RESEARCH ACTIVITIES OF THE EUROPEAN COMMISSION ON NEW FUEL CYCLE CONCEPTS

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ABSTRACT

This paper presents the objective and the research work of the eight projects partly funded by the European Commission in the field of new fuel cycle concepts, which includes three areas of work: (i) strategy studies, (ii) partitioning techniques, (iii) transmutation techniques. This field is part of the specific programme on Nuclear Fission Safety of the Framework Programme for the European Atomic Energy Community (1994 -1998).

INTRODUCTION

In its fourth five-year shared-cost research and development programme on "Management and Storage of Radioactive Waste 1990-1994", the European Commission (EC) included the study of the potentialities of partitioning and transmutation (P&T) of long-lived radionuclides. The activities partly supported by the EC were dealing with experimental work on partitioning of long-lived radionuclides from high level waste (HLW) and an overall strategy study on the potentialities of P&T for nuclear waste management. Besides, research work on P&T, mainly on partitioning experiments and fuel and target development, was carried out at the Joint Research Centre of the European Commission in Karlsruhe, the Institute for Transuranium Elements (ITU).

The main results and conclusions of the strategy study have been presented elsewhere [1]. This study has led to an order of priorities for the radionuclides to be recycled. Plutonium should have the first priority because of its very large radiotoxicity and of proliferation risks. At present, the main issue is to improve the burning rate of plutonium in reactors to decrease its stock. Americium is the most radiotoxic actinide after plutonium up to 50,000 years and also contributes to the potential radiotoxicity of HLW through neptunium formation beyond 50,000 years. The problem to be solved is to find a selective partitioning process for americium only from the liquid HLW coming from PUREX reprocessing, which does not generate unacceptable amounts of secondary waste. When removing all plutonium and americium from HLW, the resulting waste will become less radiotoxic than uranium ore after 1000 years. Fast reactors appear at present to offer the best prospects for plutonium and americium incineration, when compared to light and heavy water reactors and high temperature reactors.

Nuclear Fission Safety is one of the specific programmes of the Framework Programme for the European Atomic Energy Community (1994-1998). It has been launched by the European Union on 15 December 1994. Exploring new fuel cycle concepts is part of this programme with three research tasks: (i) strategy studies, (ii) partitioning techniques, (iii) transmutation techniques. Eight research proposals to be partly funded by the EC have been selected in 1995 by the Commission on the basis of an evaluation made by independent experts. The objective and the research work of these projects are briefly described in this paper.

STRATEGY STUDIES

Four projects cover the strategy studies.

Evaluation of Possible P&T Strategies

The first project is a global assessment of different possible P&T scenarios and of the technical feasibility of P&T techniques and of advanced fuel and target fabrication. Eleven European laboratories are participating to this study, which is coordinated by CEA Cadarache (F). Five scenarios have been defined. The first one is the reference scenario with one-through cycle for pressurised water reactors (PWRs) with uranium oxide (UO_2) fuel. The four other scenarios have the same common basis from 2000 to 2040: the reactor park consists of PWRs with UO_2 fuel and PWRs with a mixture of 70% of UO_2 fuel and 30% of mixed oxide (MOX) fuel; plutonium (Pu) is recycled once, i.e. spent UO_2 fuel is reprocessed to recover Pu for MOX fuel fabrication and spent MOX fuel is placed in interim storage. From 2040, spent MOX fuel is also reprocessed following the requirements in Pu, minor actinides are partitioned and recycled in homogeneous mode for neptunium (Np) and heterogeneous mode for americium (Am) and curium (Cm). A PWR loaded with 30% MOX fuel is replaced every year by an over-moderated PWR with 100% MOX fuel in the second scenario and by a CAPRA type fast reactor (FR) incinerating Pu in the third scenario. The fourth scenario starts like the third one until a shutdown of nuclear energy is decided: special actinide burners are then built. The beginning of the fifth scenario is similar to the third one; at a certain time, fast breeder reactors are coupled to the network with the objective of having only FRs as burners and breeders in the reactor park.

The consequences of the use of fuels with high Pu content in PWRs and FRs on partitioning will be assessed: the dissolution of spent MOX fuel in nitric acid becomes more difficult, as the content in PuO_2 increases. Reprocessing of MOX with 45% PuO_2 and uranium free fuel and the effect of inert matrices will be studied. Both enhanced "PUREX based" and/or pyrometallurgical processes will be investigated.

