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COMMISSION OF THE EUROPEAN COMMUNITIES

Multiannual Programme of the Joint Research Centre 1980-1983

1981 Annual Status Report

Safety of nuclear materials

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SAFETY OF NUCLEAR MATERIALS

1981

Research Staff: 52
Budget: 6.025.000 ECU

Projects:

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

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Joint Research Centre
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1. INTRODUCTION

The safe and economic 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.

The motivations for a Community action in the field of radioactive waste management are 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 developed 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 the 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.

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 programme Safety of Nuclear Materials is essentially dealing with long-term safety aspects of the radioactive waste management. The long-term risk of the radioactive waste is due to the presence of alpha-emitting actinides (plutonium, americium, neptunium) and of some long-lived fission products.

The JRC staff allocated to this programme constitutes the most important single group of the Community working in the area of the long-term safety of radioactive waste.

The programme Safety of Nuclear Materials of the JRC has three main objectives:

— *Evaluation of long-term risk of radioactive waste disposal*
Development and testing of models for risk analysis of the geological disposal of radioactive waste and production of the experimental data required for model application.

— *Assessment studies on alpha-waste management*
Assessment by theoretical and experimental work of the feasibility and incentives of alternative management strategies for alpha-contaminated wastes, in view of an optimization in terms of cost and long-term risk.

— *Non-destructive assay of plutonium*
Development of methodology and instrumentation for the non-destructive assay of plutonium content in waste streams.

2. RESULTS

Evaluation of long-term risk of radioactive waste disposal

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.

Development and application of models

The JRC methodology is based on the following steps:

1. Identification of any barrier able to prevent, decrease or delay the radioactivity flow towards the biosphere.
2. Modelling of the barriers in such a way that either a probabilistic or a deterministic quantitative description of the behaviour of each barrier against radioactive escape can be drawn.
3. Data collection and model application, to assess containment failure probabilities and radioactivity doses to man.
4. Sensitivity analysis, to get information on the relative importance of the various parameters.

The barriers identified are the following:

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

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 1978-1979.
Release probabilities to three different receptors (groundwater, land surface and atmosphere) were estimated over four different time periods (2,000, 15,000, 100,000 and 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 overall failure probabilities appear to be low enough (about 10^{-7} per year) to offer a large margin of safety.

During 1980 and 1981 a corresponding analysis of the consequences has been developed on the basis of release scenarios selected among those indicated as the most important by the probabilistic analysis.
The release scenario based on the creation of a permeable fracture across the repository has been particularly analysed. Np-237 has been identified as the most important isotope in governing the long-term risk; this is due to the concurrence of its very long half-life together with its large mobility and large

dose factor. Other isotopes of concern appear to be I-129 and Tc-99, while plutonium isotopes are especially important for inhalation pathways.
To take into account the uncertainties in the input data a Monte Carlo method has been utilized to generate the probability distribution of the consequences, given the distribution of the input parameters.

Concerning maximum doses to an individual of an hypothetical critical group, assessed with very conservative assumptions, the results of the study indicate that they would be in the range of 10^{-3} -10^{-4} rem/year representing 10^{-2} to 10^{-3} of the natural background dose.

During 1981 the analysis of the consequences of a glacial erosion over the repository region has been started. Preliminary dose assessments seem to indicate that the consequences of this scenario could be of some importance and therefore require to be carefully weighed against the corresponding probabilities, in order to have a comprehensive view of the risk.

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, is to obtain in about 6 years a harmonized evaluation of the main disposal options presently under study in the Community (salt, granite, clay, sub-seabed). The activity of the J.R.C. deals particularly with the clay and sub-seabed disposal options.
Coordination of the study is assured jointly by J.R.C. and shared cost action.

During 1981 the J.R.C. activity in the area of risk analysis and feasibility assessment of the sub-seabed disposal option has been considerably increased.
It is worth mentioning that from February 1981 the Commission participates as an official member in the NEA International Seabed Working Group. This involves the participation of J.R.C. and shared cost action staff in various Task Groups.
The J.R.C. staff contributes particularly to the activity of the System Analysis Task Group.
In view of a possible enlargement of the activity on seabed disposal, in the framework of the next multiannual J.R.C. plan, programme proposals are under evaluation in various research areas; in particular the role and the characteristics of high pressure facilities are being evaluated.

