THE FUTURIX-FTA EXPERIMENT IN PHÉNIX

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Abstract

In support to the European and American strategies of long-live radioactive waste transmutation in ADS or critical fast neutron reactors, an irradiation test of fuels with high contents of transuranium elements is being designed and is scheduled during the two last cycles of the Phénix fast reactor in 2007. This experiment will provide the main data concerning the behaviour under irradiation in representative conditions of different innovative fuels (oxides, nitrides, metal alloys, cermets) and will allow qualification and validation of models developed to predict their performance.

This item report focuses on recent progress.

Introduction

Europe and the USA are following similar R&D Partioning and Transmutation strategies to manage long-lived waste and especially minor actinides. Americium and Curium recycling is presently considered in dedicated reactors such as Accelerator Driven Systems (ADS) or in fast burners of the 4th generation.

This leads to specific fuel formulations, highly enriched in Minor Actinides. Fertile Uranium is excluded from ADS fuels to avoid the production of Plutonium ("non-fertile or U-free fuels"). In fast reactors, Uranium content is adjusted to achieve a conversion ratio of 0.25 or less and is therefore limited ("low-fertile fuels").

Very little information is available about the irradiation behaviour of such fuel compositions, except mixed oxide fuel $(U,Pu,Am,Np)O_2$ for which the SUPERFACT experiment provided comprehensive information. FUTURIX-FTA experiment objective is to compare in similar and representative conditions the behaviour of fuels proposed for TRU burning [1].

The FUTURIX-FTA program began in January 2003 and the irradiation will start at end of 2006. This progress report focuses on latest results.

Fuel selection

The fuel selection reflects the consensus reached on potential fuels for transmutation [1]. Eight experimental pins will be irradiated, their composition is summarized below:

Table 1. **Fuel Compositions**

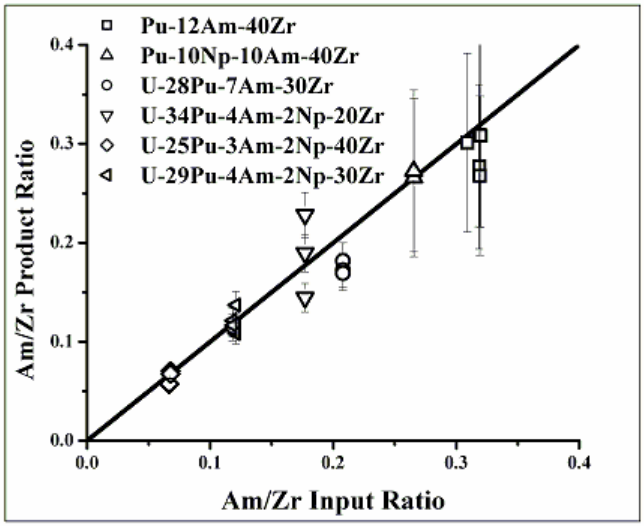
Experimental pin	**Fuel type**	**Composition**
DOE 1	Metallic low-fertile fuel	**35U 29Pu 4Am 2 Np 30Zr**
DOE 2	Metallic U-free fuel	**48Pu 12Am 40Zr**
DOE 3	Nitride low-fertile fuel	**$(U_{0.50},Pu_{0.25},Am_{0.15},Np_{0.10})N$**
DOE 4	Nitride U-free fuel	**$(Pu_{0.21}Am_{0.21}Zr_{0.58})N$**
ITU 5	Oxide U-free CERMET fuel	**$(Pu_{0.8},Am_{0.2})O_2$ + 60 vol% Mo**
ITU 6	Oxide U-free CERMET fuel	**$(Pu_{0.23},Am_{0.25},Zr_{0.52})O_2$ + 60 vol% Mo**
CEA 7	Oxide U-free CERCER fuel	**$(Pu_{0.5},Am_{0.5})O_2$ + 70 vol% MgO**
CEA 8	Oxide U-free CERCER fuel	**$(Pu_{0.2},Am_{0.8})O_2$ + 65 vol% MgO**

