STATUS OF THE FRENCH RESEARCH PROGRAMME FOR ACTINIDES AND FISSION PRODUCTS PARTITIONING AND TRANSMUTATION

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Abstract

The global energy context pleads in favour of a sustainable development of nuclear energy. It is a technology with a future since the demand for energy will likely increase, whereas resources will tend to get scarcer and the prospect of global warming will drive down the consumption of fossil fuel sources.

How we deal with radioactive waste is crucial in this context. From the start, the CEA has devoted considerable effort to management of the back end of the cycle. It furnished the process and techniques used in the La Hague facility to extract the re-usable materials, uranium and plutonium, and condition the resulting waste. Towards the end of the 1960s, it developed the process of vitrification for highly active waste that has become the world reference. French industry was responsible for the introduction of standard international practices with respect to waste conditioned during the processing of spent fuels. The specifications for the packages are approved by more than ten countries across the world.

The law of 1991 specifically gave new momentum to the research into waste by requesting exploration not only into deep geological storage repositories, but also into reducing the quantity and toxicity of the long-life radioactive elements present in the waste by separation and transmutation and studying their conditioning for long-term disposal. Over the past ten years, all of the above-mentioned research has been conducted by CEA and ANDRA in close collaboration with partners from industry (EDF, COGEMA, FRAMATOME-ANP), with the CNRS and various universities. A review of the situation ten years on indicates a number of significant results that have changed the prospects for nuclear waste management.

The paper focus on separation and transmutation R&D programme and main results over these ten last years. The massive research programme on enhanced separation, conducted by CEA and supported by broad international co-operation, has recently achieved some vital progress. Based on real solutions derived from the La Hague process, the CEA demonstrated the lab-scale feasibility of extracting minor actinides and some fission products (I, Cs and Tc) using an hydrometallurgical process that can be extrapolated on the industrial scale. The CEA also conducted programmes proving the technical feasibility of the elimination of minor actinides and fission products by transmutation: fabrication of

specific targets and fuels for transmutation tests in the HFR and Phénix reactors, neutronics and technology studies for ADS developments in order to support the MEGAPIE, TRADE and MYRRHA experiments and the future 100 MW international ADS demonstrator. Scenarios studies aimed at stabilising the inventory with long-lived radionuclides, plutonium, minor actinides and certain long-lived fission products in different nuclear-power-plant parks and to verify the feasibility at the level of the cycle facilities and fuels involved in those scenarios. Three French Research Groups CEA-CNRS carry out partitioning (PRACTIS) and transmutation (NOMADE and GEDEON) more basic studies.

Introduction

The radioactive content of the waste generated by the nuclear-power cycle constitutes a potential risk for human beings and the environment. In order to control it, the safest management methods possible must be implemented. Although that management already has a long scientific and technical history, interrogations and concerns have emerged in the public and have prompted the legislator to address the issue.

In France, the Law of 30 December 1991 sets the major orientations of the public policy in that field by prescribing the research areas to be explored. It constitutes the first global legislative instrument for the management of high-level long-lived radioactive waste. It identifies the principles by which all radioactive-waste-management methods must abide: protection of nature, the environment and health, as well as respect for the rights of future generations. It prescribes the implementation of a significant structured research programme and its associated research schedule until 2006. In addition, it also institutes a National Review Board (*Commission nationale d'évaluation – CNE*) responsible for following up research and for reporting to the government on a yearly basis.

The Law of 1991 advocates a wide exploration of solutions concerning the management of radioactive waste. With that goal in mind, three research areas have been defined:

- Area 1 relating to partitioning and transmutation deals with various potential solutions capable of reducing substantially the mass and toxicity of long-lived radionuclides as a source of risk over the long term.
- Area 2 aims at defining the conditions under which a repository, whether reversible or irreversible, may be implemented and operated in a deep geological formation, where various high-level long-lived waste packages would be emplaced. Underground laboratories are essential tools to that research programme.
- Area 3 is dedicated to research on packaging and long-term storage. It covers the development and qualification of mechanisms ensuring that the waste is kept under satisfactory safety conditions over periods of several decades, pending the availability of the management methods to be developed in Areas 1 and 2.

