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Multiannual Programme
of the Joint Research Centre
1980-1983

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Annual Status Report

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SAFETY OF NUCLEAR MATERIALS

1982

Research Staff: 52
Budget: 6.025.000 ECU

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 solutions 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 risks linked to geological disposal of radioactive waste, carried out both by theoretical assessment studies and by experimental activities aimed at providing the necessary data base (risk evaluation).
— the study of possible modifications of the fuel cycle to keep hazardours 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 the barrier approach based on the evaluation of the barriers provided between disposed waste and man.

The JRC activity in this area includes:

— development of models for the assessment of the long-term safety of disposal in geological formations (continental and marine) and their application to specific sites.

— experimental work to evaluate the ability of the waste conditioning and of the retention of the radionuclides in the geological media and environment, to act as barriers against the radioactivity migration from waste repository to man.

In the field of model development a special attention has been given in 1982 to the conceptual problem of risk quantification. Waste disposal in geological formations is based upon the geologists' 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 segregated.

The JRC approach to assess the long-term risk associated to such an option is based on these points:

a. separate assessments of release probabilities and consequences for each scenario.

b. quantification of the risk as the product of probabilities and consequences.

c. description of the uncertain parameters with time-dependent probability distributions and study of the overall uncertainty associated with the risk estimate.

This approach enables the comparison with preestablished acceptability criteria (for instance, the natural background) both in terms of dose and of risk and the comparison with other risks, both natural and induced by the technological society.

Computer codes for the implementation of the methodology have been prepared and are continuously updated and ameliorated as long as new information becomes available.

The JRC methodology is being applied, in collaboration with CEN/SCK-Mol, to the Boom clay formation in Belgium which has been selected as an experimental site to study feasibility and safety of radioactive waste disposal in clay formations.

The probabilistic analysis of a possible failure of the Boom clay formation was carried out during the early part of the programme.
Release probabilities to three different receptors (groundwater, land surface and atmosphere) were estimated over time periods from 2,000 to 250,000 years. Faulting phenomena are the principal mechanisms having the potential to cause releases to groundwater, while direct releases to land surface may be linked to erosive processes due to glacial actions; in short term, different kinds of human actions may be important.

The overal failure probabilities appear to be low enough (about 10^{-7} per year) to offer a large margin of safety.

Since 1981 consequence analysis is being carried out for the most important release scenarios, such as faulting and glacial erosion. Only a few radionuclides were found to be a potential source of hazard for the faulting scenario (^{237}Np, ^{129}I, ^{99}Tc) but with radiation exposure to the most exposed individuals of the order of 10^{-3}—10^{-4} rem/year.

Research is proceeding in the analysis of the second release scenario (glacial erosion) and on the natural evolution of the repository in absence of any disruptive event.

The results obtained on the faulting scenario are also being reviewed on the basis of the most recent laboratory results on barriers behaviour, so that the uncertainty on results obtained by this second iteration will be substantially diminuished.

In the second half of 1981 the PAGIS (Performance Assessment of Geological Isolation Systems) action has been started in the framework of the Community Plan of Action in the Field of Radioactive Waste. The objective of this coordinated study, involving the participation of several national organizations under the cost-shared action programme and the JRC, is to obtain in about 6 years a harmorized evaluation of the main disposal options presently under study in the Community (salt, granite, clay, sub-seabed).

The initial part of this action (1982-1983) is aimed at providing a detailed data base for the risk analysis exercise. Reference sites have been selected for the four types of formations, and the necessary geological and hydrological data are being collected and reviewed. The study of those parts of risk analysis which are not site specific, such as waste reference data, some aspects of major disruptive geological events and biosphere modelling, will be carried out in common.

Coordination of the study is assured jointly by JRC and shared cost action. In addition, the JRC has assumed several tasks particularly on the preparation of detailed barrier modelling. A special effort is also being done by JRC to assure harmonization of the adopted risk analysis methodologies.

Since 1981 the JRC activity in the area of risk analysis and feasibility assessment of the sub-seabed disposal option has been considerably increased.

The Commission participates as an official member in the NEA International Seabed Working Group.

In view of a possible enlargement of the activity on seabed disposal, in the framework of the next multiannual JRC plan, programme proposals are under evaluation in various research areas.