Transmutation in PWRs and FRs will be assessed by computing actinide consumption and core safety parameters for advanced MOX fuels (highly enriched in Pu and/or containing Np) and/or specific targets containing americium and/or curium. Pu consumption will be optimised, while keeping the core safety parameters at a reasonable level. The moderator-to-fuel ratio of full MOX fuel cores in PWRs will be increased to maximise the Pu consumption. "U-free" PWRs and FRs will be studied.

The fabrication processes of the advanced fuel and target will be investigated: sol-gel fabrication and MOX fuels adapted to enhanced moderation and full core loading. The consequences of the presence of more radioactive nuclides in the fuel on the dose rates received by the workers during the different steps of fuel fabrication and handling will be evaluated.

Finally, the long term risk and residual dose to man from different waste to be disposed of in an underground repository will be compared: spent PWR UO_2 fuel at high burn-up, spent PWR MOX fuel, vitrified HLW, actinide depleted vitrified HLW, separated actinides conditioned in advanced matrices, spent FR MOX fuel after multiple recycling. The retardation efficiency of three geological barriers (clay, hard rock, salt) will be investigated.

Supporting Nuclear Data for Advanced MOX Fuels

Belgonucléaire (B) coordinates this project, which involves six European research institutions altogether. The objective of this project is to provide the strategy study above with more accurate nuclear data for the scenarios aiming at reducing the waste toxicity in MOX recycling schemes either in PWRs or FRs.

The accuracy of strategy studies involving the use of PWRs with enhanced moderation and full MOX loading will be assessed. The experimental basis is provided by the analysis of the composition of MOX fuel after irradiation at very high burn-up (80 GWd/t) in the overmoderated BR3 reactor. Experimental data are also available for MOX fuel irradiated in the Saint Laurent B1 PWR. These data will be compared with the results of calculations using the European JEF 2.2 library. Sensitivity and uncertainty analyses will be performed on the basis of the JEF 2.2 library and the EAF transmutation and uncertainty file.

A similar exercise will be performed for FRs. The experimental data are provided by the measurements of the isotopic composition of samples irradiated in Phenix and KNK-II in Karlsruhe. These data will be interpreted with the European JEF 2.2 library.

A critical analysis and intercomparison of the nuclear data files for the isotopes ^{240}Pu, ^{242}Pu and ^{241}Am will be performed especially in the range of resonances.

The corrected nuclear data will be introduced into working libraries for transport and radionuclide inventory computations. The computational techniques of the different partners will be compared to integrate all results in a common data base. Finally, the uncertainty margins of the isotopic compositions will be estimated for the P&T strategy studies.

Thorium Cycles as Nuclear Waste Management Option

The third project is coordinated by ECN Petten (NL) and involves six other European laboratories. The objective is an assessment of the thorium (Th) fuel cycle to limit nuclear waste production and to burn waste. This study will cover the major aspects of the thorium fuel cycle, i.e. mining, fuel fabrication, reactor operation, reprocessing, waste disposal and non proliferation.

The work on mining consists of a study of the quantities of tailings for available thorium minerals, the extraction performances and the composition of radionuclides in the extraction waste. The health impact on workers, the potential radiotoxicity of the generated waste and possibly the residual short and long term risks to the public will be also calculated.

The different types of thorium oxide fuels will be reviewed with special emphasis on the radioactivity of fresh and recycled fuels. Mixed oxide fuels will be considered with uranium, protactinium (Pa) and plutonium.

Three types of thorium fuelled reactors will be assessed: a PWR core with minimum actinide waste production; a PWR to burn as much as possible plutonium and minor actinides (MA) (Np, Am, Cm); a FR as an energy generating system with low actinide production or with high capability to burn Pu and MA. Limits imposed by reactor safety parameters, neutron economy and maximum achievable burn-up will be taken into account.

Concerning reprocessing, the THOREX process will be reviewed on the basis of previous experience with special attention to partitioning of Th, U, Pu and Pa.

The long term residual risk of geological disposal of the different HLW generated by the three types of reactors considered above will be computed. The basic migration parameters of Pa and Th will be measured in order to assess their behaviour in two generic geological environments (clay and granite).

The technical proliferation characteristics of Th and U based fuel cycles will be compared. The basic problems (neutron emission and heating rates) associated with the fissile ^{233}U and ^{239}Pu isotopes will be addressed.