Long-term stability of conditioned waste

The J.R.C. activity in this area is essentially directed to study radiation damage effects and leaching phenomena in borosilicate glasses.

During 1981 studies have been carried out, using a HV electron microscope, on electron irradiated borosilicate glasses. It has been confirmed that the observed phenomena of bubble formation start at dose rates much higher than those expected during the storage of high level wastes.
Other information on the basic aspects of radiation damage is being obtained by means of irradiation experiments on silica.
During 1981 a critical review of all the results, reported in the literature, concerning radiation damage in borosilicate glasses, has been completed and published. The conclusions of this review are quite reassuring; in fact it is stated that the existing values of the examined properties indicate that the effect of radiation will not greatly influence the stability of the glass at the doses and dose rates expected.

During 1981, in the area of leaching of borosilicate glasses, the major effort has been 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.
Thus it is envisaged a deeper integration between the studies of leaching and radionuclide migration.

The J.R.C. intends also to contribute to a better knowledge of the basic mechanisms of leaching which is required for a correct evaluation of the long-term stability of borosilicate glasses for various repository conditions. In this connection an experimental facility is being set-up for the measurement of the relevant surface quantities which determine the mechanisms of leaching.

Interaction of radionuclides with geological media and environment.

The J.R.C. activity in this area has been mainly directed to investigate the migration behaviour of the radionuclides following the leaching of vitrified waste in a repository. Thus the leachate of the borosilicate glasses, containing the radionuclides to be studied, has been used in the laboratory experiments.

The tests carried out at the J.R.C. in the past years have indicated that the presence in the leachate of colloidal forms, which can be subjected to filtration in the soil layers, has an important role in governing the radionuclide migration in the short term; the tests have also indicated that the formation of carbonate complexes of actinides, which are not retained by geological media, can favour their migration and consequently increase their potential risk.

Thus the J.R.C. experiments during 1981 have been mainly directed to study formation and behaviour of colloidal forms and carbonate complexes. The work initially limited to actinides has now been extended to investigate the behaviour of technetium.

The other important line of activity concerns the relations between chemical forms and bioavailability, this aspect being investigated in the framework of collaborations with laboratories working in the shared cost action programme Radiation Protection.

During 1981 the work on colloidal forms included laboratory experiments on the retention and subsequent release of radionuclides in columns of various media, characterization of colloids and development of a model for deep bed filtration.

The laboratory experiments were carried out in such a way to simulate the conditions existing in the porous media overlaying the Boom clay formation.

In the area of carbonate complexes the work concerning compositions and stability constants of Am complexes has been completed; on the basis of the results obtained it is possible to evaluate the relative importance of the various soluble speçies of Am in groundwater.

It is worth mentioning that the possibility to increase the effort in the area of the interaction of the radionuclides with geological media and environment, is being envisaged, in order to apply the J.R.C. methodologies to various geological formations and to increase the contribution to the PAGIS action.

Assessment studies on alpha-waste management

The long-term risk of radioactive waste is mainly 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 most important waste stream is the high level waste (HLW), from the first extraction cycle of the reprocessing plants, which contains most of the fission products and by-product actinides (americium, curium, neptunium) present in the spent fuels and a small fraction of plutonium and uranium.

Other important alpha-waste streams are:

— residues from fuel dissolution

— cladding hulls

— various liquid and solid wastes produced in the operation of reprocessing and Pu-fuel fabrication plants.

They are included in the broad category of the medium level wastes (MLW).

The J.R.C. 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.

The assessment has shown that the reduction in 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 of partitioning and transmutation 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.

On the basis of these considerations the J.R.C. effort was progressively reoriented towards an assessment of the procedures of alpha-waste management aiming at their optimization, taking into account various parameters and in particular cost and long-term safety.

As first step of the study a Baseline Waste Management Strategy (BWMS) has been defined. The introduction of alternative options in this reference strategy will be evaluated in terms of cost/benefit.

During 1981 the option based on the application of the OXAL process both to HLW and different categories of MLW has been evaluated.

