TRANSMUTATION STUDIES IN FRANCE, R&D PROGRAMME ON FUELS AND TARGETS

M. Boidron[1], N. Chauvin[1], J.C. Garnier[1], S Pillon[1], G. Vambenepe[2]
[1]Direction des Réacteurs Nucléaires, CEA, Cadarache 13108 Saint Paul Lez Durance Cedex, France
[2]EdF, 12-14 Av. Dutrievoz, 69628 Villeurbanne Cedex, France

Abstract

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

For the line one, and in close link with the partitioning studies, research is carried out to evaluate the transmutation potential of long-lived waste in appropriate reactors configurations (scenarios) relying on current technologies as well as innovative reactors. Performed to evaluate the theoretical feasibility of the Pu consumption and waste transmutation from the point of view of the reactor cores physics to reach the equilibrium of the material fluxes (i.e. consumption = production) and of the isotopic compositions of the fuels, these studies insure the "scientific" part of the transmutation feasibility.

For the technological part of the feasibility of waste transmutation in reactors, a large programme on fuel development is underway. This includes solutions based on the advanced concepts for plutonium fuels in PWR and the development of specific fuels and targets for transmutation in fast reactors in the critical or sub-critical state.

For the waste transmutation in fast reactors, an important programme has been launched to develop specific fuels and targets with experiments at various stages of preparation in different experimental reactors including Phénix. Composite fuels as well as particle fuels are considered. This programme is presented and recent results concerning the preparation of the experiments, the characterisation of the compounds properties, the thermal and mechanical modelling and the behaviour of U free fuels are given.

1. Introduction

For the management of high level and long live radioactive waste and in the frame of the first line of research identified in the French Law of December 91, the potential of a partitioning and transmutation strategy to reduce the quantity and the toxicity of the waste is to be evaluated (along with the other alternatives that are disposal in deep geological formations and long term interim surface or subsurface storage [1]). The research is centred on minor actinides (americium, curium, neptunium) which represent the majority of the long-term radiotoxic elements in the waste, once plutonium has been extracted, and certain fission products with a very long-lived isotope, relatively abundant and potentially mobile (technetium, iodine and caesium). The objective of the partitioning studies is to develop chemical processes to obtain advanced partitioning of radionuclides to complete the partitioning of uranium and plutonium. The development of extracting molecules and the validation of the basic concepts, which corresponds to the stage of scientific feasibility of this research, is currently underway, and the process validation (technical feasibility) is to be achieved for 2006.

Within the same time, the objectives of the transmutation programme are to evaluate the transmutation potential of long lived waste in appropriate reactor configurations (scenarios) relying on current technologies (Pressurised Water Reactor and Fast neutrons Reactors) as well as innovative reactors (with dedicated systems such as the accelerator driven systems) and also to study the materials to be used for the new type of fuels suitable for transmutation in order to define the first elements of adequate solutions.

After a general presentation of what is considered in the R&D programmes on transmutation and a short review of the scenarios studies, emphasis is led on the presentation of the research carried out on the materials needed for the future fuels necessary for a transmutation strategy.

2. Strategy for a long-term work programme

Three steps can be identified before a possible industrial development of the transmutation strategy, the first being the stage of scientific feasibility in which the possibility of transmutation is evaluated on the basis of the reactor cores physics, the second including detailed studies to obtain elements of technical feasibility in terms of fuel cycle impacts, safety and economic considerations and fuel development, and the third dealing with the industrial feasibility with a stage of demonstration in representative conditions of the chosen technologies.

Studies on transmutation deal with the two first steps with a special emphasis on the fuels studies, the development of fuels being one of the key points to reach the objectives of waste transmutation.

Feasibility of the transmutation have been considered in different parks of reactors taking into account in the first part of the scenarios the reactors of current technology (PWR and FR) ensuring electricity production as well as incineration of waste, and in the second part, the reactors dedicated to transmutation with high content of waste that can be either critical or ADS. The third part to be considered are the other innovative technologies and cycles like molten salt reactors and pyrochemistry that can be alternatives to the other ways. A first review of the results including scientific and technological feasibility is planned in 2001 for the scenarios dealing with reactors of current technology, in 2003 for the innovative reactors and in 2005 for the alternative ones.

Since plutonium is both a recyclable energy material and the main contributor to potential long-term radio toxicity, the start point of all the scenarios is the management of plutonium in the fleet of reactors and the research on transmutation is connected to the studies linked to the plutonium consumption (links identified in the Capra Cadra programme [2]), the development of advanced

concepts for plutonium consumption [3,4], and particle fuels [5] and the research on new nuclear technologies for the future. In addition to competitiveness, the new types of reactors will have to present marked progresses in terms of minimisation of natural resources, safety and reduction of waste production. These requirements induce research to develop fuels with good thermal characteristics, able to sustain high temperatures, to reach high specific power and high burn ups. The solutions to be found for the transmutation have to be connected to these developments

3. Transmutation scenarios

The scenario studies were performed to insure the theoretical (or "scientific") feasibility of the Pu consumption and waste transmutation from the point of view of the reactor core physics to reach the equilibrium of the material fluxes (i.e. consumption = production) and of the isotopic compositions of the fuels.

Taking the open cycle as a reference (Reference case in Table 1), five families of scenarios have been considered in agreement with the French National Commission of Evaluation; three scenarios rely on existing technology (PWR of the Franco-German European Pressurised Reactor–EPR type) using plutonium and optionally ensuring the incineration of minor actinides (Case 1); fast neutron reactors of the European Fast Reactor-EFR type ensuring the multirecycling of plutonium and minor actinides (Case 2) (optionally the mono-recycling of minor actinides); a combination of PWR (UOX and MOX) and fast neutron reactors (Case 3) to burn the plutonium and incinerate, according to the variety, the minor actinides and some long-lived fission products). The other two cases use innovative technologies (combination of PWR [UOX] reactors and dedicated systems such as the ADS); a double component park considers PWR and dedicated fast reactors (Case 4), and in the "double strata" system (Case 5), the first stratum contains PWR and fast neutron reactors that multirecycle the plutonium, and in the second stratum, the hybrids transmute the minor actinides and long-lived fission products.

The characteristics of the various reactor fleets considered are summarised in Table 1.

Table 1. **Description of scenarios**

Scenario	**PWR UO_2**	**PWR MIX**	**EFR**	**EFR**	**Dedicated FR U free Pu+Am+Cm (double component)**	**ADS (U free) (Mainly Am-Cm recycling) (double strata scenario)**
Ref.	100%					
1		100%				
2			100%			
3	44%			56%(1)		
4	79%				21%	
5	46%			49%(2)		5%

(1) Incineration in moderated targets.
(2) Pu recycling only.