Impact of the Accelerator-based Technologies on Nuclear Fission Safety (IABAT)

Kungliga Tekniska Högskolan (S) is coordinating this research contract, which involves a total of eleven European research institutions. The possibilities of Accelerator Driven Systems (ADS) for safe energy production, minimum waste production and nuclear waste transmutation will be assessed in this project. The objectives of the IABAT project are to perform system studies on accelerator driven hybrid systems, to assess accelerator technology, to obtain basic data on nuclear reaction cross-sections and on radiation damages at the spallation target walls and to study the radiotoxicity of the ADS fuel cycles.

The system studies on ADS will address several items: a safety and economical assessment; the physics of molten salt systems with fast and thermal neutron spectrum; the stability, time response and dependence of dynamics on subcriticality; a 2D/3D neutron kinetics study; an actinide incineration system with liquid lead as an actinide carrier; spallation target optimisation with respect to transmutation efficiency; estimation of the spallation product yields and of the radiotoxicity of the spallation target.

The technological aspects of accelerator technology related to construction, proton current and energy levels will be assessed. A cost analysis will be performed for linear and circular accelerators with an energy of e.g. 1GeV and a current of 10 mA.

As the high energy transport codes are not very reliable in the 100 MeV energy region, basic nuclear data are necessary. Evaluated nuclear data files will be assembled for protons on lead and isotopes for energies up to 200 MeV. Some cross sections above 20 MeV will be measured and validated for neutron and proton induced reactions (e.g. fission of Pb, Bi and ^{238}U and transmutation of some nuclides). The measurement of yields for ^{233}U thermal fission and ^{232}Th fast fission will be performed. The radiation damages at the lead spallation target enclosure walls will be estimated.

Different aspects of possible fuel cycles for ADS will be addressed. The radiotoxicity of the residual waste and the non-proliferation resistance of the reprocessing technology will be investigated for a LWR waste transmutation system based on molten salt fuel and thermal neutron spectrum. Different thorium fuel options will be considered for an ADS with liquid lead coolant/carrier. The radiotoxicity of the thorium based ADS fuel cycle will be evaluated for thermal, epithermal and fast systems.

PARTITIONING TECHNIQUES

Experimental work on partitioning is carried out in two projects.

New Partitioning Techniques (NEWPART)

Seven European laboratories are participating to this project, which is coordinated by CEA Marcoule (F). The objective of this project is to develop processes for the separation of minor actinides from very acidic aqueous solutions containing HLW without generating secondary solid waste, while avoiding the problems encountered previously (e.g. radionuclide precipitation and difficulties in the back-extraction of actinides from the solvent).

To meet this objective, two principles have to be applied: (i) the molecules (extractants, diluents, aqueous soluble reagents) used in the processes must be totally incinerable, once they have been spent; in other words, they are converted into gases, which can be released into the atmosphere; (ii) the very acidic liquid HLW to be processed must not be neutralised.

There are two possible routes for the separation strategy:

- route N° 1 has two extraction cycles, the first one to co-extract the actinides and the lanthanides from the very acidic liquid HLW, and the second one to separate the actinides from the lanthanides in aqueous acid solutions, 0.05 to 0.5 mole/L. Some work will be done on the Am/Cm separation.
- route N° 2 has a single cycle, which extracts selectively the actinides, leaving the fission products and lanthanides in the very acidic aqueous solution (> 2 mole/L). This route corresponds to the second step of route N° 1, except that the separation is done at a higher acidity, which makes it much more difficult from an experimental point of view. Its basic criteria must be carefully established using molecular design, synthesis and testing of new extractants.

It is proposed to use weak bases such as diamide reagents for the first cycle of route N° 1 and to develop the DIAMEX process further. For the second cycle of route N° 1 and for route N° 2, tripyridyltriazine (TPTZ) derivatives and new heterocyclic N donor atom extractants will be investigated.

Extraction and Selective Separation of Long Lived Nuclides by Functionalized Macrocyles

This research project involves CEA Cadarache (F) as coordinator and eight European universities. Its objectives are to synthesise and test new macrocyles for the selective extraction of strontium and actinides from MLW (after a sufficient decontamination, the resulting waste could be disposed of in surface repositories) and for the partitioning of trivalent actinides (Am, Cm) from trivalent lanthanides in HLW.

The work programme of this project has three different items: (i) synthesis of macrocycles; (ii) measurement of the extracting properties of the macrocycles; (iii) molecular modelling and X-ray and NMR structures of the complexes formed by the extractant and the radionuclide.