The Law stipulates that, by 2006, the research programme shall provide to the legislator and public authorities the necessary elements to get an overall idea of the situation and to decide whether it is appropriate to create a repository for high-level long-lived radioactive waste.

This paper presents the large amount of results dealing with partitioning and transmutation that have been accumulated since the beginning of the national programme by the CEA and other important French R and D or industrial agencies, such as the CNRS (French National Council for Scientific Research), EDF (Électricité de France), COGEMA or FRAMATOME-ANP.

Overview of priorities for the partitioning and transmutation studies

The national programme for partitioning and transmutation is defined as a function of relevant priorities assigned in the framework of each research area, with due account to the fact that the objective is to provide the French government and Parliament by 2006 with sound elements that will allow them to implement a strategy for the management of long-lived radioactive waste. Those priorities include:

- to study the feasibility of various partitioning processes constituting a prerequisite common core for more specific management possibilities for long-lived radionuclides; to select and optimise those processes in relation to the efficiency of the actinide-burning systems and of the transmutation systems for long-lived fission products, and the foreseeable specific types of packaging for those various products; to study the reprocessibility of fuels and targets for transmutation purposes, particularly pyrochemical processes;

- to study scenarios aiming at stabilising the inventory with long-lived radionuclides, plutonium, minor actinides and certain long-lived fission products in nuclear-power-plant geometries (the so-called “single-stage” approach) and to verify feasibility at the level of the cycle facilities and fuels involved in those scenarios; to examine also elimination possibilities from that inventory;

- to assess different innovative reactor systems that ensure a good control of high-level long-lived waste with a significant effort dedicated to hybrid systems, by contemplating a mixed (“dual-stage”) set of nuclear power plants, where a small number of specialised burner reactors would burn up the long-lived radionuclides generated by nuclear-power reactors, and by examining the possibilities of reducing significantly the waste inventory;

- to study the feasibility of an hybrid-system demonstrator.

Programmes and results for partitioning and transmutation of actinides and fission products

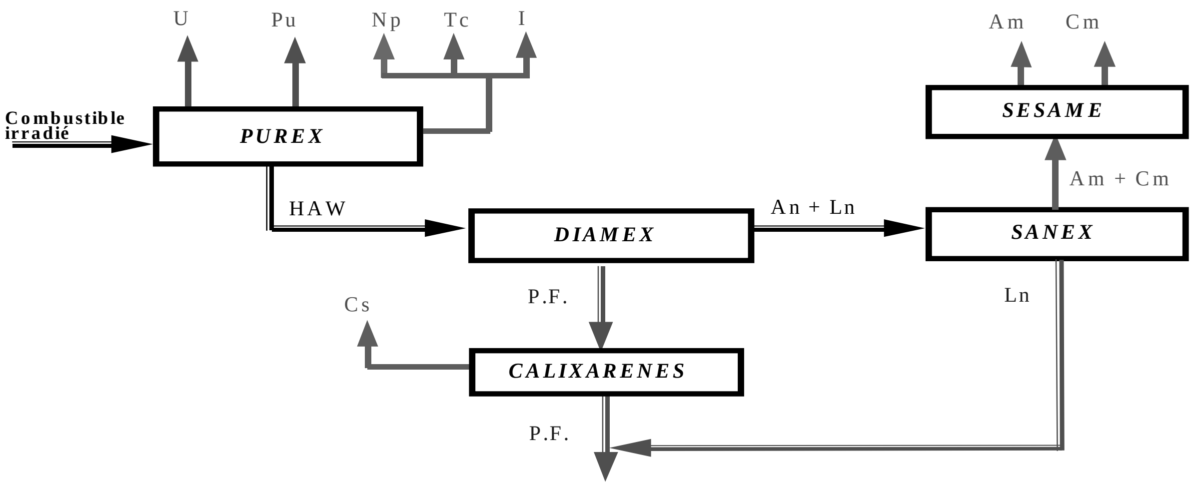
The overall objective is to investigate solutions capable of reducing the quantity of long-lived radioactive waste resulting from energy generation, by separating it (through chemical-separation processes before the reprocessing of spent fuels) from the other elements it contains and by transmuting the largest possible part of the isolated waste under a neutron flux either inside present or future critical (such as gas cooled thermal-spectrum or fast-spectrum) reactors or innovative systems (notably, hybrid systems) in order to transform it as soon as possible into stable (non-radioactive) atoms. [1]

