# STATUS OF THE CEA PROGRAMME ON FUELS FOR TRANSMUTATION

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## Abstract

For the management of high level and long life radioactive waste, a large and continuous research and development effort is carried out to provide a wide range of scientific and technical alternatives according to three lines: partitioning and transmutation, disposal in deep geological formations and long term interim surface or subsurface storage.

For the technological part of the feasibility of waste transmutation in reactors, a large programme dedicated to the development of fuels and materials is underway. It includes studies in the field of materials and composite microstructures and involves experiments in different reactors.

Since the last OECD/NEA Information Exchange Meeting in Madrid on December 2000, the progress of this programme is presented and updated.

## Introduction

For the management of high level, long life radioactive waste and in the frame of the research dealing with the French Law of December 91, the partitioning and transmutation (P&T) strategy to reduce the quantity and the toxicity of the waste is being evaluated along with other alternatives which are disposal in deep geological formations and long term interim surface or subsurface storage. [1]

The P&T research is focused on minor actinides (MA: neptunium, americium, curium) which represent the major long term radiotoxic elements in the waste, and on a few fission products which are very long life isotopes, abundant and potentially mobile (technetium, iodine and cesium). [2]

The aim of the transmutation programme is to evaluate the transmutation performances with different reactor configurations (scenarios) relying either on well-known technologies reactors (Pressurised Water Reactors (PWR) and Na-cooled Fast neutrons Reactors (FR)) or on innovative reactors (Gas-cooled reactors) and/or Accelerator-driven Systems (ADS). [3]

After a general presentation of the R and D programme, emphasis is put on the research carried out on fuels and materials for PWR (called EPR in the future) and FR (called EFR) in order to insure the technical feasibility of the concepts.

## Concepts and scenarios

Scenario studies are performed to insure the theoretical (or "scientific") feasibility of the waste transmutation from the point of view of the reactor core physics. The aim is to demonstrate the possibility to reach the equilibrium of the material inventory (consumption = production) and to calculate the isotopic compositions of the actinides incorporated in the fuels. Since Pu is both a recyclable energetic material and the main contributor to the potential long-term radio toxicity, all the transmutation scenarios include the Pu management.

As a main achievement of the scenario studies, the scientific feasibility of the waste transmutation has been established, according to different concepts:

- homogeneous recycling of Pu and minor actinides either in the 100% EPR park (with a $^{235}U$ enriched fuel) or in the 100% EFR park. The latter allows additional transmutation of long life fission products (LLFP) in moderated targets;
- in the mix EPR – EFR park with homogeneous recycling of Pu and Np, and transmutation of Am and Cm in moderated targets (heterogeneous recycling). [4]

The studies to reach the scientific feasibility of the waste transmutation using innovative systems (gas-cooled reactors or ADS) are going on, in a parallel work programme. [5]

After the scientific feasibility, a second step is to obtain the technical feasibility in terms of fuel developments, fuel cycle impacts, safety and economics.

For the homogeneous recycling, many elements already exist and only limited complements are requested.

For the heterogeneous recycling, the target material, U, Pu-free and with a high content of minor actinides, can be either a solid solution of actinides or a dispersed fuel. Dispersed fuel is inclusions of an actinide compound dispersed in a matrix that must be as inert as possible towards neutrons as well as stable under irradiation and able to cope with very high fission rates reached in the inclusions (once through concept).

Because of the technological discontinuity with regards to U, Pu oxide fuels, a large R and D effort is needed, covering the manufacturing process and the characterisation of the basic properties of the non irradiated materials, the realisation of experimental irradiation and post irradiation examinations.

That is the goal of the going-on programme which is conducted with industrial partners in the frame of various international collaborations and where the irradiation in PHENIX take an important place.

This paper will describe general studies made to improve our knowledge in different fields such as inert matrix, fissile and structural materials or manufacturing processes. Then it will deal with studies on the homogeneous mode and those concerning the heterogeneous one, to finish with the description of what is done about dedicated fuels.

## General studies

### *Experiments linked to nuclear data*

In order to improve the knowledge of the cross-sections of the various interesting nuclei, two specific experiments are being prepared, in which a large number of separated minor actinide and long life fission product isotopes in milligram quantities will be irradiated in well known flux conditions, fast or locally moderated spectrum: PROFIL R in a quasi-standard PHENIX sub assembly and PROFIL M is a rig to be introduced in a moderated $^{11}B_4C$ device.