Fuel fabrication

Metallic fuels

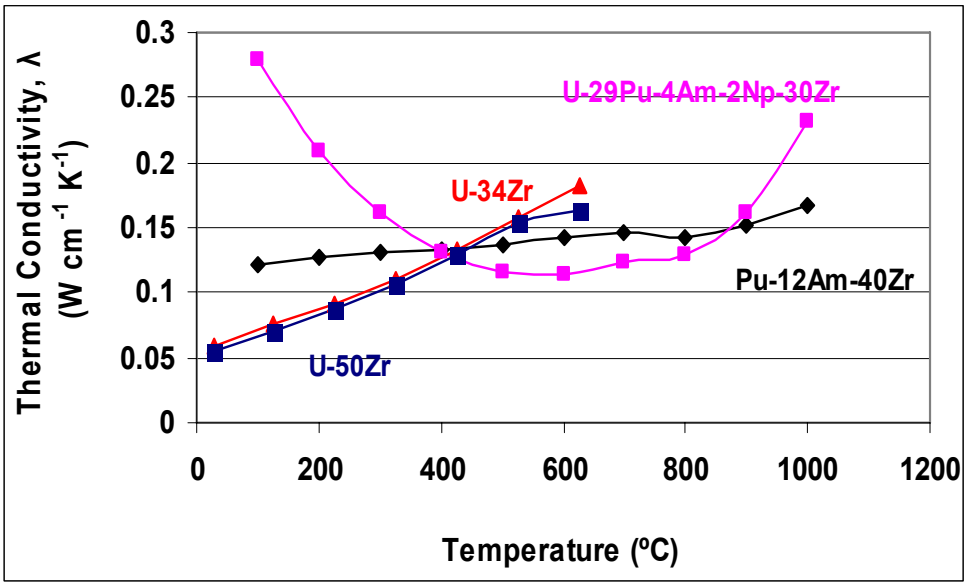
A specific fabrication process (modified arc melter earth) has been successfully developed to accommodate the high volatility of Am metal [2]. Am losses are negligible as shown below.

Figure 1. **Arc melter and Am retention results**



As-cast microstructure examinations show that fuel DOE 2 (PuAmZr) is monophased, with an even distribution of elements. DOE 1 is multiphased, homogeneous, with some precipitates enriched in Zr. Thermal properties are being measured and the first results are coherent with reported values.

Figure 2. **Measured thermal conductivity**



Fuel Cladding Chemical Interaction (FCCI) tests at 650 °C between fuels DOE 1 and DOE 2 and standard Phénix cladding (15-15 Ti ε) have been conducted. No interaction was observed, but a slight interdiffusion of Fe and Ni between DOE 2 (PuAmZr) and the cladding was observed after 7 h at 650 °C. The depth of this diffusion zone will be characterized.

Nitride fuels

The fabrication process is still under development. It is based on the preliminary synthesis of $(Pu,Am,Zr)O_2$ followed by a carbothermic nitridization into (Pu,Am,Zr)N. High volatily of AmN imposes a limit on temperatures. Sintering temperatures and times are being adjusted to reach the desired target density (about 85 % TD) and the level of impurities (for the moment about 5 % of the non-metal fuel is Carbon).

Oxide fuels

Although the CERCER and CERMET fuels are being manufactured by powder metallurgy, the fissile material $(Pu_xAm_{1-x})O_2$ (x=0.2 ; 0.8) has been prepared by a oxalic co-precipitation process. This wet route is a good alternative to minimize heavy contamination due to minor actinides especially if curium is concerned.