The studies cover minor actinides (americium, curium, neptunium) that, except for plutonium, represent most of the long-term radiotoxic inventory of nuclear waste and certain fission products having a long-lived isotope, a high relative abundance in the spent fuel as well as chemical properties that make them potentially mobile: iodine, caesium and technetium. Since plutonium is both a recyclable energetic material and the main contributor to potential long-term radiotoxicity, all scenario studies considered must be consistent with the policies contemplated for the long-term management of plutonium. [2]

As a complement to the “Partitioning-Transmutation” reference strategy, studies are also undertaken on the alternative aspect of “Partitioning-Conditioning”, where partitioned – yet “untransmutable” – radionuclides would be conditioned in new specific matrices. Conditioning studies concern particularly the development of new specific matrices for partitioned radionuclides. Since their chemical form is known, it is possible to integrate them very closely in a defined crystalline form and thus achieve a high durability level. Programmes cover in first priority very-long-lived fission products such as iodine 129 and caesium 135. The goal of the research programme is to bring all those processes to the technical-feasibility stage by 2006, while combining them with an industrial-feasibility assessment.

The reference approaches of the programme on extensive partitioning are based on extraction during the liquid phase, either by adapting the PUREX process used in the industry to reprocess fuel or in developing new PUREX-downstream complementary extraction processes (Figure 1). The scientific feasibility of these processes (development of extracting molecules and validation of basic designs at lab-scale) have been established in 2001 with values of separation as high as 99.9% for recovered americium and curium from HLW). Americium and curium are partitioned by new extracting molecules specifically developed for that purpose with the French process which has been selected at the end of the scientific feasibility stage; caesium is separated with calixarene-crown molecules, whereas neptunium, iodine and technetium are extracted by technical adaptations of the existing PUREX process which is already used at La Hague plant. The next goal of the study programme for partitioning processes will be to reach the technical feasibility stage before 2005 (complete process validation) with the assessment of the industrial feasibility (implementation conditions, production of secondary waste, safety conditions, costs, etc.).

Figure 1. **Diagram of separation processes**



Alternative processes are also being investigated, notably those involving pyrochemistry for the reprocessing of transmutation targets (hardly soluble in aqueous nitric acid solution) and some innovative fuels of future reactors.

That research on extensive partitioning relies on basic studies in theoretical chemistry and on models, notably in association with the PRACTIS Research Group that includes representatives from the CNRS, the CEA, ANDRA (National Radioactive Waste Management Agency) and EDF.

The purpose of transmutation programmes is to assess the possibility of eliminating long-lived radioactive waste through critical reactors (present or future) or innovative sub critical hybrid systems.

With regard to critical reactors, programmes are divided into three major themes:

- studies on reactor cores and transmutation neutronics in existing PWR-type reactors with evolutions of current management modes (evoluted CORAIL MOX assembly, APA concept RMA improved-moderation core, plutonium on an enriched-uranium support MIX fuel) and in future gas cooled thermal or fast-flux reactors;

- experimental studies on fuels and targets, including in particular an experimental irradiation programme;

- basic physics in order to complete and improve nuclear data (effective sections of nuclear reactions) and nuclear-reaction models. That task requires an evaluation at each step of uncertainty.

Programmes on transmutation within innovative systems are conducted notably in the framework of the GEDEON Research Group that includes representatives from the CEA, the CNRS and EDF. FRAMATOME-ANP and ANDRA are also members of its Scientific Board.