The scientific feasibility of plutonium management and waste transmutation have been established for the different cases [2,6]:

- With homogeneous recycling of Pu and MA in the EPR reactors (Case 1), in the case of the homogeneous multirecycling with a ^{235}U enriched fuel (MIX) and in the EFR reactors (Case 2) that allow additional transmutation of LLFP in moderated targets.
- In the EPR – EFR park (Case 3) with homogeneous recycling of Pu and Np, and transmutation of Am and Cm in targets placed in a moderated neutron spectrum in the fast core.
- In the double component hypothesis (Case 4), around 20% of dedicated systems are needed to ensure Pu and MA consumption.
- In the double strata (Case 5), around 50% of the first stratum are EFR reactors burning Pu, 5% of dedicated systems assuming MA transmutation.

In terms of reduction of the radio toxicity of the ultimate waste in the case of ingestion, the results are roughly of the same order for the different scenarios, depending on the elements considered, with a reduction factor of 3 to 10 for Pu consumption alone, and a reduction factor of 100 for Pu and Minor Actinides management, by comparison to the open cycle (reduction factor of one).

Nevertheless, the scenarios are not equivalent if one considers the amount of recycled masses, Scenario 1 leading for example to large amounts of Pu and MA and especially of curium which have to be taken into account when considering the technical feasibility and especially the impact on the cycle.

These results lead to consider the development of MA fuels for PWR (with multi-recycling of Pu), for fast reactors (with either mono-recycling of MA and LLFP in moderated targets or multi-recycling in quasi standard fuels) and also for dedicated reactors. The R&D programme is presented below according to these three items, after a review of the available results.

4. R&D programme on materials for transmutation

The aim of the R&D programme is to obtain elements of technical feasibility for the fuels to be used in the different strategies to contribute to the evaluation of the scenarios in 2006.

Aside the homogeneous recycling (in PWR and FR) which requires standard fuels with a low content in minor actinides, the heterogeneous recycling (in FR and ADS) leads to consider fuel with a high content of minor actinides but without uranium to prevent the formation of “new” actinides. This type of fuels can be either solid solutions of actinides or composite fuels with an actinide compound in a matrix support that must be as inert as possible towards neutrons to be stable under irradiation and able to reach very high fission rates up to 90%, far above the standard ones (see Table 2).

Table 2. **Objectives for transmutation in fast neutron spectrum**

	Targets	**Standard fuel in FR**
Composition	Inert matrix ~1-7g.cm^{-3} MA	(U, Pu)O_2
Fast fluence (n.m^{-2})	10-40 10^{26} (moderated or fast spectrum)	20 10^{26}
Linear power (W/cm)	Min.: 10, max: 400	400
Temperature range	500-2 000	2 200
Fission rate (%)	30% ->>90%	17.5%
Prod. helium (cm^3.g^{-1} of fissile phase)	36 (FR = 85%, AM~1g/cm^3)	0.15
Prod. fission gases (cm^3.g^{-1} of fissile phase)	20.6 (FR = 85%, AM~1g/cm^3)	3.6
Dose on the cladding in dpa	200	150

For these U free fuels, that represent a technological discontinuity with regards to U and Pu oxides, development is needed in different areas, first with the characterisation of the basic properties of the fuels components, and for the elaboration of the fabrication process, then with the realisation of experimental irradiations and post irradiation examinations to obtain elements of the behaviour under irradiation and also in term of simulation to prospect the behaviour of new concepts.

As the different phases require the use of shielded nuclear installations, the time needed to define a solution usable for the industrial level, will be around 15 years and more. For the specific case of fast neutron reactors, a first step will be reached before 2010 with the identification of the performance potential of the tested solutions. This will allow the definition of a second step for 5 to 10 years to reach the ultimate objectives fixed to the selected concepts.

This leads to privileged generic and basic research and, along with the present experimental programme, to develop the simulation of irradiated elements and fuels (including specific irradiation tools) and to share this development in international collaborations.

The programme detailed below covers a large fields of applications and is presented according to the technology considered with in first, the hypothesis of transmutation in PWR for which the R&D is to be connected to the projects under consideration to burn plutonium, and in second the actions linked to the use of fast neutrons reactors which offer determinate advantages for a transmutation strategy (ratios of fission to capture more favourable than in thermal flux, availability of neutrons) and have proven their capacity to use plutonium. Before the presentation of the experimental programme in FR, a status of the knowledge of the behaviour of the composite fuels is made, to point out the fields of research. In third the research for the fuels to be used in ADS is starting with work beginning on the characterisation of the elements of interest.

5. Fuels studies for transmutation in pressurised reactors

The solution considered in the calculations is the homogeneous mode on the basis of the MIX fuel [4], with a fuel composition of 2.7% plutonium, 0.3% americium, 0.4% curium and a uranium enrichment of 4.5%, in a standard UO_2 EPR fuel rod geometry.

Another option using a basis of standard fuel rod geometry and UO_2 fuel is the Corail concept [3] in which around 30% of MOX type rods are set with enriched UO_2 rods in a standard PWR assembly.

To prevent the formation of ^{239}Pu from ^{238}U captures, two other options are under investigations using an inert matrix in which the plutonium and actinides can be dispersed. This concept is investigated considering:

- Standard geometry for the rods that are of two kinds, one including UO_2 fuel and others containing composite fuel (Duplex concept).
- A modified geometry with annular rods and composite fuel.

This last concept is studied to develop an Advanced Plutonium Fuel Assembly (APA) [4] and recent developments [7] have shown the possibility to integrate actinides in the fuel. The objective of stabilising the plutonium inventory is reached assuming 29% of APA EPR, 36% APA EPR being needed to stabilise the plutonium inventory together with Am and Cm transmutation.

The R&D work is concentrated on the development of the appropriate concept for Pu consumption in PWR, the fuel to be developed for this purpose will have to take into account the possibility to burn also minor actinides. Boiling water reactors that constitute a growing part of the reactors in the world and that have the capacity to use plutonium will also be considered in order to assess their ability to recycle plutonium and to transmute the waste when compared to the EPR one.

The high temperature reactors characterised by high thermal efficiency and the capacity to offer inherent safety may be used to burn plutonium in a complementary way. Their possibilities are under investigation [5] and their contribution regarding the objectives of the fuel cycle will be assessed.

6. Fuel for transmutation in fast reactors – elements of behaviour under irradiation

In conventional reactors (PWR, FR) the addition of limited amounts of Am, Np, Cm in the standard fuel in the whole core (homogeneous mode) is not supposed to affect deeply the fuel behaviour. For the UPu type of fuel of fast reactors, the fuel behaviour is not too much affected by less than 2 wt% of minor actinide (MA) addition as was confirmed by the SuperFact [8] experiment in the fast reactor Phénix where (U, Pu, Np)O_2 and (U, Pu, Am)O_2 were successfully irradiated until a fission rate of 7% (32% transmutation rate).