The oxalic coprecipitation of Am and Pu followed by the calcination of the precipitate in oxidizing atmosphere is the method adopted to obtain a solid solution of $(Pu_{1-x}Am_x)O_2$ type. The sintering of this mixed oxide in reducing atmosphere leads to $(Pu_{1-x}Am_x)O_{2-y}$.

$$x_1(Am^{3+},3NO_3^-) + x_2(Pu^{4+},4NO_3^-) + (3x_1 + 4x_2)/2\ H_2C_2O_4 + nH_2O \text{ ----->}$$

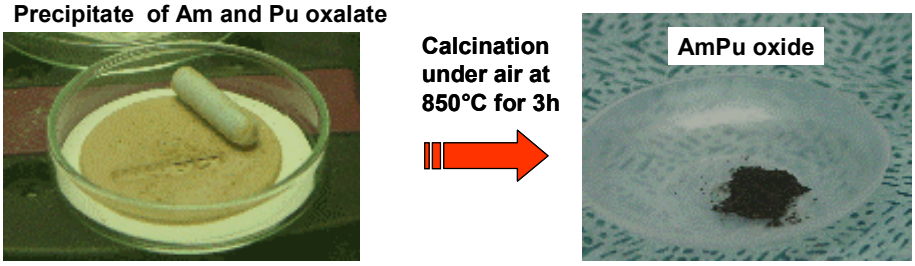
$$x_1Am(C_2O_4)_{3x1/2},n_1H_2O + x_2Pu(C_2O_4)_{4x2/2},n_2H_2O$$

The development of the operating conditions ($[HNO_3]$, $[H2C_2O_4]$) of $(Pu_{0.8}Am_{0.2})O_2$ synthesis was realized at the laboratory at the half gram scale (fig. 3)[3]. Analysis by X-rays diffraction shows the formation of a crystalline cubic structure with a cell parameter between AmO_2 and PuO_2.

After transposition of the experiment in shielded cell (continuous mode), two batches of approximately 25 grams of mixed oxide $(Pu_{0.5}Am_{0..5})O_2$ and $(Pu_{0.2}Am_{0..8})O_2$ were produced. The single-phase structure was controlled by an X-Ray analysis.

The particle size distribution covers a narrow range with over 99% of the grains smaller than 7 µm and the specific area value offer good performance for the pressing step [4].

Figure 3. **$(Pu,Am)0_2$ powder from Am and Pu oxalate**



After running a test with inactive materials to demonstrate the validity of the process, the fabrication of the CERCER has been undertaken (figures 4 and 5). The oxide is mixed with inert MgO, pressed and sintered at high temperature in a dedicated furnace set up in a shielded cell.

Goods results were obtained with few out-of-specification pellets. The homogeneous structure of the final product shown by optical microscopy (figure 6) meets the specifications.

Figure 5. **Fabrication process**

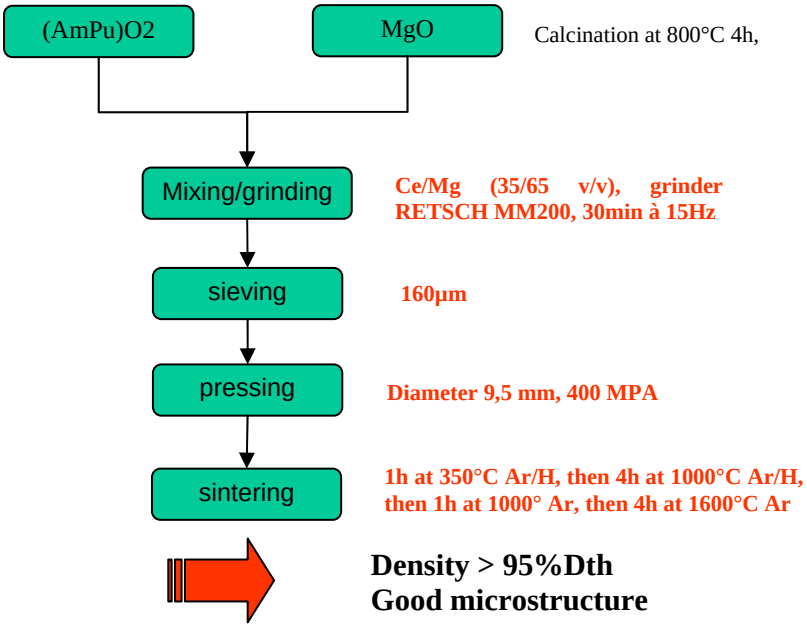
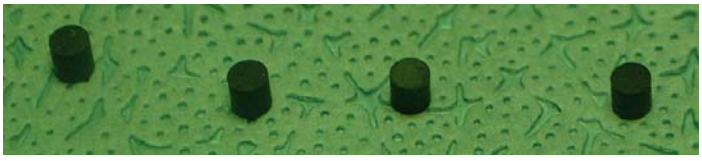
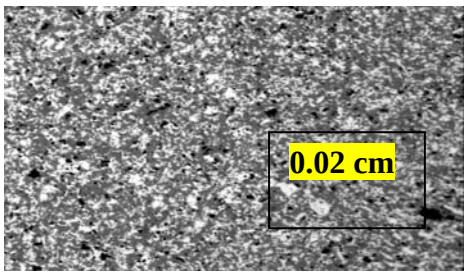