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

In the area of leaching of borosilicate glasses, the major effort is being spent to study the leaching in normal repository conditions i.e. with the glass in contact with limited amount of water and the backfilling material.

The results of the experiments have indicated that leaching and transport of radionuclides are strictly related.

An analysis of experiments conducted in simulated geological disposal conditions (in plastic clays) has shown that the weight losses of the glass can be correlated with diffusion phenomena.

In particular weight losses data seem to be correctly described by calculating the diffusion of silica in the leaching solution, assuming that at the contact with the glass the SiO_2 content in the leachant reaches the saturation point.

The availability of radionuclides for migration in the surrounding environment depends on parameters which are specific for the different radioisotopes. 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).

Cs, Tc and Am have been taken as typical representatives of the three classes, and model validation experiments are under way.

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.

From the on-going study, two main processes have been identified which govern the transport in the geosphere: the retention of microcolloids, present in the glass leachate, by homogeneous deep bed filtration mechanisms, and the formation of soluble anionic species which migrate with minimum interaction at the velocity of the water vector.

The first process is characteristic of the behaviour of the transuranic nuclides in the short-term, since the geological media effectively retain the leached colloids.

The second process may be viewed as the controlling factor over the long-term. The formation of transuranic anionic species in fact becomes the rate determining step in the subsequent dispersion of these nuclides.

In the case of technetium on the contrary soluble anionic species are present immediately in the vitrified waste leachate under the simulated natural conditions adopted. However, it should be noted that a different situation may exist under more reducing systems such as that found in deep clay formations and certain types of sea sediments.

In order to verify this assumption, percolation experiments with technetium and neptunium are being performed in special glove-boxes under anoxic conditions.

In the framework of a collaboration with DG XII Radiation Protection Programme of the Commission, a study has been made to investigate the biogeochemical behaviour of plutonium and americium and the relations between chemical speciation and bioavailability.

Two points arise from these studies: firstly the changes in chemical speciations of plutonium in-situ (and hence its potential bioavailability) may be important. That transuranic chemical speciation is a dynamically active system must therefore be taken into account when estimating the environmental fate of these nuclides. The second point is the ability of living organisms to affect the speciation so that under certain circumstances feed-back mechanisms from biota may be of potential importance in controlling the fate of transuranics.

C. CHEMICAL SEPARATION OF ACTINIDES

The long-term risk of radioactive waste is largely due to alpha-emitters (plutonium, americium, neptunium). In the nuclear fuel cycle (mainly in reprocessing and Pu-fuel fabrication plants) a large variety of alpha-wastes are produced.

The JRC performed during the past years an important assessment concerning actinide separation from HLW and their transmutation in nuclear reactors (partitioning and transmutation). The assessment, based on laboratory experiments and theoretical evaluations, was essentially completed during 1980.

Verification experiments on the feasibility of chemical separation have been continued at a reduced scale throughout 1982, largely in cooperation with CEA-Fontanay-aux-Roses, in order to confirm at the scale of several kilograms of spent fuels the flow sheets developped by tracer experiments. A substantial confirmation of the flowsheet, has been obtained in the fully-active runs.

The assessment study on feasibility and cost-benefit of nuclear transmutation has clearly shown that the reduction of the long-

term risk of geological disposal, which is the incentive for partitioning and transmutation, is small, irrespective of the transmutation option considered. In addition, considering the large implications on the fuel cycle and the long time needed for its implementation, a reduction of the potential long-term risk of geological disposal appears to be more readily achievable by improving the techniques of alpha-waste reduction, conditioning and disposal.

Studies initiated in 1981 and completed in 1982 have shown that methods developped for high level waste partitioning may be adapted to medium level waste streams. In particular the OXAL flow sheet has been modified successfully to include the decontamination of both spent organic solvent and other medium level streams of reprocessing plants.

An extensive use of oxalic acid as reagent process for both high-level and medium level waste streams may give raise to waste management schemes based in separate disposal of α— and non—α streams which may be worth of further investigations.

In order to scale-up the chemical separation studies on medium level streams, the equipment of a series of hot cells of the ESSOR reactor (ADECO cells) was considered in 1981 and a detailed design of a multipurpose chemical facility (named PETRA) was carried out in 1982. The facility is 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 α—waste reduction can be developped and verified at a semipilot scale.