### *Experiments linked to structural materials*

Concerning structural materials used to build the irradiation device some irradiation such as ELIXIR or PNC 5 and 6 are on way in the PHENIX reactor. They will bring important information on the behaviour under irradiation of specials stainless steels in order to select a well known and the most appropriate one.

### *Experiments on moderators*

In the field of moderators, [4] it has been shown that a locally moderated spectrum in fast neutron reactors is needed to improve the transmutation rate. This leads to the design and realisation for the PHENIX irradiation programme of two specific devices. The choice was two solid moderator materials for which sufficient knowledge and manufacturing capabilities existed: $^{11}B_4C$ and $CaH_x$. The DMC-1 device contains boron carbide and the DMC-2 one contains the calcium hydride. Both devices are now fabricated and ready to be irradiated as soon as PHENIX restarts.

A complete characterisation of the $CaH_x$ (chemical reactivity, thermal properties, dissociation temperature in different atmospheres…) has been realised. This work was initiated in the so-called MODIX programme devoted to the irradiation of hydride moderators but is now postponed.

***Selection of matrix***

Matrices selected for composite fuels or targets must be as inert as possible towards neutrons as well as stable under irradiation. Selection is made with different steps in the approach:

- First, the requirements for the matrix are to gather good thermal and mechanical properties and sufficient chemical stability in the course of the actinide phase evolution.
- The initial selection takes into account available data, [6] essentially out of pile properties, data on in-pile behaviour being rather scarce.
- Then a second step involves uranium oxide target to simulate the later MA target. It is useful to study fast neutrons interaction, effects of fission products (FP) and some gas production in order to optimise microstructures. The actinide dispersion is especially tested with the introduction of macro masses to concentrate radiation effects due to the fission fragments or alpha decay in the surrounding of the macro masses.
- Finally, irradiation with MA targets are performed to test the chemical interactions between the compound and the matrix and the behaviour of the material especially concerning the management of the helium production (coming from the alpha decay of americium) which could lead to an important swelling of the pellets and then to a clad failure.

The outcomes drawn from the first irradiation, especially T3 in HFR, TANOX and THERMHET, [7,8] in SILOE, are gathered below:

- If spinel, $MgAl_2O_4$, behaves very well under high fast neutron fluence (more than $22.10^{26}$ $n.m^{-2}$), [9] fission products recoil and $\alpha$ decay lead to severe damages in the matrix. [10]
- To avoid the material swelling, the target must operate at a temperature high enough to favour defects recovery and gas diffusion.
- The macro dispersion design turns out to be efficient to limit the damage of the matrix and has to be compared with the micro dispersed one. [11]
- Magnesia, MgO, is still considered as a promising candidate as matrix material.

Further experiments are planned in PHENIX reactor in order to broaden the experimental database especially on the reference matrix MgO.

- MATINA 1A: MATINA 1 [13] was an irradiation experiment carried out up to a fast fluence of $2.10^{26}$ $n.m^{-2}$ (E>0.1Mev) of various matrices (MgO, $MgAl_2O_4$, $Al_2O_3$, $Y_3Al_5O_{12}$, TiN, W, V, Nb, Cr). Some of them (MgO, $MgAl_2O_4$, $Al_2O_3$) were mixed with $UO_2$ (micro dispersion) to study the effects of FP. In 1996, 2 pins ($MgO+UO_2$, $MgAl_2O_4+UO_2$) were discharged for destructive examinations in view of the matrix selection for the ECRIX experiments. The remaining pins were re-introduced in PHENIX (MATINA 1A) for continuation of their irradiation up to $6.10^{26}$ $n.m^{-2}$. This irradiation will bring important data on magnesia at high fluence, what is missing at present.
- MATINA 2-3 will take into account the main results of the earlier irradiation. It will include the study of magnesia alone, with micro and macro dispersed inclusions of uranium oxide and the effects of the temperature on defect recovery and gas diffusion. The device will contain also two other interesting compounds, known to have a good structural stability under irradiation: Yttrium-stabilised zirconia and cerium pyrochlore compounds.