The main problems are concentrated in the “heterogeneous recycling”, in which MA targets are loaded with a high content of actinides in some areas of the core. These so called targets are U-free fuels in order to reduce waste production and the support of the MA compound is a matrix such as a ceramic or a metal as inert as possible regarding neutron interaction.

6.1 Requirement and design of MA targets

The aim of such inert matrix fuels (IMF) is to be efficient for the transmutation and to allow a good level of safety in case of incident or accident the requirement for a transmutation strategy being fission rate up to 90% with a MA content of ~1-2 g cm^{-3} (low part of the range compared to the requirement of 7-8 g cm^{-3} for ADS fuels with fission rate around 30%).

The fissile atoms and the support matrix can either form a solid solution or be integrated in a composite fuel like ceramic inclusion in ceramic matrices (Cercer) or ceramic inclusions in metal (Cermet), the respective composition and concentration of the different parts of these composites

being adjusted to take into account the different effects induced by irradiation. The main requirements are good thermal and mechanical properties for the matrix and chemical stability in the course of its evolution for the actinide phase. Possible ceramic or metal candidate materials have been selected with criteria concerning their basic properties (thermal and mechanical properties, activation with neutrons, chemical compatibility with neighbouring materials,…) and their behaviour under irradiation.

The criteria prevailing for the selection have been initially considered on the basis of the available data [9] that concern essentially out of pile behaviour, data being rather scarce in the field of the behaviour under irradiation in the adequate neutron energies and fluxes.

Under irradiation in reactor, three main sources of damage have to be considered for the IMF: fast neutrons interaction, effects of fission fragments and alpha decay products (alpha particle + heavy recoil atom). These energetic particles produce damage through electronic and atomic interactions and the consequences, that depend on the material, may affect significantly bulk properties: changes of lattice parameter, phase changes, amorphization, swelling, evolution of thermal and mechanical properties.

Furthermore, MA fuels for transmutation will have a specific behaviour under irradiation when compared to standard fuels: the power evolution history will not be constant and will vary with a factor of 10 (or more), the total quantity of gases (fission gases + helium) will be higher of factors of some hundreds, and the maximum burn-up level to be reached is above 90% of the Fissile Initial Metal Atoms which is far above the usual levels of standard fuels (see Table 2 for comparison).

In order to design specific IMF concepts adapted to the objectives of transmutation, the R&D programme must cover first the basic materials qualification (properties, fabricability, behaviour under irradiation) and also the in pile test of the concept itself. For the material qualification under irradiation, the experiments are designed to study the different effects:

- Fast neutron fluence with experiment like Matina (see below).
- Fission products with ion irradiations in accelerators, this part must be developed after the first experiments on the spinel [10].
- Alpha interaction with helium implantation experiments started in the frame of the Efttra programme.
- Effect of fast neutrons and fission products in irradiations like Matina and Efttra T3, T4 ter.

Global experiments are then performed to test the in pile behaviour of the concept i.e. the evolution of the different elements and of the composite itself.

6.2 Irradiation results and qualification of the concepts

The main results in this field come from the irradiation programme that was conducted in the frame of the European collaboration Efttra (where three main field were identified, materials for transmutation, either matrices and actinides compounds, and test of target concepts) [11] and also from experiments performed in the Siloe and Phénix reactors. The main results are synthesised below.

6.2.1 Inert matrices

6.2.1.1 $MgAl_2O_4$

If spinel behaves very well under high fast neutron fluence [12,13] (more than 22 10^{26} $n.m^{-2}$), fission products recoil or alpha decays product severe damage in the matrix. The large programme on this material has given results for different irradiation conditions. The Efttra T4 (effect of alpha decay due to Am + fission) [14] and Tanox and Thermhet irradiations [15,16] (effects of fission only) illustrate the swelling and the modification of the material (Figures 1a and 1b). This quite unsatisfactory behaviour have not been observed in the case of fuels based on macromasses concept (Thermhet fuel with fissile inclusions of 100 to 300 µm in diameter, Figure 1c) and operating in a different temperature range (above 1 000°C) like Thermhet and Matina [17].

Due to the complexity of the different effects and of their respective interactions, behaviour of spinel under irradiation is not fully understood up to now. Nevertheless, the use of the spinel matrix for transmutation may still be a solution in the case of a concept of macro dispersed fuel tailored to take into account the irradiation damage and fuel swelling therefore operating at a sufficiently high temperature to favour defects recovery and gas diffusion.

Figure 1a. **Clad diameter change of Efttra T4**

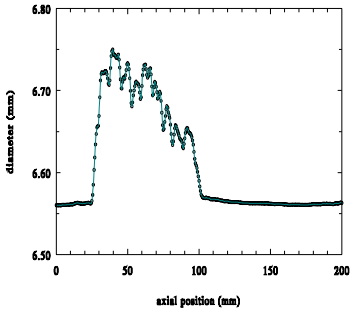


Figure 1b. **Micro dispersed fuel of Thermhet**

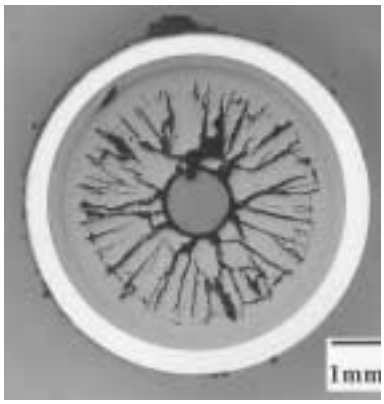
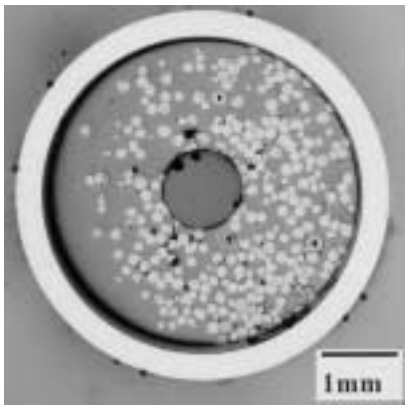


Figure 1c**. Macro masses of Thermhet**



6.2.1.2 MgO

The Matina experiment allowed also to test magnesia up to 1.4% FIMA and 2 10^{26} $n.m^{-2}$ [17] of fast fluence (effect of neutrons without fissions) and the pellets appearance was very closed to the fresh ones. This lead to consider MgO as a good candidate for fast reactors conditions. Its behaviour will be tested in the future Ecrix experiment in the micro dispersed form and also in the Camix experiment with macro dispersed americium oxide.

6.2.1.3 Al_2O_3

The average swelling of 1.9% in volume measured in the Efttra T2bis irradiation for only 0.46 10^{26} n/m^2 of fast neutron fluence and the one of 28% for a fast fluence of 17 10^{26} n/m^2 in Santenay in Phenix confirm the poor interest of alumina for transmutation since fast neutrons induce extensive dislocation-loop formation and swelling.