Figure 4. **High temperature oven in a shielded hot cell**

Figure 6. **Final Pellets of $(Am_{0,5}Pu_{0,5})O_2$ and with homogeneous structure**



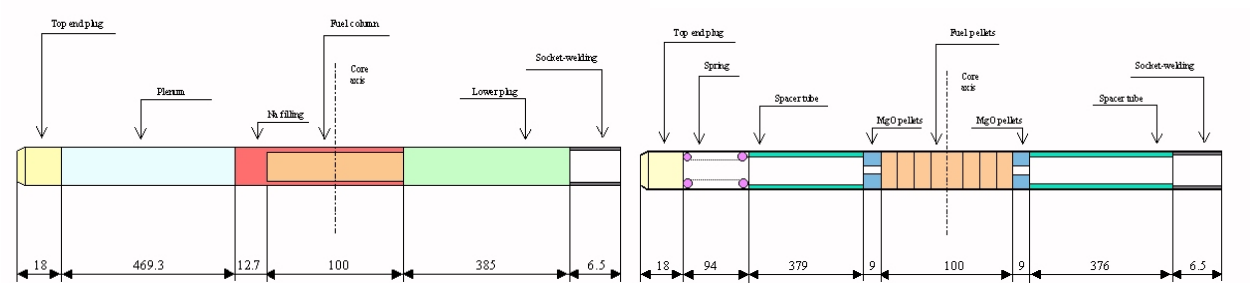


Irradiation design

Pin design

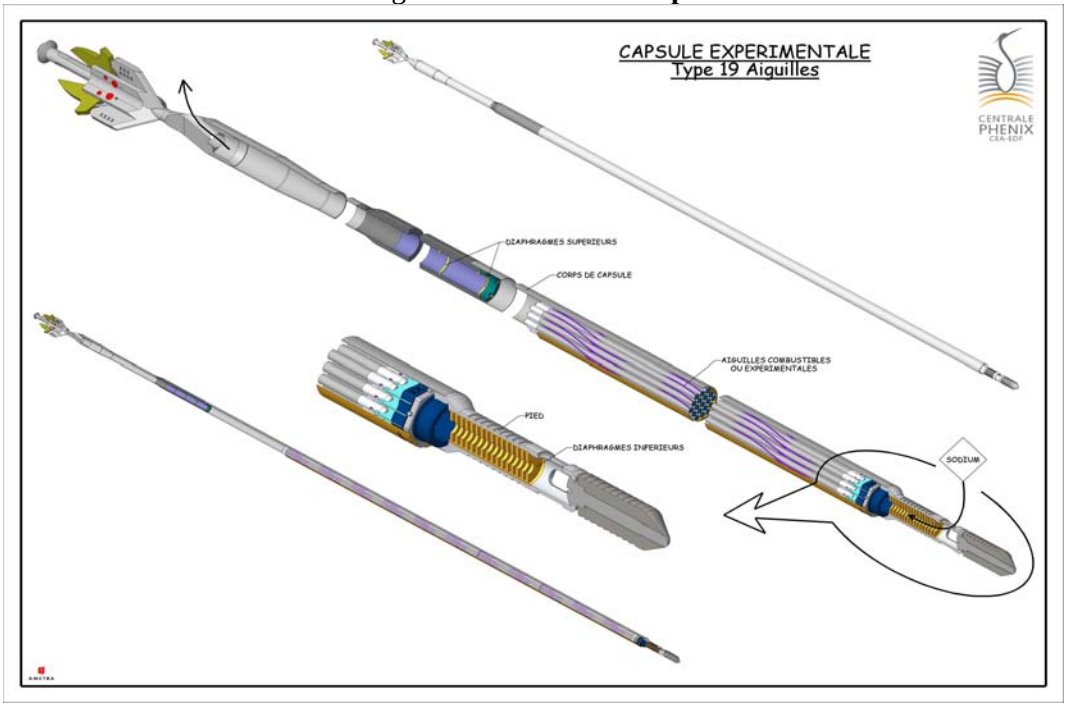
There is two different pins designs: metal and nitride fuels are sodium bonded, whereas oxide CERCER and CERMET fuels are helium-bonded.