### *Selection of the minor actinides compounds*

Concerning the selection of MA compounds, first experiments (T4 or ECRIX) were logically focused on americium dioxide ($AmO_{2-x}$). They are now extended to new and more complex Am compounds, like zirconia-based solid solutions, which should present attractive properties as thermal stability of the cubic phase. For example, the pyrochlore form $Am_2Zr_2O_7$ is being characterised in the Oak Ridge National Laboratory in the frame of a bilateral collaboration.

In each case we pay attention to the manufacturing process which can be influent on the results and which is very important on the economical point of view.

## Fuels for homogeneous recycling in fast reactors

In this case limited amounts of Np, Am, Cm are introduced in the standard fuel to limit the impact on the fuel performance.

As confirmed by the SUPERFACT [14] experiment in PHENIX where (U, Pu, Np)$O_2$ and (U, Pu, Am)$O_2$ with 2% respectively Np and Am, were successfully irradiated at medium burn-up (6,5at% and a transmutation rate of 36%), the addition of limited amounts of Np, Am, Cm in the standard fuel should not affect deeply the FR oxide fuel behaviour.

The TRABANT 1 pin irradiated in HFR with 5% of Np in a high Pu content oxide fuel gave similar conclusions.

Performance of a metallic fuel with a low content of minor actinides (Np+Am+Cm) and rare earths, will be studied in the METAPHIX experiments conducted in the scope of a contract with ITU on behalf of the Japanese CRIEPI. METAPHIX 1,2,3 are three 19-pins rig capsules, each loaded with three experimental sodium bonded pins containing a metallic alloy UPuZr with 2 to 5% of minor actinides and/or rare-earths. The target burn ups are 2, 7 and 11at%. [15] This irradiation will take place in PHENIX.

## Fuels for heterogeneous recycling

### *Incineration of minor actinides*

#### *Requirement and design of MA targets*

In the case of the heterogeneous recycling, MA targets are loaded with a high content of actinides in some areas of the core. Fast neutrons reactors offer determinate advantages for the transmutation strategy because ratio of fission to capture is more favourable than in a thermal flux and a lot of neutrons are available.

Targets have specific features when compared to standard fuels: the linear heating rate raises largely during the irradiation, the helium production is much higher than the conventional fission gases production, and the burn-up to be reached in the actinide inclusions is about 90% of the initial heavy atoms in a once through recycling. These targets are U-free fuels in order to prevent the formation of new actinides and the MA compound is diluted in a ceramic or metal matrix (see above).

All this represents a technological discontinuity with regards to U and Pu oxides cycle. So development is needed in different areas, first with the characterisation of the basic properties of the fuel components, and for the elaboration of the manufacturing process, then with the realisation of experimental irradiation and post irradiation examinations to obtain elements on the behaviour under irradiation.

*Outcomes drawn from the first irradiation*

Some irradiation of dispersed fuel have already been done as EFTTRA T4 and T4bis. [10] Pins contained americium oxide target dispersed in a spinel matrix. The pins were successfully fabricated and irradiated in the High Flux Reactor in Petten. They reached respectively 28% and 72% fissile initial metal atom (FIMA) with a transmutation rate close to 100%. They allowed to underline the large impact of the helium production on the target performance because of the large swelling as a consequence of radiation damage and gases accumulation. Design improvement (porous target, Pu loading), selection of new matrix (Y-St $ZrO_2$, MgO) are then be proposed to take into account this experience feedback and will be tested in the future experiments.

The work performed up to now, has brought numerous results in terms of dispersed fuel manufacturing and characterisation of their basic properties as well.

*Further experiments*

The ECRIX [16] programme consists in two irradiation, identical as regards the material constituting the americium target but different as regards the target irradiation conditions: the irradiation neutron spectrum will be moderated by two different materials: $^{11}B_4C$ for ECRIX B and CaHx for ECRIX H.

The fuel of the two ECRIX [17] pins is a composite target with americium oxide "micro-dispersed" in the MgO matrix. The manufacturing of the pellets has been made using powder metallurgy. The objective is to reach a fission rate of 30at%.

Compared to the other experiments, the simultaneous implementation of a fast neutron reactor flux, a neutron spectrum converter and an americium target gives these experiments a prototypic aspect.