6.2.1.4 ZrO_2

Due to its low heat conductivity, the element was not identified at the start as a reference, but its fairly good structure stability under irradiation (in its stabilised form), makes it a possible candidate that will be tested in the Camix experiment (see below).

6.1.2.5 Other matrices, CeO_2, $Y_3Al_5O_{12}$, Y_2O_3, TiN, W, Nb, V, Cr

Most of these materials have been tested under neutron irradiation [11,15,17] but the examination being still underway, their potential as matrices remain to appreciate.

6.2.2 Actinide compounds

6.2.2.1 NpO_x

For the experiments planned in the Super Phénix reactor, pellets have been successfully fabricated under the form of standard fuel loaded with some 2 wt% of Np to test the homogeneous way and NpO_2 inclusions were dispersed in $MgAl_2O_4$, Al_2O_3 and MgO for the heterogeneous recycling. The decision to stop the Super Phenix operation brought the irradiation projects to an end in 97 and the new programme planned in Phénix have been concentrated on the main problems, the efforts being centred on americium compounds.

6.2.2.2 AmO_x

Americium oxide has a very complex phase diagram and the compound may show a high oxygen potential or a high chemical reactivity towards its environment (like the matrix in contact or other elements like sodium) [18], furthermore the thermal conductivity of AmO_x is very low. This simple oxide form has been considered in the first experiments (Efttra T4, Ecrix) and an alternative is now proposed with a solid solution of AmO_2 and ZrO_2.

6.2.2.3 (Am, Zr)O_x

Some zirconia based solid solutions present very attractive properties and analogy with $UZrO_2$ suggests a good behaviour towards radiation. $(Am_{0.5}, Zr_{0.5})O_2$ and the pyrochlore form, $(Am_2, Zr_2)O_7$, were both considered and characterised [19] in the Oak Ridge National Laboratory in the frame of the CEA/ORNL collaboration that may be extended to the same targets with curium in the place of americium. This type of compound will be tested in the Camix experiment.

6.2.3 Target concepts

The first targets were based on a concept of a micro dispersion of the actinide phase in the host matrix and a lot of experiments have been performed based on two types of matrices: MgO + AmO_x, $MgAl_2O_4$ + AmO_x.

Magnesia based targets have been fabricated for the Ecrix experiments in Phénix, and samples were used for properties measurements of the composite (thermal characteristics, oxygen potential, melting point, heat capacity, thermal expansion and diffusivity) as well as specific tests like compatibility with sodium in collaboration between ITU and CEA. The irradiation will give indication on the behaviour of the magnesia-based concept as a candidate for americium transmutation.

The first fabrication of this type of fuel was made by the impregnation process and then irradiated in the High Flux Reactor in Petten in two irradiations: Efttra T4 and T4bis [11,14] where fission rates

reached respectively 38.5% and 70% FIMA with a transmutation rate closed to 100%. This gives indication that technical feasibility of transmutation is possible but the pellet have swollen considerably, as a consequence of radiation damage and gases accumulation (Figure 1a).

Figure 2a. **Calculated effect of fission and alpha particles on the damaged volume of the matrix**

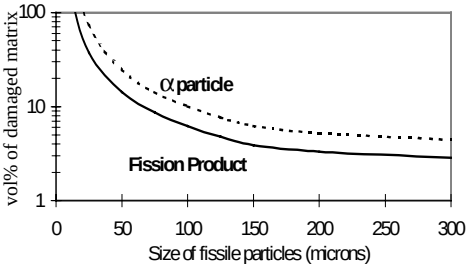


Figure 2b. **Macro particles of UO_2 dispersed in a spinel matrix**

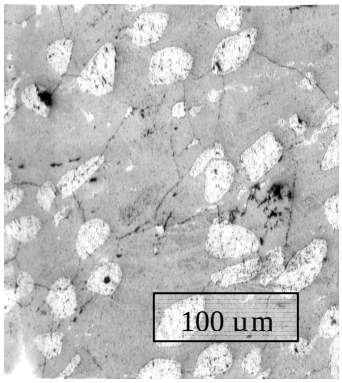
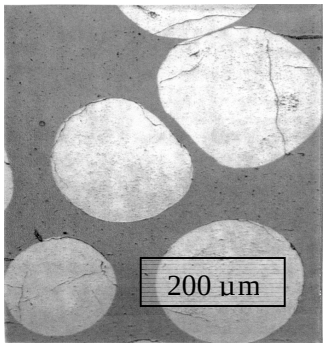


Figure 2c. **Macro particles of UO_2 dispersed in MgO**



These results and data of other experiments indicate that the target concept is to be improved to reach the ambitious objectives for transmutation scenario (fission rate >90%) namely in taking into account the radiation damage and gas production. Improvement of the dispersion is researched with the introduction of macro masses (100-300 µm diameter) [20] to concentrate radiation effects due to fission fragments or alpha decay in a small shell (Figure 2a). On a spinel-based matrix the product answers the requirements (puerility and size of the particles, homogeneity of the target, absence of cracks) as shown in Figure 2b, but the process is still under development for magnesia to obtain an acceptable composite (see cracks in Figure 2c) with other innovative options of the fissile particles.

The experiments Thermhet and Efttra T3 were the first tests of this concept that will be introduced in the Camix experiment.

6.3 Modelling

The irradiation behaviour of fuels for homogeneous recycling is simulated with the usual code Germinal used for standard fuel, completed with specific models for helium production or evolution of minor actinide isotopes. For targets, a heterogeneous modelling (see Figures 3a and 3b) is used with a finite elements code (Castem) to calculate thermal and mechanical behaviour taking into account the fissile-bearing phase and the matrix.

The experiments Thermhet [21,22] and Efttra T4ter [23] were calculated with a good agreement measurement/calculation.