Figure 7. **Pin design (sodium bonded and He bonded pins)**



The fuel column is 10 cm high. The eight experimental pins, along with either the standard oxide Phenix or “dummy” steel pins are inserted in an irradiation capsule (19 pins).

Figure 8. **Irradiation Capsule**



Irradiation conditions

The capsule will be inserted in the 4th ring of the reactor core. Linear power, burnup and transmutation rates are listed below.

Table 4. **Linear power, burnup and transmutation rates**

Experimental pin	Composition	Maximum linear power (W/cm)	Burnup after 240 EFPD (at %)	Am transmutation rate (at %)
DOE 1	**35U 29Pu 4Am 2 Np 30Zr**	275	7,0	20
DOE 2	**48Pu 12Am 40Zr**	320	9,0	17
DOE 3	**$(U_{0.50},Pu_{0.25},Am_{0.15},Np_{0.10})N$**	300	5,5	20
DOE 4	**$(Pu_{0.21}Am_{0.21}Zr_{0.58})N$**	260	10	20
ITU 5	**$(Pu_{0.8},Am_{0.2})O_2$ + 50 vol% Mo**	370	13	20
ITU 6	**$(Pu_{0.23},Am_{0.25},Zr_{0.52})O_2$ + 50 vol% Mo**	130	9,6	20
CEA 7	**$(Pu_{0.5},Am_{0.5})O_2$ + 70 vol% MgO**	160	9,2	20
CEA 8	**$(Pu_{0.2},Am_{0.8})O_2$ + 65 vol% MgO**	130	9,2	20

CERMET fuel ITU 5 has the highest linear power because of the Pu content and isotopic composition. On the contrary, CERCER fuels operate at low power. Power generally decreases with irradiation time when the Pu content is higher than the Am content. For CEA 8 fuel, power is maximal at end of irradiation due to Pu formation from Am neutron capture.

DOE 2 fuel has been moved (vertical offset to the core midplane) to limit the linear power to 320 W/cm, so as to achieve irradiation conditions comparable to the ATR Reactor (DOE Advanced Fuel Cycle Initiative).

Fuel behaviour and design

Preliminary design has been completed for DOE fuels 1 to 4. The sodium bond leads to low central temperatures in normal situations and accident scenarios, even with an initial large fuel/clad gap. Conservative assumptions have been made on initial fuel properties, when experimental data are scarce (most notably Am-bearing fuels).

For metal fuels, the limiting factor appears to be the temperature at fuel surface/cladding interface, during a postulated hypothetical accidental scenario of pump-diagrid rupture, where high sodium temperatures are reached during a long (~100 h) transient. For nitride fuels, margins to melting are high in all accident scenarios.

Conclusion

Preliminary design work will be completed in 2005 for CERCER and CERMET fuels, and final design will be completed in 2006 after the fabrication of the actual fuel pins. It will also benefit from the first results obtained in the ATR Reactor. The FUTURIX-FTA experiment will provide in 2008 important data on the behaviour of different innovative fuels for minor actinides transmutation.

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