The experimental irradiation programme is based, for a large part, on tests in the fast flux Phénix reactor which power restart is planned at the very end of 2002. Flux conditions of Phénix are well suited to allow the studies of irradiation damages of fuels and targets for transmutation under representative conditions of fast and partly moderated flux, which are considered to be the most efficient for transmutation of minor actinides and some long live fission products. A first phase of experiments, ready at the power restart time, will include:

- targets ECRIX B and H, made of pellets containing americium oxide microdispersed in an inert magnesia matrix, under fast (B) and partly moderated (H) neutron flux;

- 3 sub assemblies METAPHIX 1, 2 and 3 containing experimental pins with metallic UPuZr fuel and dispersed minor actinides, inside an agreement with the Japanese CRIEPI agency;

- irradiation ANTICORP 1 consisting of 3 pins containing pure metallic ^{99}Tc;

- PROFIL R for the measurement of capture sections of fission products, lanthanides, actinides isotopes under fast flux neutrons.

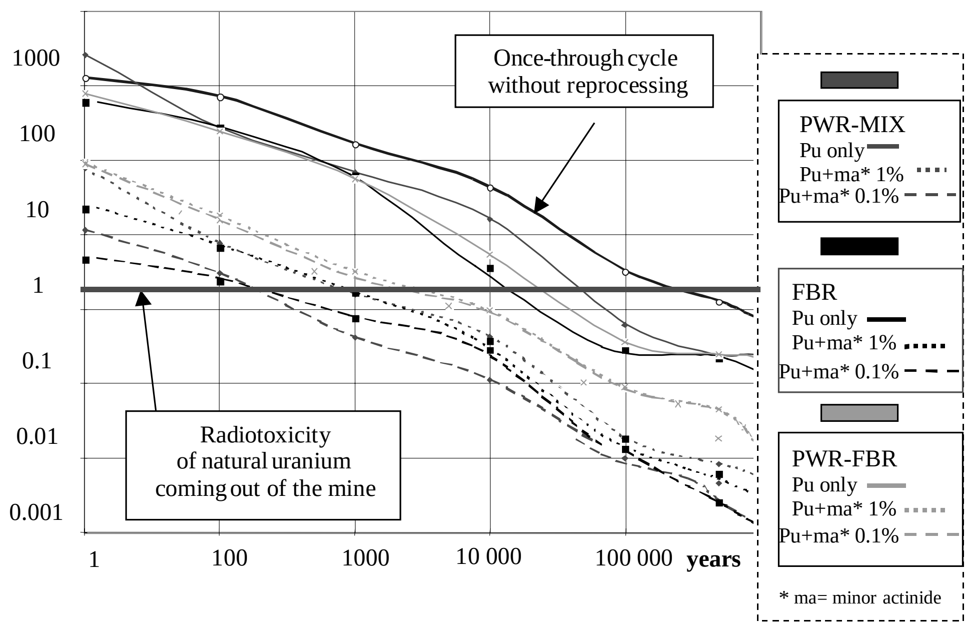
A second phase, which will integrate the knowledge gained with the experimental irradiations carried earlier in the thermal French SILOE and European HFR and the R and D results about fabrication process (macrodispersion and new matrix materials) will start in 2004; a joint CEA-DOE-ITU experiment (oxide, nitride and metallic ADS fuels) is also planned beginning in 2005.

Started in 1998, a reflection on innovative nuclear systems has resulted in a summary report on hybrid systems where the project of a European demonstrator was indicated. The CEA and the CNRS instituted a co-ordination group to prepare a documented report that was presented to the National Review Board in February 2001. This Development Plan is in the process to be updated with taking into account recent results and necessary evolutions for implementation of large and heavy cost experiments. Progress has been gained about the nuclear basic data still necessary to control the neutronic coupling and to design an ADS transmutation core (MUSE experiment corresponding to the first coupling of the CEA MASURCA reactor with a low power neutron source) and about the understanding of the behaviour of liquid spallation target with the specific irradiation and corrosion conditions to which the window (between beam and core) is submitted; a preliminary design of an experimental ADS is in progress inside the framework of an European project partly supported by nuclear industrials such as FRAMATOME and ANSALDO. Next steps will be the implementations of the 1 MW MEGAPIE lead-bismuth spallation target scheduled in operation in 2005, the construction of a demonstration experiment at a power of few hundreds of kW based on the Italian TRIGA reactor driven by a cyclotron accelerator with a solid tungsten target due to operation in 2007 and the possible implementation of the more powerful and representative experimental ADS MYRRHA facility in Belgium. All these projects are being developed with a clear support of the EURATOM Agency.