Figure 3a. **3D idealised meshing of a pellet with a periodic distribution of inclusions**

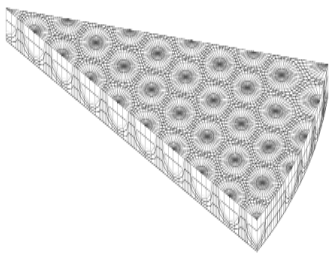


Figure 3b. **2D meshing deduced from a metallography**

6.4 Conclusion on the behaviour under irradiation

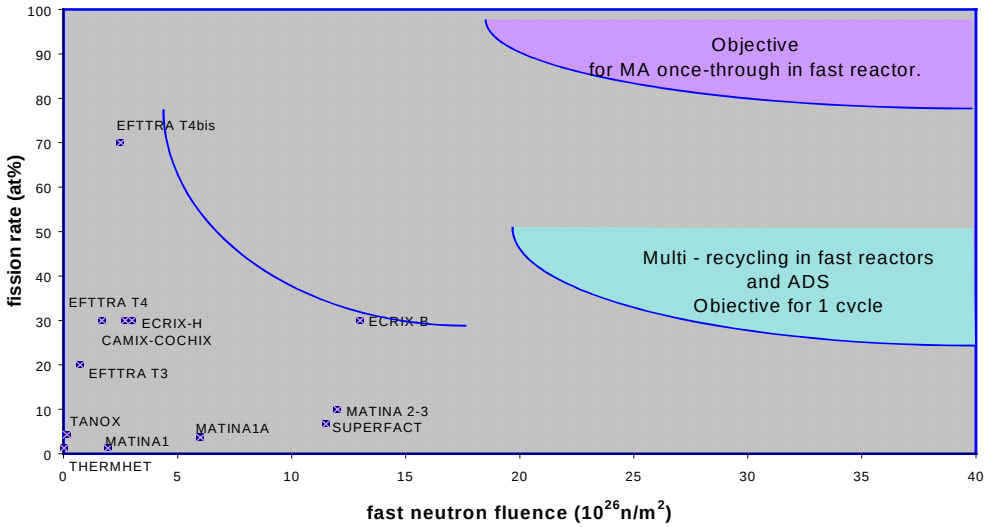
The work performed up to now, have brought numerous results in terms of fabrication of this new types of fuels, characterisation of the basic properties, behaviour under irradiation of the different components and of composite fuels.

With a start point based on simple chemical form of the fissile elements (AmO_2 particle) directly mixed with the matrix to obtain a micro dispersed composite, the different results obtained have lead to an optimisation of the concept with a greater complexity of the various parameters, the optimisation elements being [9,20]:

- The choice of a spherical host phase with size ranging between 50 to 300 µm in diameter.
- A MA compound in a stabilised phase of the type $AmZrYO_2$.
- A matrix material leading to a composite with acceptable thermal and mechanical characteristics and that can sustain a not too complex fabrication process. In this domain the possible choices rely on spinel, MgO and zirconia.
- The management of matrix damage and gas production, the later being still a matter of discussion the choice going from complete retention to complete release of fission gases. Calculations are still to be made to evaluate the possibilities of new ways of porosity repartition (porous matrix or “jingle” concept) and of coated particles.

In addition to the experimental programme described below, an important work remains to be done, in close relation with the reactor choice, in order to collect the necessary data. Figure 4 shows that after the present experimental phase will be completed and new designs are defined, a second phase will be necessary to reach the final objectives fixed to the fuels.

Figure 4. E**xperiments for MA transmutation**



7. Irradiation programme for fuel development in Phénix

The irradiation programme is designed to cover various objectives related to the behaviour under irradiation of the fuel rod, and more generally of the different materials entering in the concept (actinide or long-life fission product targets, moderators...):

- For MA transmutation in the homogeneous mode and considering the major acquired knowledge on the fuel of fast neutron reactors, no major problem is expected and the objective of the experiments is to evaluate the performance of the fast neutron reactor fuel in the presence of a low percentage of minor actinides.

- For MA in heterogeneous mode, the developments of actinide target-fuels is at its beginning. The objectives of the first experiments are to test the various possible solutions for the elements (inert matrix, americium compound...) and for the composite (dispersion fraction and mode of the americium composite...).

- Similarly, for the LLFP and the solid moderating materials selected in the incineration concepts, the behaviour under irradiation of the composites with the most interesting physical and chemical properties is to be studied.

7.1 Incineration of MA in the homogeneous mode

For the homogeneous mode, in addition to the experience already acquired with SuperFact [8] based on a fast neutron reactor standard $UPuO_2$ fuel, the performances of a metallic fuel with a low percentage of minor actinides (neptunium + americium + curium) and rare earth, 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-rod type rigs capsules with three experimental rods. The target burn-up are 2, 7 and 11 at%, respectively. The rods are at Phénix dimensions, cladded in AIM1 with a fuel constituted of a metallic alloy UPuZr (+minor actinides) and a sodium bond. Each rig will contain a reference rod (without minor actinides), a rod with a MA content of 2% and a rod with a MA content

of 5% [24]. The target irradiation conditions for the metallic fuel rods at the beginning of their irradiation are a maximum heat rating of 350 W/cm, a nominal TNG cladding temperature of ca. 580°C. The 3 capsules will be introduced in the reactor in the year 2001.

7.2 Incineration of MA in the heterogeneous mode

For the heterogeneous mode and to optimise the irradiation possibilities of Phenix, several aspects will be studied simultaneously:

- Damage of the matrices by irradiation separating the effects of neutron effects, fission products and alpha particles.
- Behaviour under irradiation of the various americium composites and/or of the composite and matrix arrangement.

7.2.1 Selection of the matrix

Matina 1 allows the irradiation up to 2 10^{26} $n.m^{-2}$ (fast flux) 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) with UO_2 micro inclusions to study the effects of fission products. Dismantled in 96 at the end of the 49th operating cycle, Matina 1 supplied 2 rods subjected to destructive examinations to provide elements for the selection of the matrix for the Ecrix H and B experiments.

The remaining rods were re-introduced in Phénix awaiting for the continuation of the irradiation Matina 1A up to 6 10^{26} $n.m^{2}$

In addition to Matina 1A, the experimental base on the reference matrix MgO will be broaden:

- On the one hand, by taking into account the microstructure of the composite as a parameter to be optimised (the “macrodispersed” concept).
- On the other hand, by irradiations up to more significant fast fluences.

Furthermore, the investigation field will be enlarge to include matrices that were not (or slightly) present in Matina 1.The matrices considered for this purpose are stabilised zirconia ZrO_2, tungsten as a metallic matrix, the manufacturing and characterisation work on these materials will possibly be shared with other R&D entities.

Both objectives are taken into account in the Matina 2-3 irradiation project i.e. a 19-rod rig with ca. a dozen experimental rods containing Cercer and Cermet composites planned to begin its irradiation in 2003 up to a fast fluence of ca. 10 10^{26} $n.m^{-2}$.

7.2.2 Selection of the americium composite – composite optimisation

The Ecrix programme includes two irradiations, 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 CaH_x for Ecrix H. The two experiments have required the development of two specific irradiation rigs planned to be available beginning of 2001 and specific calculations (Figures 5 and 6a) to take into account the flux modifications [25].

