THE PREPARATION OF THE EFTTRA-T5 AMERICIUM TRANSMUTATION EXPERIMENT

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Abstract

For the EFTTRA-T5 experiment, the irradiation of which is planned in the High Flux Reactor in Petten, Americium-containing zirconia-based inert matrix targets are envisaged, both with and without addition of Plutonium. The power of an Am-target (without Pu) is very low at BOI and slowly increases with irradiation time, because fissile nuclides are formed during irradiation. The addition of Plutonium increases the initial power of the target, such that a high fuel temperature can be obtained at Beginning Of Irradiation (BOI).

In this paper the technical preparations of the EFTTRA-T5 experiment will be explained. The calculations of temperature and power profile, leading to the definition of the actual composition of the EFTTRA-T5 targets are presented. In addition, the results of the fabrication tests for the targets are discussed.

Introduction

Americium is one of the radioactive elements that contributes to a large part of the radiotoxicity of spent fuels; transmutation of long-lived nuclides like ^{241}Am by irradiation in nuclear reactors is therefore an option for the reduction of the mass and toxicity of nuclear waste. The EFTTRA group is focussing on the development of transmutation targets for Americium, with emphasis on the fabrication aspects and irradiation behaviour. As became clear from the EFTTRA-T4 and T4bis irradiation experiments, which were made with targets of $MgAl_2O_4$+11wt% ^{241}Am, the release or trapping of helium is the key issue for target design. Significant volume swelling was observed in these experiments and was attributed to the production of helium which is characteristic for ^{241}Am transmutation. These findings led to the conclusion, that for the new EFTTRA T5 experiment helium should be released from the target. This release of helium may be achieved when the fuel temperature remains sufficiently high during the complete irradiation. Alternatively, release paths for helium can be created by inclusion of tailored open porosity in the target.

Since 1992 the EFTTRA (Experimental Feasibility of Targets for Transmutation) group is involved in the field of transmutation of waste compounds [1] in order to reduce the radiotoxicity of the long-lived radionuclides obtained after partitioning from the nuclear waste. Three radionuclides: technetium-99, iodine-129 and americium-241, have been selected for the EFTTRA research programme. In this paper the study on transmutation targets for americium is discussed. Two types of target can be considered for actinide transmutation:

- A solid solution target in which the actinide material is directly added in fresh fuel (e.g. MOX).
- A composite target, in which the actinide material is dispersed in an inert matrix. This matrix can be either a ceramic or a metallic material.

In the framework of the EFTTRA collaboration, the composite target has been investigated with respect to fabrication processes and behaviour of the targets under neutron irradiation. Concerning the specific case of americium, targets for transmutation consist of a ceramic matrix (commonly oxide ceramic) in which the minor actinide is homogeneously dispersed. This minor actinide can be either incorporated in the crystal lattice of the inert matrix material to form a solid solution, or can be made of americium compound particles (AmO_2 for example) heterogeneously mixed with the inert matrix material. In the latter case, the size of the particles of americium compound dispersed in the matrix can be very small (around 1 μm). In this case we speak of a “micro-dispersed” target. These targets are produced using a wet route, by infiltration of green pellets for example, [2] or a dry route, by a high energy mixing of the powders of inert matrix and americium compounds. [3] Another option consists of spherical shape particles with a large diameter (around 100 μm) that are embedded in the inert matrix material by a soft blending process. The “macro-dispersed” targets thus fabricated aim at minimising the damage volume caused by the fission product recoil in the inert host material.

The transmutation scheme of ^{241}Am is shown in Figure 1, from which can be remarked:

- Americium transmutation requires high neutron fluence for transmutation of all actinides present. The main fissile atom in the transmutation chain of ^{241}Am is ^{239}Pu. Three neutrons are required to induce fission of ^{239}Pu formed from ^{241}Am.
- ^{242}Cm emits a helium atom during decay. In the EFTTRA-T4 [2] and EFTTRA-T4-bis, [4] experiments (both containing AmO_x + $MgAl_2O_4$), it was observed that these fuels showed strong swelling during irradiation, which was attributed to the large helium production in these fuels. A large swelling of the pellets can cause unacceptably large mechanical interaction between the fuel and the cladding. Characteristics of the EFTTRA-T4 and EFTTRA-T4-bis experiment are shown in Table 1.