The OXAL process had been experimentally investigated in the previous years in connection with the actinide separation from HLW. During 1981 the experimental work at the J.R.C. was extended to consider the application of the OXAL process to the separation of plutonium from MLW streams.

OXAL can be applied for the recovery of actinides from all process liquid waste streams which result from the reprocessing and fabrication of nuclear fuels and it is potentially extendable to solid waste arisings, leading to a practical process scheme for a fully integrated alpha waste management strategy.

The process scheme is highly compatible with the present Purex technology and provides considerable advantages concerning:

— reduction of the amount of alpha wastes with a consequent reduction of the associated hazard.

— more flexibility in the utilization of the Purex process.

The proposed process scheme is presently subjected to an extensive engineering evaluation.

The experimental activity on the OXAL process, at the laboratory level, has been essentially completed during 1981. In order to make possible the verification of the OXAL process on a larger scale, the equipment of concrete cells, available in the ADECO laboratory, has been considered. The safety aspects have been discussed with the licencing authorities and cost evaluations have been carried out.

A decision on the operation will be taken during 1982. In the case of a negative decision it is planned to reduce the experimental activity on alpha waste management and to concentrate the J.R.C. effort on the assessment studies. This would make possible an equivalent increase of experimental

effort in the area of interaction of radionuclides with geological media and environment.

Non-destructive assay of plutonium

The J.R.C. is developing methodology and instrumentation for non-destructive assay of plutonium content in waste streams. Methods based on neutron and gamma measurements are applied.

The methodology proposed by the J.R.C. for non-destructive assay of plutonium has been published during the past years in the form of guides which constitute an important contribution to the rationalization of the waste monitoring problems. During 1981 the process of revision of these guides has been continued, taking into account recent results, and the publication of a revised version is planned for the end of 1982.
On the other hand the J.R.C. methodology is being applied and tested in the evaluation of the waste monitoring system of the Dounreay Nuclear Power Development Establishment (DNPDE) in the framework of a collaboration between DNPDE and J.R.C.

The Dounreay waste monitoring system includes monitors based on passive and active neutron assay and gamma assay. DNPDE provides the operational data and J.R.C. provides the mathematical modelling and interpretation.
During 1981 Part 1: Interpretation Models and Calibration Methods, of the final report of the experiment has been prepared. It is planned to prepare the other parts of the final report during 1982.

In the area of instrument development the effort has been directed during 1981 towards the setting-up of a new detection system for passive neutron assay.
The system based on the pulse to pulse time correlation analysis may constitute a major improvement in comparison with the systems presently applied.
During 1981 a high efficiency detection head has been set-up and the computerization of the system has been essentially completed.
The system is now being subjected to a series of tests both at Ispra and in nuclear plants in order to evaluate the effective possibilities of application in different working conditions.

The J.R.C. is also contributing to the organization of an interlaboratory comparison on the monitoring of drums containing Pu-contaminated wastes. This exercise, organized in the framework of the shared cost action programme, groups the J.R.C. Ispra and other 5 Community laboratories.
The scientific secretariat of the overall exercise has been assigned to the J.R.C. Ispra.

3. CONCLUSIONS

The J.R.C. 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.

The J.R.C. is providing an important contribution to the Community effort in this area.
In fact a multidisciplinary group of about 50 people is engaged in studies connected with long-term safety; the J.R.C. activity takes adavantage from the very strict connections existing between theoretical evaluations and experimental work.

Strict relations have been established between the J.R.C. and the shared cost action programmes Management and Storage of Radioactive Waste and Radiation Protection.
An important example of effort integration between J.R.C. and national laboratories is offered by the PAGIS action, started in 1981, with the objective of developing an harmonized evaluation of the main disposal options presently under study in the Community.

The PAGIS action will constitute a point of reference for the J.R.C. activities on risk evaluation both in the theoretical and experimental area.
Specific collaborations have been established with national laboratories, which make possible the verification of the J.R.C. results and their transfer to Community organizations.
In this connection it is worth mentioning in particular the collaboration with CEN/SCK, Mol for the application of J.R.C. risk analysis methodology on the Boom clay formation and the collaboration with DNPDE, Dounreay for the evaluation of the waste monitoring system.

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