Figure 5a. **Ecrix B rig**

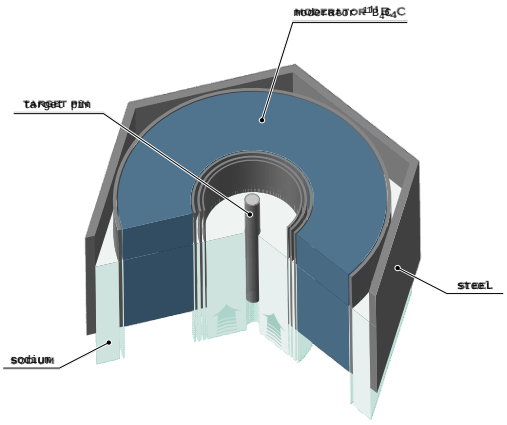
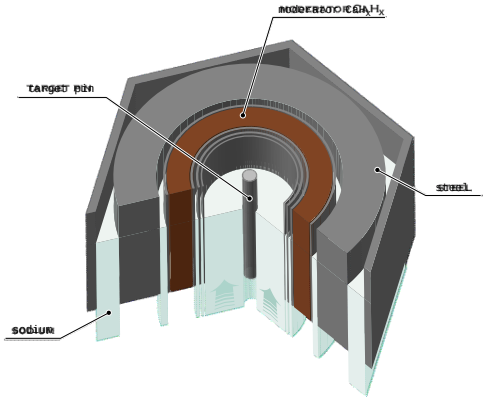


Figure 5b. **Ecrix H rig**



The fuel of the two Ecrix rods is a composite target with americium oxide “micro-dispersed” in the MgO matrix. The objective is to reach a fission rate of 30 at%. The required duration to reach this objective is 700 Effective Full Power Days for Ecrix B and 450 for Ecrix H with respective maximum linear power of ca. 50 and 70 W/cm without the uncertainty. The introduction in the reactor of the two experimental capsules is planned for the year 2001.

The manufacturing of the pellets has been completed in Atalante (Figure 6b) [26], and the two rods will be ready by end of 2000. 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 that goes beyond the sole objective of studying the behaviour of the target

Figure 6 a. **Evolution of the volumic power in Ecrix B and H (top)**

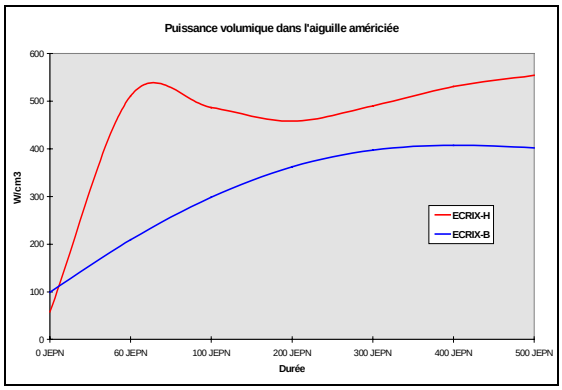
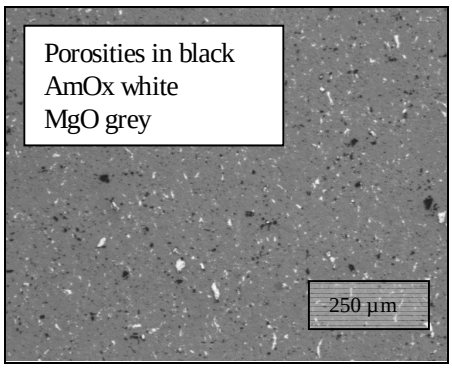


Figure6 b. **Micrograph of the Ecrix fuel**



To open the field of investigation, a further irradiation is planned to select the optimised composite and the americium targets. A research axis will concern the stabilisation of the americium oxide in a cubic structure thanks to the addition of a stabilising element with composites of the $(Am,Zr,Y)O_{2-x}$ type. The Camix irradiation (Composites of AMericium in PHÉNIX) will cover the optimisation of the actinide compound. 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 $ZrYO_2$ and MgO). Such a process applied to different matrices is to be operational

for the Cochix irradiation (optimised target in Phénix) that should start in end 2002 with the objective of a fission rate equivalent to that of Ecrix i.e. ~30 at%.

7.3 Incineration of long life fission products

In this domain, and after the first results obtained in the Efttra T1 (Tc,I) and T2 (Tc) experiments [11], studies are concentrated on iodine and technetium (transmutation of caesium requiring both chemical and isotopic separation, the reference strategy is direct disposal).

Two experiments are planned in Phénix. The first experiment Anticorp 1 includes 3 rods containing technetium 99 in the metallic form, to study the behaviour of the material under irradiation for a transmutation atomic rate of 15% corresponding to an irradiation of 350 EFPD in a CaH_x moderated device planned to start its irradiation beginning of 2002.

A second LLFP transmutation experiment focused on iodine transmutation is planned in 2003 using the natural isotope (the use of ^{129}I is still considered) to study the behaviour under irradiation of various possible iodine compounds (effect of the neutron damage and chemical evolution of the compound). This programme will be conducted in close partnership with NRG.

7.4 Experiment on moderators

The need of a locally moderated spectrum in fast neutron reactors leads to the design of two specific devices for the Phénix irradiations using the first moderators of acquired fabrication and so easily available $^{11}B_4C$ and CaH_x. Research on other moderators will continue in the Modix irradiation (MODerator in PHÉNIX) planned in 2002, to test hydride-moderating materials (CaHx and possibly YHx) up to a significant fast fluence of 10 10^{26} $n.m^{-2}$.

The temperature stability of the compounds is the subject of an experimental out of pile programme (studies of the dissociation temperature in different atmospheres).

7.5 Experiments linked to nuclear data

In order to improve the knowledge of the cross-sections of the various nuclei involved in the corresponding transmutation chains, under neutron flux conditions representative of the considered recycling mode (fast or locally moderated spectrum), two specific experiments will be launched each containing rods with a large number of minor actinide and long-life fission product isotopes separated into small quantities of several milligrams: Profil R in an otherwise standard sub assembly in the fast flux of the internal core and Profil M using a moderated $^{11}B_4C$ device.

8. Experiments in other reactors

CEA is involved in several R&D international collaborations aimed to develop technologies for transmutation.

First of all, the Efttra collaboration including CEA and other European organisations (ITU, NRG, EDF, IAM, FZK) working jointly on P&T issues. A valuable set of data has already been gained from the experiments performed in the HFR reactor of Petten [11]. Various inert matrices have been tested with and without inclusions of fissile material together with experiments on LLFP.

A CEA/Minatom work programme is underway in Russia. The Bora Bora irradiation in Bor60 is designed to test various fuel concepts developed in the frame of Capra Pu management programme with the test of pelleted PuO_2-MgO that will bring data on the behaviour of this composite. The Amboine project – in its initiating phase - deals with the feasibility of the Vipac concept for Am transmutation based on AmO_2 MgO. The irradiation of such a target is planed in a second phase.

The determination of fundamental proprieties of zirconium pyrochlore as the host phase for Am-Cm in the composite target is underway with the contribution of Oak-Ridge National Laboratory [19].