Finally, the potential performances of partitioning-transmutation are assessed globally through scenario studies. Different major scenario families are considered. The first families call upon current technologies: PWR/EPR-type facilities using plutonium in MIX fuel and burning minor actinides as an option; fast-breeder isogenerators ensuring the multirecycling of plutonium and minor actinides (monorecycling of minor actinides as an option); mixed PWRs (UOX and MOX) and fast-breeder reactors burning plutonium and, with certain variations, minor actinides and certain long-lived fission products. The other families rely on innovative technologies: mixed PWRs (UOX) and hybrid systems burning plutonium, minor actinides and long-lived fission products; "dual-stage" reactors where PWRs and new innovative high temperature gas cooled thermal then fast reactors ensure the multirecycling of plutonium and the transmutation of past and existing nuclear wastes.

A first appraisal of the scenarios with current technology reactors has been presented in 2001 in order to illustrate the achievements made so far on the stabilisation – and even reduction – perspectives over time of the quantities of long-lived radionuclides (Figure 2) resulting from energy generation and to quantify secondary waste that may be low, without being totally inexistent. These results indicate that the multi recycling of plutonium offers a first advantage by reducing the potential radiotoxicity of ultimate waste by a factor of 3 to 10, compared to once-through cycle, and that partitioning and transmutation of minor actinides would bring a further advantage reflected by a global reduction factor in the order of 100, depending on the partitioning/transmutation modes considered.

Figure 2. **Radiotoxicity after various partitioning and transmutation operations**



Conclusion

In France, spent nuclear fuels are reprocessed in order to recover uranium and plutonium. The resulting waste are conditioned in line and then, temporarily stored. For further waste management, the Parliament has launched in 1991 a research programme, which must converge in 2006. It has first to deal with the existing wastes and those that have yet to be produced by the existing energy production systems. Then, it has to answer the question whether it is possible to reduce the radiotoxic potential of the ultimate waste. The answer is a probable "yes", provided that important modifications are made on the reactors and fuel cycle systems, which means important investments. It will take time, several tens of years, as the nuclear energy production system is relatively young. However, if nuclear energy production is to be continued, the evolution is without doubt in the direction of reducing the long-term radiotoxic potential in the waste. Partitioning and transmutation is an inescapable channel.

Transmutation of separated radionuclides, mainly plutonium and minor actinides, could be done in existing PWR, although the time to reach equilibrium may be rather long. Fast reactors could do the job more efficiently. They would be either critical or driven by an accelerator.

CEA has chosen Gas Cooled Reactors as champions for the future generation of reactors. CEA is strongly involved in the US-started Generation IV forum. First, aiming at recovering knowledge on High Temperature Reactors, it aims at developing a fast neutron version of the concept. Ability to transmute minor actinides will be a part of the requested properties.

However, one must keep in mind that PWR can be operating in France up to the end of this century. Although ADS cannot compete with critical reactors for energy production, there is a niche where they have a possible role in burning the minor actinides that are produced by PWR. A fuel containing a large proportion of americium looses the quasi-magical properties of the mix U5-U8 in stabilising the neutrons dynamics of critical reactors (Doppler effect and delayed neutrons). In this context, CEA is wishing to share the development of a demo ADS somewhere in the world in collaboration large enough to cover the cost of it. From its own point of view, CEA is wishing to put most of its effort in the development of energy producing reactors.

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