Figure 1. Transmutation scheme of ^{241}Am

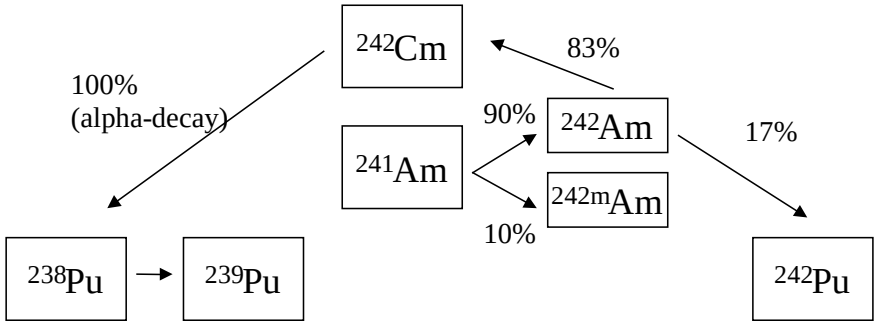


Table 1. **Characteristics of the EFTTRA T4 and T4 bis irradiation experiments in the HFR**

	T4	**T4 bis**
Full power days	358	653
Fast fluence E>0.1 MeV (m^{-2})	16.8 10^{25}	21.3 10^{25}
Actinide ratio EOI/BOI (%)	72.1	28.2
^{241}Am ratio EOI/BOI %)	3.7	<1
Min/max Fission power ($W.cm^{-3}$)	30/272	28/300

Objectives of the EFTTRA T5 experiment

The goal of the EFTTRA T5 experiment is the optimisation of the performances of Am-containing targets on the basis of results obtained with previous experiments carried out, such as T4 and T4bis. The main issue of the T5 experiment is to release helium from the target in an early stage of irradiation. Two different approaches are followed to reach early helium release:

- **Provide release paths** by creating open porosity i.e. release paths to the plenum gas. In the EFFTRA T5 test matrix a composite target with MgO matrix containing a network of open porosity is included (pin 1).
- **Increase target temperature** in order to promote the release of helium from the matrix. Americium or americium/plutonium zirconia based solid solutions (pins 2 and 3) are included in the test matrix to study the effect of temperature increase. The role of the plutonium in association with americium is to increase the temperature of the target at the beginning of irradiation.

In addition to the above-mentioned targets a Ceramic-Metal target (CerMet, pin 4) is included. Pin 4 will be a “cold” target. It serves as a comparison to the above-mentioned special design of the oxide inert matrices. The main characteristics of the EFTTRA T5 targets are described in Table 2.

The T5 irradiation experiment is planned in the high flux position C7 of the HFR, which has a thermal flux of about $1*10^{18}$ $m^{-2}s^{-1}$ and a total neutron flux of about $6*10^{18}$ $m^{-2}s^{-1}$. The use of a high flux position is required in order to transmute a substantial fraction of the ^{241}Am, within the duration of the HFR irradiation. Table 2 shows the proposed test matrix for the T5 irradiation.

Table 2. **Test matrix for the EFTTRA T5 experiment**

Pin no.	**Composition**	**Micro structure**	**Density** (g/cm^3)		**Gap** (μm)	**Boundary condition**
			Am	Pu		
1	$MgO - Am_2Zr_2O_7$	Composite 10-50 μm	0.7	0	*tbd*	Central temp. maximum 1 000°C
2	$(Am, Zr, Y)O_{2-x}$	Solid – solution	0.7	0	100	
3	$(Am, Pu, Zr, Y)O_{2-x}$	Solid – solution	0.7	*tbd*	100	Central temp. 1 700°C at BOI
4	$(Am, Zr, Y)O_{2-x}$ + metal	Composite	*tbd*	0	100	Central temp. maximum 700°C