On the mid-term, new realisation in Japan, in the frame of bilateral collaboration agreements with JNC and JAERI are under discussion, exchanges being considered on oxide compounds for the homogeneous mode and also on the properties of nitrides.

A general view of the various components of the experimental programme is given in Table 3.

Table 3

Homogeneous dispersion of MA	Matrices behaviour, effect of neutrons	Effect of neutrons and FP, composite study	Effect of n + FP + alpha particles, Am compound and composite study	PFVL studies	General data
SuperFact; Trabant 1 pin 2 (oxide); **Metaphix 123** (metal)	Santenay, T2,2bis, Matina 1, **1A,** T3	Matina 1,**1A,** Tanox,T3,T4ter, Thermhet, **Bora Bora**	**T5, Ecrix B H, Camix Cochix, AmO2Vipac**	T1,T2bis, **Anticorp 1 2**	**Profil R M** (cross-sections) **Modix** (moderators)

Done, **under preparation**

9. Transmutation in dedicated systems

The impact of the fuel design on the transmutation in dedicated reactors has been largely investigated firstly in the frame of the European Capra Cadra project [2], then in the frame of the "Fuel and Fuel Processing (FFP)" sub-group of the European Technical Working Group on ADS [27].

Taking into account the innovative specifications of such "dedicated" fuels, a systematic analysis of the different actinide compounds have been done, in order to select the most promising candidates with the current knowledge. Presently, an R&D programme proposal is being submitted to the European Commission in the frame of the 5th framework programme. It aims at extending the basic properties knowledge, assessing the synthesis and reprocessing processes and optimising the design of such innovative fuels.

9.1 Specifications

Compared to conventional fuels (UOX or MOX), dedicated fuels are distinguished by:

- The absence of fertile uranium (U-free fuels or Pu-based fuels) to enhance the transmutation efficiency (no new actinide formation). In that case, inert matrices may be considered as support of the actinide phase in replacement of uranium as in the case of transmutation in fast reactors. Considered in a strategy of multi recycling and reprocessing, these fuels may include plutonium which can be interesting for the choice of the compounds and that lead to more stable irradiation conditions than in FR.

- Their high minor actinides (Np, Am, Cm) content (3 to 4 times the one of composite fuels for FR) with the ratio Pu/MA varying from 1:5 to 5:1. That leads to enhance the radioactivity level of the virgin fuel compared to conventional fuels and to request for the fabrication step remote handling and special protection to shield gamma and neutron radiation.
- High burn-up to decrease the fuel cycle cost, but less than in the case of transmutation in FR (30% against up to 90%). As for the "once through" fuels, dedicated fuels must accommodate a large fission gas and helium production and a high level of radiation damage. The impact on fuel design and materials choice is thus very significant but to the difference of FR, the reactor parameters are not fixed and that leads to some additional possibility in the choice.

9.2 Actinide compounds selection

According to the fuel specifications above, a classification of the different fuel types, from the less promising to the most one, is proposed taking into account the current knowledge on minor actinide compounds, which is unfortunately very sparse and poor.

- Metallic fuels, based on metal actinide alloys, are considered as the less interesting candidates. because of the low melting point of the major constituents (Np and Pu melts at 640°C), the expected limited mutual solubility of the actinides and the risk of stainless steel clad-fuel eutectic reaction at low temperature (410°C). Even if some improvements may be put forward such as a large Zr addition to enhance the fuel margins to the melting, considerable uncertainties remain on the actinides alloys metallurgy and on the fabrication processes (because of the high volatility of Am and Pu).
- Carbide fuels are known to have good thermal and mechanical properties and have shown in the past relatively good performance. However, the complex phase relations, especially between the sesqui- and monocarbides and the highly pyrophoric nature of these compounds make them less interesting than the other classes of refractory compounds (nitrides and oxides).
- Nitride fuels are attractive because of their expected good thermal properties and their ability to form solid solution whatever the minor actinide content. The major uncertainties concern the risk of dissociation at high temperature, which could be a critical issue in case of severe accident and the americium nitride vaporisation, which could complicate the fabrication step and limit the running temperature in pile. Large swelling under irradiation is also a specific feature of the nitride fuels, which should involve technical developments to accommodate it. If improvements to meet all the requirements can be considered like operation at low temperature, the development of such fuels must have to overcome the problems of temperature stability and also the technical and economical problem of the nitrogen 15 enrichment to avoid the ^{14}C formation.
- Oxide fuels are probably the most promising candidates since they offer a logical extension of the current MOX fuel technology. Although the thermal properties of actinide oxides are not so favourable compared to nitrides, they should be improved by using support matrices (oxide or metal) with good thermal properties. The experience developed in Europe on composite targets could be directly applied to dedicated fuels. Other engineering solutions (e.g. annular pellets, annular pins with internal cooling, specific coated particles) could be also developed. Finally, in spite of thermal weakness, improved oxide fuels should be considered not only as the "safest" solution if we take into account all the basic knowledge accumulated for 30 years on MOX fuels and the great synergy with other programmes, but also the best compromise if one consider the entire fuel cycle (in terms of fabrication and reprocessing command, reactor safety approach,…).

9.3 Programme for dedicated fuels

From this analysis based on the current knowledge on actinide compounds, nitrides and oxides are thought to be the most attractive support to the transuranium elements in ADS. 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.

The "Confirm" programme [28], which started in September 2000 in the frame of the 5th PCRD, is the first one devoted to the nitride fuels. The (Pu/Am, Zr)N compounds will be synthesised and characterised and (Pu, Zr)N will be irradiated in the Stüdvisk reactor. In parallel, modelling development is performed to predict the performance of such fuels.

The "Future" programme, which will be proposed to the European Commission in January 2001, will be focused on the oxide fuels. The (Pu, Am; (Zr))O_2 will be synthesised and characterised and composite fuels based on (Pu, Am; (Zr))O_2 will be studied. Modelling codes for the homogeneous and heterogeneous fuels will be developed.

Both programmes should allow by 2004 to collect information enough to judge the feasibility of such fuels and to influence the next R&D programmes.

10. Conclusion

The efforts made since several years for the development of the fuels necessary to insure a transmutation strategy lead to acquire knowledge on the basic data for the different materials needed, the fabricability of the composites, and the first elements about their behaviour under irradiation for MA and FP compounds.

The experimental programme planned in Phenix have been consolidated and the preparation work have reached marked milestones like the irradiation devices fabrication, the realisation of the first experimental pins with AmO_2 MgO composite, the various calculations and safety files necessary for the start of the experiments planned in 2001 (Metaphix 1,2,3; Ecrix B,H; Profil R). The first experimental results together with the knowledge acquired during the preparation of this first phase allow the definition of an optimisation of the targets that will be introduced in a second phase in 2003. Results from this programme together with those coming from other irradiations and of international programmes will be use to elaborate the first elements of technical feasibility of the fuels for transmutation in 2006.