A final decision on the construction of the facility will be taken in 1983.

The assessment of the cost-benefit of alternative strategies for medium level waste disposal has been continued throughout 1982 and several schemes arising from the application of OXAL are being evaluated. The lack of sufficiently developped engineering data base for such assessment studies is limiting the progress of this action. Efforts are being done to improve and computerize the JRC waste management data base.

D. ACTINIDE MONITORING

The JRC is developing methodology and instrumentation for non-destructive assay of plutonium content in waste streams. The activities are performed within the frame of the EURATOM Advisory Laboratory.

Each of the envisaged five chapters of the Guide «Monitoring of Plutonium Contaminated Solid Waste Streams» has been published during the past years. Nevertheless each chapter is subject to continuous revision following the developments of methodologies for Pu-waste monitoring. The revision of the first two chapters (Planning of Monitoring Systems and Principles and Theory of Radiometric Assay) are completed, the third on passive gamma assay techniques is drafted and the chapters IV and V (Passive and Active Neutron Assay) are now updated to the latest developments. A new edition is planned for 1983.

The JRC assists in the implementation of a non-destructive assay system for monitoring of Pu-contaminated waste at the Dounreay Nuclear Power Development Establishment (DNPDE) in the framework of a collaboration between DNPDE and JRC. For the applied neutron and gamma techniques mathematical models were elaborated, nuclear data prepared and a software package is in preparation permitting an on-line interpretation of measurement data.
A seminar at the University of Lyon is in preparation to inform interested people from other bodies on the achieved results.

Considerable success has been achieved in the area of passive neutron assay. The time correlation analyser connected to a high efficiency detector head came into operation at the beginning of 1982. An assessment of the various systems for passive neutron assay has also been carried out. Four different types of time-correlation analysis were considered in the assessment. These are: the variable Dead-Time Counter; Shift Register; Pulse-To-Pulse and the Pulse Fluctuation Analysis.

A common interpretation model based on elementary probability theory and basic nuclear data is applied to all the various time correlation analysis. Measurements have been performed on JRC plutonium samples. The results show clear evidence for the advantages of the P.T.P. and P.F.A. measuring methods. The development of a hardware version of the time correlation analysis as plant instrument has been started.

The JRC assists in the implementation of a non-destructive assay system for plutonium waste monitoring in support of plant operators or control autorities. On the other hand for further development, the JRC is interested in pilot plants test measurements in various plants applying non-destructive radiometric techniques developed at Ispra. These tasks have been performed in collaboration with DNPDE-Dounreay and AERE-Harwell (Great Britain); ALKEM-Hanau (Germany); CNEN-Casaccia (Italy).

The JRC is also contributing in the organization of an interlaboratoy comparison of non-destructive assay monitors to be carried out in the framework of the share cost action programme.

3. CONCLUSIONS

The JRC programme Safety of Nuclear Materials is essentially dealing with long-term safety aspects of radioactive waste which are of major importance for the solution of waste management problems. Such aspects are intrinsically international and ideally tailored to a Community Institution such of the JRC, since they take advantage of the interdisciplinary nature of the JRC and of its indipendence of national constraints. The links established with national Institutions such as CEN/SCK Mol, DNPDE UK and CEA-Fontanay-aux-Roses as previously mentioned assure an easy flow of information with individual countries.

Actions such as PAGIS, to which the JRC gives an important contribution, are also means for orienting and discussing the various aspects of Community and national research, and for obtaining an optimized balance between the activity which the

Community executes in the JRC and that carried out in national laboratories by means of cost-shared actions.

The main action of the JRC in relation to the waste management deals with the evaluation of long-term risks or geologic disposal, which is considered as one of the major issues of nuclear energy.

In addition to the activity on risk evaluation of geologic disposal the JRC carries out activities which are related to fuel cycle optimization. They aim at indicating possible improvements in the many and complex operations of the fuel cycle, in order to obtain waste arisings which are optimized from a waste management point of view.

Actinide separation from waste and extensive recycling of the separated actinides is a waste management option on which the JRC has been active for almost a decade and has acquired expertise and reputation. While the cost-benefit of such operations on high level waste appears doubtful the application of chemical partitioning to other waste streams appears as a promising way to maintain the α-waste arising at a reduced level without excessive sophystication of the technology of the fuel cycle.

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