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Multiannual Programme of the Joint Research Centre 1980-1983

1980 Annual Status Report

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SAFETY OF NUCLEAR MATERIALS

1980

Research Staff: 52
Budget: 5,235,000 ECU

Projects:
- Risk evaluation
- Protective barriers
- Actinide separation
- Actinide monitoring

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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 by means of a direct action programme developed at the Joint Research Centre (Programme Safety of Nuclear Materials) and by means of an indirect action programme.

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 geological disposal 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 modeling relies upon mathematical treatment of the retention phenomena which accompany the solution migration through soil columns.
4. Environmental mobility and biological availability of radioelements; this barrier is particularly effective for most of the long-lived alpha-emitters; it is modeled in 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, 25,000, 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 a corresponding preliminary exercise of consequence analysis has been developed on the basis of two release scenarios selected among those indicated as the most important by the probabilistic analysis.

Preliminary results of the study seem to indicate Np-237 to be the risk-governing isotope in the long term. Important contributions are also due to I-129, Tc-99, Pu-239, Pu-240 and Am-243.

The existence of small mobile fractions of actinide elements, which appear to be capable of migrating without retention in the subsoil, can become the risk-governing factor. However, the extent of these fractions seems to depend on various factors, as water composition and velocity, so that the figures given (0.1% for plutonium and americium, and 2.5% for neptunium) must be considered only as orientative, and require to be utilized with some caution.

Preliminary results indicate, however, that maximum doses to individuals, assessed with very conservative assumptions, would be in the range 10^{-3} - 10^{-4} rem/year.
This would be quite acceptable, representing 10^{-2} - 10^{-3} of the natural background dose.

In order to improve contacts between the risk analysis activity of the JRC and similar activities which are carried out by national laboratories in the framework of the indirect action programme a Risk Analysis Subgroup was formed. The group meets periodically to exchange ideas and experience, to discuss risk analysis approaches and to promote supporting experimental research. The JRC keeps the secretariat of the group.

During 1980 a considerable reinforcement of indirect action activities on safety analysis has been devised, so that a joint effort JRC-D.G.XII is being developed to obtain a coordinated set of studies at Community level, with the objective of developing risk analysis models for the various types of geological formations.

During 1980 the JRC has increased its participation in the NEA International Seabed Working Group which is studying the option of disposal of high level waste (HLW) into the deep ocean seabed.

The JRC represents the Commission in the Working Group.

Long-term stability of conditioned waste

The JRC activity in this area has been essentially directed to investigate radiation damage effects and leaching phenomena in vitrified wastes.

The radiation damage studies are being conducted by means of tests on borosilicate glasses and more fundamental studies on silica.

From the results obtained at the JRC it appears that radiation damage cannot deteriorate the stability of the borosilicate glasses in a significant way.

Concerning leaching of vitrified waste, the experiments conducted in presence of a large excess of water, typical of accidental repository conditions, have been completed during 1980.
The results obtained at the JRC indicate that the temperature is the leaching governing factor; in fact the temperature influences not only the initial value of the leaching rate but also the ability of the surface layer to act as a diffusion barrier.

During 1980 a series of tests has been initiated to investigate the leaching behaviour of the glasses in conditions similar to those existing in a repository. In normal repository conditions the amount of water in contact with the glass is very limited; in addition, if a sufficient time is passed so that the container is completely dissolved, the glass is in contact with the backfilling material and the corrosion products of the container.

It is widely recognized the need for a better knowledge of the basic mechanisms of leaching to make possible correct evaluations of the long-term stability of borosilicate glasses in varying repository conditions. To this purpose a new activity has been started during 1980 aimed at the characterization of the glass surfaces in connection with the leaching process.

Interaction of radionuclides with geological media and environment

Objective of the JRC activity is to contribute to a better understanding of the interaction of radionuclides with geological media and environment, following an eventual release from the waste repository. The retention of radionuclides in the geological media and the environmental dilution and segregation processes can constitute valid barriers against the transfer of radionuclides from waste repository to man.

Experimental studies are in progress to investigate the migration behaviour of actinides and Tc-99 leached from a glass matrix, under conditions similar to those of natural groundwater.
It is assumed that, as a consequence of a postulated accidental event, water penetrates the repository and partially dissolves the glass containing actinides and technetium, moving afterwards through a surrounding porous formation.

In order to simulate in the laboratory the expected conditions of glass leaching and underground transport, a water pathway is established which flows over the glass and then through soil columns (glauconite sand sampled from the Antwerp sands overlaying the Boom clay formation).

These migration experiments have been complemented by various tests aimed at the identification of the chemical forms of the radionuclides in the leaching solutions.

