of the programme «Safety of Nuclear Materials» is essentially addressed to find a satisfactory solution of a major issue of nuclear energy: the final elimination of the waste generated by the nuclear fuel cycle.

Two potential and complementary solutions are studied:

— the safe elimination of wastes by burying them in deep geological formations

— the optimization of the back-end of the fuel cycle from a waste management point of view.

In the first area of research the JRC is active both in the theoretical aspects of risk analysis and in the experimental validation of the models utilized for the evaluation.

The development of the risk assessment code LISA has been a major achievement in 1983. This code will be a basic tool for safety assessment also for the following years. The Community action on Performance Assessment of Geological Isolation Systems (PAGIS), jointly coordinated by JRC and by the shared-cost action programme provides a fruitful ground for interchange and discussion of the Community research in this area.

Actions such as MIRAGE (MIgration of RAdionuclides in the GEosphere), which has become fully operational in 1983, perform the same function for what concerns related experimental actions.

Although disposal in continental geological formations is considered as the main option for the final elimination of radioactive waste, the safety aspects of disposal of waste in deep oceanic sediments are also studied by the JRC, as an alternative option which is worth being explored at least on a scientific basis.

For what concerns studies on optimization of the back-end of the fuel cycle from a waste management point of view, the JRC is active both by theoretical evaluations of possible alternative routes for waste treatment and conditioning, and by related experimental research. Such research has been conducted in previous years essentially at laboratory scale. The decision of setting-up a facility (PETRA) by which such studies can be carried out at kilogram scale on fully active materials has been a major break-through towards a realistic assessment of alternative waste treatment.

It is expected that such a facility will also be of major help in the characterization of fully active conditioned waste, which is a prerequisite for its safe storage and disposal.

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