The synthesis of various extractants such as bis-crown ether and calixarene derivatives will be investigated for the removal of strontium. Calixarenes bearing phosphine oxide and diamide groups will be synthesised for the extraction of actinides. Macrocycles (calixarenes, resorcarenes, and others) with soft and hard donor groups will be studied for the actinide/lanthanide separation.

The extracting properties of these compounds will be thoroughly investigated by determining the complexation constants, measuring the distribution coefficients of the nuclides of interest and carrying out transport experiments through supported liquid membranes.

These experimental results will be compared with molecular mechanics and molecular dynamics simulations to understand the selective complexation of cations and to provide guidance for the synthesis of macrocycles. The measurement of NMR and X-ray structures of the complexes between the macrocyclic extractant and the cation, both as crystal and in solution, will provide additional data for modelling.

TRANSMUTATION TECHNIQUES

Two projects are dealing with transmutation techniques. The neutrons are produced in a nuclear reactor in the first project and by spallation using an accelerator in the second one.

Joint EFTTRA Experiment on Am Transmutation

This project aims at demonstrating the feasibility of ^{241}Am transmutation in a high thermal flux reactor. For this purpose, an irradiation experiment of a target containing ^{241}Am embedded in an inert matrix (e.g. cerium oxide or spinel) will be performed in the High Flux Reactor (HFR) at Petten.

The project includes different steps: fabrication of the target (stainless steel cladding encapsulating pellets made of a mixture of americium oxide and inert matrix); preparation of the irradiation facility; irradiation of the target during one-and-half year; non-destructive analyses (i.e. visual inspection, γ-ray spectrometry and tomography, neutron radiography); destructive post-irradiation examination (after de-cladding, the sample will be prepared for microscopic examination and a part of it sent to chemical analysis to determine its isotopic composition and hence the transmutation rate); interpretation of the results.

The work will be carried out by the EFTTRA (Experimental Feasibility of Targets for TRAnsmutation) group, which consists of six European laboratories and institutes. The coordinator is ECN Petten (NL).

Neutron Driven Transmutation by Adiabatic Resonance Crossing (TARC)

This project is coordinated by CERN and involves five other European research institutions. Its main objective is to develop both theoretically and experimentally a new method, the adiabatic resonance crossing (ARC), which enables to enhance strongly the capture rate of neutrons by the radionuclides to be incinerated. An experimental test will be carried out with a neutron spallation source driven by the CERN proton synchrotron. The transmutation of the radionuclides exposed to the neutron flux will be detected by the de-excitation γ-rays and the decay of the daughter nuclei with electronic methods. A first test will be made with ^{99}Tc, for which the predicted cross sections are sufficiently accurate.

The peak cross sections for neutron energies corresponding to the resonance region are much larger than for the other neutron energies. For instance, the cross section of ^{99}Tc is 4000 barn at the peak of a resonance at 5eV, but it is only 20 barn at thermal energy. Access to the resonance region can be achieved by using a transparent medium where the neutrons lose their energy in very small decrements following successive nuclear encounters (i.e. the lethargy is minimal) and their absorption is very small. In such a medium, the neutron flux per unit energy is correspondingly larger and the neutron energy decreases slowly through the resonances, leading to a very large probability of capture during crossing. Lead appears to be one of the best elements which satisfy these criteria.

All the measurements will be performed in a large lead volume with a spallation target (initially the lead itself) and several types of detectors. The radionuclides to be transmuted will be introduced in the lead assembly through thin channels. A narrow hole brings the proton beam from the CERN proton synchrotron deep inside the structure. Protons create neutrons after interaction with the lead.

The first task is to construct the lead assembly and to equip the proton beam line with instruments. Then, different types of experiments on ARC will be carried out. The transport properties of lead for neutrons at different energies will be studied. An appropriate formalism and new time-efficient Monte Carlo simulation methods for parallel computers will be developed for ARC. Finally, a conceptual design of an incinerating device based on ARC will be made.

CONCLUSION

The three areas of the research activities on new fuel cycle concepts are covered with more emphasis on strategy studies and balance between the two experimental areas. These research activities gather different scientific fields from basic chemistry (partitioning) to nuclear physics and accelerator technology. About forty different European research institutions are participating to the projects, which have started in 1996. It is expected that, at the end of the specific programme on Nuclear Fission Safety in three years from now, the scientific community will have a clearer picture on the possibilities offered by the different options investigated to reduce efficiently the radiotoxicity of nuclear waste.

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REFERENCE

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