The results obtained at the JRC indicate that the reversible ion exchange mechanisms, normally utilized in the risk evaluation studies for modeling radionuclide migration, do not give a correct description of the phenomena. In fact, on the basis of the JRC results, filtration of microcolloids appears as the migration governing mechanism.

The JRC experiments have also indicated the existence of a small fraction of the actinide elements which seems to be able to migrate without retention. The actinide mobile species have been interpreted as complex carbonate ions. The determination of the stability constants of the various complexes formed is under way.

The JRC studies have indicated that also the behaviour of the radionuclides in the environment is strongly dependent on their physico-chemical form. The JRC activity in this area is strictly connected with the studies carried out in the framework of the indirect action programme Radiation Protection.

In this area a jointly sponsored CEC/IAEA Technical Meeting on the "Behaviour of transuranics in the aquatic environment and sediment water interfaces - Techniques for identifying chemical speciation" was held at the JRC Ispra on March 1980.

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 (BPAs) (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).

During 1980 the assessment studies on the chemical separation of actinides from HLW and their transmutation in nuclear reactors (partitioning and transmutation) has been essentially concluded and the JRC effort has been progressively transferred on the assessment of management procedures for MLW.

Assessment studies on partitioning and transmutation of actinides

In the Community the favoured solution for the management of HLW is the conditioning in vitrified form and the disposal in geological formations.
However, the JRC was charged to assess the feasibility and incentives of the advanced waste management option based on the chemical separation of the actinides from HLW and their transmutation in nuclear reactors. In fact this strategy had been proposed as a possible solution of the problems of the long-term risk of geological disposal.
The work at Community level in this area was progressively concentrated at the JRC - Ispra.

The activity started at the JRC Ispra in 1973 and was developed during the plans 1973-1976 and 1977-1979. A total of about 100 men.year was employed in this activity. Conclusions of the JRC assessment were drawn during 1980.

The JRC activity has been developed following two main lines:

— Laboratory studies and engineering assessment of chemical flow-sheets for the actinide separation from HLW. Three processes have been studied based on solvent extraction (HDEHP and TBP processes) and oxalate precipitation (OXAL process).

— Assessment of the nuclear transmutation of the actinides based on reactor physics calculations, conceptual design of fuel elements, evaluations of cost, risk and impacts on the nuclear fuel cycle.

Some aspects of the overall assessment have been studied by national laboratories under JRC contract. It is worth mentioning the reactor physics calculations for FBRs and HTRs carried out by CNEN, Casaccia and KFA, Juelich respectively, the verification on a larger scale by CEA, Fontenay-aux-Roses of the flow-sheets developed by the JRC and the assessment by AGIP Nuclear, Milan on the use of sol-gel processes for the fabrication of actinide bearing fuels.

The conclusions drawn by the JRC on the basis of the various studies can be summarized as follows:

— Partitioning and transmutation are feasible in the sense that a solution to overcome the technological problems may probably be found if a very large effort of research, development and demonstration is provided during a few decades.

— While reactor problems do not appear particularly severe, the reprocessing and fabrication processes will require a large effort of adaption and development due to the technological and safety problems related to the high neutron emission and heat release and the very small critical masses of some BPAs.

— In the HLW partitioning the radiation dose commitments to the solvent are far beyond those considered as today acceptable.
Severe problems arise in the fuel element fabrication process for heterogeneous recycling due to the high neutron emission and decay heat release. These problems are less severe for homogeneous recycling.
Also the problems connected with the fuel behaviour in the reactors appear less important in the case of an homogeneous recycling.

— Among the various strategies considered for transmutation, the homogeneous recycling in FBRs appears to be the best choice both for transmutation efficiency and for compatibility with the FBR fuel cycle facilities anticipating the availability of an advanced fuel cycle technology both for reprocessing and MOX fabrication (remotely operated shielded facilities).

— The relative increase of the fuel cycle cost is limited. The increase of short-term risk due to routine radiation exposure can probably be kept at low levels; the increase of the accidental risk is difficult to be evaluated.

— The reduction in the long-term risk of geological disposal which is the incentive for partitioning and transmutation appears to be small, irrespective of the transmutation option considered.
In fact the reduction of the long-term risk is limited by the accumulation of the fuel and by-product actinides losses and by the presence of long-lived fission products.

— 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.

In the framework of the activity of OECD Nuclear Energy Agency, in the field of radioactive waste, the Commission has been chosen as leading organization for the studies on partitioning and transmutation of actinides.

Thus the JRC has organized at Ispra in March 1977 the First Technical Meeting on the Nuclear Transmutation of Actinides and in April 1980 the Second Technical Meeting on the Nuclear Transmutation of Actinides. These were the most important international meetings organized on this subject.