Definition of fuels answering all the needs will require an additional research of long duration integrating the choices made in the scenarios taking into account external parameters such as cost, industrial feasibility, reactor park evolution and public acceptance. In this context, the necessity to cover a large domain of parameters, will lead to favour international co-operation and to privilege analytical experiments owing a better understanding of the phenomena together with some technological ones. The identified concepts will need to be tested in experimental reactors before considering an industrial phase.

If the necessary developments for an homogeneous transmutation using the present fuels as a start, appear available in around ten years time, the development of composite fuels in technological discontinuity when compared to the present fuels, needs continuous efforts on matrices, actinide compound and fuel conception. The development of dedicated fuels will require specific characterisation of the potential fuels adapted to the working parameters of ADS and taking into account the evolution of the strategy in this domain. The use of different cycles like molten salts must be appreciated in the course of the other actions made in these domains on a long-term schedule.

REFERENCES

[1] P. Bernard, B. Barré, N. Camarcat, M. Boidron, B. Boullis, J.M. Cavedon, D. Iracane. *Progress in R&D Relative to High Level and Long-lived Radioactive Wastes Management: Lines 1 (Partitioning-Transmutation) and 3 (Conditioning, Long-term Interim Storage) of the 1991 French Law,* Global'99, Jackson Hole (USA).

[2] A. Vasile, G. Vambenepe, J.C. Lefèvre, K. Hesketh, W. Mashek, C. De Raedt, D. Haas. *The Capra – Cadra Programme,* Proceedings of Icone 8, 8th Conference on nuclear engineering, April 2-6, 2000, Baltimore, USA.

[3] S. Aniel, M. Rohart, A. Puill, J. Bergeron, G. Rouvière. *Plutonium Multi-recycling in PWR: the Corail Concept*, Proceedings of Icone 8, 8th Conference on Nuclear Engineering, April 2-6, 2000, Baltimore, USA

[4] A. Puill and J. Bergeron. *Advanced Plutonium Fuel Assembly: an Advanced Concept for Using Plutonium in Pressurised Water Reactors,* Nuclear Technology, Vol. 119, Aug. 1997.

[5] X. Raepsaet, F. Damian, R. Lenain and M. Lecompte, *Fuel Cycle Performances in High Temperature Reactor,* Global'99, Jackson Hole (USA).

[6] J.P. Grouiller, J.L. Guillet, H. Boussier, J.L. Girotto, *Nuclear Materials Recycling in Conventional or Advanced Reactors: a Scenario Study*, Global'99, Jackson Hole, USA.

[7] H. Golfier, J. Bergeron, A. Puill, M. Rohart, *Multi-recycling of Plutonium and Incineration of Americium, Curium, and Technetium in PWR,* Safewaste 2000, Montpellier, France.

[8] N. Chauvin, J.F. Babelot. *Joint CEA/ITU Synthesis Report of the Experiment SuperFact 1,* Rapport EURATOM 1999.

[9] N. Chauvin, R.J.M. Konings, H.J. Matzke, *Optimisation of Inert Matrix Fuel Concepts for Americium Transmutation,* Journal of Nuclear Materials, 1999, 274,105-111.

[10] R. Fromknecht, J.P. Hiernaut, H.J. Matzke, T. Wiss, *He-ion Damage and He-release from Spinel $MgAl_2O_4$ Radiation Effect in Insulators*, Proc. Nucl. Instrum., Method in Phys. Research. Jena, July 18-23, 1999, Paper P 03.28.

[11] D. Haas *et al., The EFTTRA European Collaboration for the Development of Fuels and Targets for Transmutation: Status of Recent Development,* Conference Global'99.

[12] E.A.C. Neeft *et al., Neutron Irradiation of Polycrystalline Yttrium Aluminate Garnet, Magnesium Aluminate Spinel and A-alumina.* Journal of Nuclear Materials, 1999, 274, 78-83.

[13] K. Fukumoto *et al.,* Journal of Nuclear Materials, 32 (1995) 773-778.

[14] R.J.M. Konings *et al.*, *The EFTTRA T4 Experiment on Americium Transmutation*, Technical Report EURATOM 1999.

[15] A. Mocellin, Ph. Dehaudt, *Composite Fuels Behaviour Under and After Irradiation*, Technical Committee Meeting AIEA Moscow, 1-4 oct.1996.

[16] V. Georgenthum *et al.*, *Influence of the Microstructure for Inert Matrix Fuel Behaviour. Experimental Results and Calculation*, Global'99.

[17] N. Chauvin, T. Albiol *et al.*, *In-pile Studies of Inert Matrices with Emphasis on Magnesia and Magnesium Aluminate Spinel.* Journal of Nuclear Materials, 1999, 274, 91-97.

[18] S. Casalta, H.J. Matzke, C. Prunier, *A Thermodynamic Properties Study of the Americium-oxygen System*, Global'95.

[19] Ph. Raison, D. Haire, T. Albiol, *Materials Relevant for Transmutation of Americium and Experimental Studies on the Pseudo-ternary System, AmO_2-ZrO_2-Y_2O_3,* Journal of Nuclear Materials, to be published.

[20] N. Chauvin, R.J.M. Konings, H.J. Matzke, *Optimisation of Minor Actinide Fuels for Transmutation in Conventional Reactors (PWR, FR),* Conference Global'99.

[21] V. Georgenthum *et al.*, *Experimental Study and Modelling of the Thermo-elastic Behaviour of Composite Fuel in Reactor – Emphasis on Spinel Based Composites*, Paper accepted for Progress in Nuclear Energy.

[22] V. Georgenthum, CEA/EDF PhD thesis, Université de Poitiers 2000.

[23] R.J.M. Konings, K. Bakker *et al.*, Journal of Nuclear Materials, 1998, 254, 84-90.

[24] *Fabrication of UPuZr Metallic Fuel Containing Minor Actinide*, Global'97 Conference, October 5-10, 1997, Yokohama, Japan.

[25] J.C. Garnier, N. Schmidt *et al.*, *The ECRIX Experiments*, Global'99 Conference, 29 Aug.-3 Nov. 1999, Jakson Hole, USA.

[26] Y. Croixmarie *et al.*, *The Ecrix Experiment*, Global'99.

[27] M. Salvatores, *Transmutation: a Decade of Revival. Issues, Relevant Experiments and Perspectives*, 6th Information Exchange Meeting on Actinide and Fission Product Partitioning and Transmutation, Madrid, Spain, 11-13 Dec. 2000, EUR 19783 EN, OECD/NEA, Paris, France (2001).

[28] J. Wallenius *et al.*, 6th Information Exchange Meeting on Actinide and Fission Product Partitioning and Transmutation, Madrid, Spain, 11-13 Dec. 2000, EUR 19783 EN, OECD/NEA, Paris, France (2001).