To open the field of investigation, a further irradiation is planned to select the optimised composite and the americium compounds. A research axis will concern the stabilisation of the americium oxide in the cubic structure of the yttrium stabilised zirconia $(Am,Zr,Y)O_{2-x}$ . The CAMIX irradiation (Composites of AMericium in PHÉNIX) will cover the optimisation of the actinide compound. [18]

A second axis of the investigation will concern the dispersion mode of the americium compound in the inert matrix considering the macro dispersed concept (with $(Am,Zr,Y)O_{2-x}$ in MgO). Improvement is expected with this concept which must concentrate radiations effects due to fission fragments or alpha decay in a small shell around the macro masses. Such a process is to be operational for the COCHIX irradiation (Concept Optimised target in PHENIX). [18]

The manufacturing of the “macro-dispersed” type composite is now controlled with uranium and transposition to americium is the challenge in view of the fabrication for the CAMIX-COCHIX experiment.

It is also important to quote the T5 irradiation planned in the High Flux reactor in Petten. [19] Its main goals are the management of matrix damage, the choice of a stable compound and the management of gas production. This last point is still a matter of discussion, the choice going from complete retention to complete release of fission gases. For the T5 experiment, a porous MgO matrix will be studied.

### *Incineration of long live fission products*

The way of transmutation which was chosen is to load the LLFP in a locally moderated spectrum of a fast reactor. Studies are focused on iodine and technetium (for cesium the reference strategy should be direct disposal).

In addition to first experiments T1 and T2 in HFR, [20] the ANTICORP-1 experiment in PHENIX will include in one rig 3 pins with metallic technetium 99 already manufactured by ITU. The irradiation in a $CaH_x$ moderated device makes the conditions for a real prototypic experiment.

Others studies, in close partnership with NRG, are made on various possible iodine compounds.

## Dedicated fuels

Some scenarios studies deals with reactors dedicated to burn the waste. Dedicated fuels contains essentially MA but also some degraded Pu for neutronic consideration and are preferably free of uranium to avoid the formation of new actinides.

From the analysis based on the current knowledge on actinide compounds, nitrides, e.g. (Pu, MA, Zr)N, and oxides, e.g. (Pu, MA)$O_2$+MgO, are thought to be promising candidates for the dedicated reactors. However, a large R&D programme is needed to enlarge our knowledge on such compounds and to optimise the fuel design to make it sure and safe. That is the objective of the FUTURE and CONFIRM European programmes, respectively devoted to oxide and nitride fuels.

An irradiation project in PHENIX, called FUTURIX, is currently investigated to test in similar and representative conditions the different candidates thought to have a good potential: they are the fuels studied in the CONFIRM and FUTURE programme, but also metallic fuels (Pu-MA-Zr) developed by ANL. This irradiation project should be a collaboration between CEA, ITU, DOE and JAERI.

A CEA/MINATOM work programme is in progress in Russia. The BORA BORA irradiation in BOR60 is designed to test various fuel concepts developed in the frame of CAPRA Pu management programme including the test of $PuO_2$-MgO and (Pu, Zr)N. This experiment will bring data on the behaviour of these fuels [21] which have a great interest for the dedicated fuels programme.

Furthermore, the AMBOINE project in its initial phase deals with the feasibility of the VIPAC concept for MA transmutation and allows to investigate innovative solutions to the problem of the curium management. [22]

## Conclusion

Efforts made for several years to develop fuels and materials in order to prove the technical feasibility of the different transmutation strategies, lead to acquire knowledge on selected materials and concepts: basic properties, manufacturing process, performance under irradiation. The success of the T4bis experiment, that reached performance close to the scenario requirements, is full of promise and outcomes.

The experimental programme planned in PHENIX, has been consolidated and the preparation work has reached marked milestones like the irradiation devices manufacturing, the realisation of the first experimental pins with $AmO_2$/MgO composite, the various calculations and safety files necessary to the start of the experiments planned in the beginning of 2003 (METAPHIX 1,2,3; ECRIX B,H; PROFIL R).

The first experimental results allow an optimisation of the targets that will be introduced in a second phase, from 2004. The necessity to cover a large domain of parameters (including various neutron spectrum and reactor types) leads to use analytical experiments owing a good understanding of the phenomena together with some technological ones. Results from this programme together with those coming from other irradiation and of international programmes will be used to answer to the technical feasibility of the different concepts tested for the minor actinides transmutation.

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