In particular during the 1980 meeting the results of the JRC assessment and of a similar assessment carried out in the U.S.A. were presented; the conclusions of the meeting were similar to the above reported JRC conclusions.

The results of the JRC assessment will be assembled in a final summary report during 1981.

Assessment studies on medium level waste (MLW) management

The medium level waste (MLW) category is composed of a large variety of materials with heterogeneous chemical and physical characteristics and considerable volume arisings.

Aim of the JRC assessment is to contribute to the optimization of the MLW management procedures by means of theoretical evaluations and experimental activities. In the optimization mainly cost and long-term risk have to be taken into consideration. Thus the activity is being carried out in a strict relation with the risk analysis studies.

The activity in this area has been started during 1980.

The setting-up of a baseline waste management strategy (BWMS) has been initiated; the introduction of alternative options in the BWMS will be evaluated in terms of cost/benefit in view of the selection of the most convenient approach.
The laboratory work has been directed to evaluate alternative management procedures to be applied to liquid MLW.

Current management schemes for this type of wastes, rely on volume reduction and for most cases on the use of non-specific low-cost matrices (cement, bitumen) for immobilization.
Limits may however be put to the volume reduction factor by the salt content of some effluents while the low leaching resistance of cement and bitumen matrices may impose a heavy burden on repository selection for a safe long-term isolation.

The JRC approach, which is being experimentally tested, considers the introduction of simple modifications in the processes with a view:

— to obtain the bulk of the waste materials with a low alpha content (working hypothesis 100 nCi/g)
— to have the alpha-emitters in a small fraction which may be subjected to specific conditioning or to recycling.

At present the laboratory work concerning liquid MLW is directed to evaluate the feasibility of plutonium separation utilizing oxalate precipitation processes, which were previously set-up for actinide separation from HLW.

In order to make possible the verification on a larger scale of the management schemes, developed by the JRC, the equiping of concrete cells, available in the ADECO laboratory, is being considered.
During 1980 a preliminary safety report for the utilization of the cells, has been prepared. A decision on the operation will be taken at the middle of 1981 on the basis of an assessment of benefits and costs.

Non-destructive assay of plutonium

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

The JRC has published guides for plutonium monitoring which constitute an important contribution to the rationalization of the waste monitoring problems.
These guides are kept under continuous revision.

The methodology proposed by the JRC is being applied for the evaluation of the waste monitoring system of the Dounreay Nuclear Power Development Establishment (DNPDE) in the framework of a collaboration between DNPDE and JRC.

The Dounreay waste monitoring system includes monitors based on passive and active neutron assay and gamma assay. DNPDE provides the operational data and JRC provides the mathematical modelling and interpretation. During 1980 the work has particularly progressed in the evaluation of the monitors based on passive neutron assay.

In the area of passive neutron assay considerable development work is in progress at the JRC.
In particular it is worth mentioning the work on the pulse to pulse time correlation analysis, which may constitute a major improvement in comparison with the systems like the Variable Dead Time Counter (VDC) and Shift Register, presently in use.
These systems have been introduced because of their electronic simplicity and ease of implementation rather than the fact that they represent a good algorithm for analysis. The advent of microprocessors means that a more sophisticated algorithm can be considered as suitable for operational implementation.

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

3. CONCLUSIONS

The JRC programme Safety of Nuclear Materials is essentially dealing with long-term safety aspects of the radioactive waste which are of major importance for the solution of the waste management problem.

The JRC is giving 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 and the connection between assessment activities and experimental work are well established.

Strict relations have been established between the JRC and the indirect action programmes Management and Storage of Radioactive Waste and Radiation Protection.
In particular a joint effort JRC-DG XII is being developed to obtain a coordinated set of studies at Community level, with the objective of developing risk analysis models for the various types of geological formations.

Specific collaborations have been established with national laboratories, which make possible the verification of the JRC results and their transfer to Community organizations.

An example of the JRC contribution to the Community effort is the application of the JRC risk analysis methodology on the Boom clay formation. This is the first complete risk analysis exercise, including probabilistic and deterministic assessment, which is carried out on a specific site in the Community.

Another interesting example is the application of the JRC methodology, for non-destructive assay of plutonium, in the setting-up of the waste monitoring system of the Dounreay Nuclear Power Development Establishment.

In some cases the JRC has been charged to perform assessments of general interest in the Community: this was the

case of the assessment on actinide partitioning and transmutation.
The JRC has worked for several years in this area and the conclusions were drawn during 1980.
The JRC has given an important contribution in defining an internationally agreed evaluation of feasibility and incentives of this advanced management strategy; in fact the JRC has contributed both with a large number of experimental and theoretical studies and with the organization of international meetings.

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