The remark *tbd* (*to be determined)* in Table 2 means that this parameter (Am, Pu density or gap size) should be adjusted such that the boundary condition is met. The test matrix contains both solid solution (pin 2, 3) and composite compounds (pin 1, 4). In the composite targets MgO (pin 1) and stainless steel or molybdenum (pin 4) are chosen as a matrix since these materials are known to have a reasonable radiation resistance and a rather high thermal conductivity. The design of pin 1 and pin 4 is such that the effect of the difference in fuel temperature on the fuel behaviour and the helium release can be studied. For both pin 2 and pin 3 $(Zr,Y)O_2$-based compounds are used, since these have a good radiation resistance and do not interact with water. The plutonium concentration in pin 3 will be chosen such that the fuel temperature of pin 3 is about 1 700°C at Begin Of Irradiation (BOI). Due to the absence of plutonium in pin 2, the fuel temperature of pin 2 will be much lower than that of pin 3. This large temperature difference will allow studying in detail the impact of temperature on helium release.

The cladding of the pins will be 15-15Ti steel with an inner diameter of 5.65 mm and an outer diameter of 6.55 mm. The fuel pins will be filled with one bar helium gas. During irradiation the pins will be contained in stagnant liquid sodium with a temperature of 300-500°C. The fuel cladding and the fuel central temperature will be measured with thermocouples.

The total americium concentration in the pins 1-3 will be identical (0.7 g/cm^3) in order to enable a comparison of the various fuel concepts. The americium concentration of pin 4 will be chosen such that central temperature reaches a maximum of 700°C. The main reasons of the choice and design of the new targets are summarised below.

Target 1: the composition of this target is close to the EFTTRA T4 and T4 bis targets. The spinel matrix is replaced by magnesia (MgO) because of its high resistance to irradiation damage and amorphisation. Moreover, tailored porosity will be included in the MgO matrix, to provide channels for helium release and accommodate the swelling. The maximal temperature of the target will be set to ~1 000°C by adjusting the initial size of the gap between target and cladding. This relatively high temperature is chosen on the basis of two criteria:

- The possibility of annealing of irradiation damage in target.
- The release of helium outside the americium compound particles.

The use of a pyrochlore oxide, $Am_2Zr_2O_7$, as support for minor actinide, results from conclusions of recent studies performed jointly by the Oak Ridge National Laboratory in USA and the CEA-Cadarache centre in France. [5] These conclusions point out that the pyrochlore structure is expected to be quite stable under irradiation. The pyrochlore phase is a fluorite type structure with a double unit

cell, and ordered deficiency of oxygen atoms. In addition, the pyrochlore phase can be fabricated using a dry process with standard devices commonly used in powder metallurgy. Furthermore, given the trivalent state of americium, the pyrochlore structure is expected to have low oxygen potential, minimising the chemical reactivity with the matrix and the cladding materials. The diameter of $Am_2Zr_2O_7$ particles will be in the range between 10 and 50 µm. Smaller sizes would give rise to chemical interaction between MgO and pyrochlore particles, whereas larger sizes are favourable to cracks in MgO matrix during fabrication.

Target 2: The cubic (Am, Zr, Y)O_{2-x} phase is known to present a very high stability under irradiation. This target no.2 is to be directly compared with target no.3. The only difference between the two targets is the presence of Pu in Target no.3, and thus the temperature. The Yttria-stabilised Zirconia (YSZ) is a refractory ceramic with a high melting point that allows high operating temperature. The zirconia-based materials are known to have a low thermal conductivity. Besides, the americium insert in the YSZ crystal lattice can play the role of stabilising agent of the cubic phase, as yttrium.

Target 3: This target has the same composition as Target No.2 with a complementary addition of plutonium in the solid solution: (Am, Pu, Zr, Y)O_{2-x}. The plutonium is added to increase the initial power of the target, such that a high temperature can be obtained at beginning of irradiation. With such a design the formation and accumulation of helium bubbles can be avoided at an early stage, which improves the behaviour of the target.

Target 4: The use of a metallic matrix, such as stainless steel or Molybdenum, will increase strongly the thermal conductivity of the target. Compared to the CerCer concept, the operating temperature of such a CerMet is expected to be low. This property allows the insertion of a larger amount of actinide in the target, without increasing dramatically the temperature of the target. In addition, it is known that the metal matrices show good fission gas retention. Whether or not this has implications for the swelling is a subject of investigation.

Fabrication of targets

Target 1

A preliminary phase of R & D has been put in place by CEA, in order to fit the fabrication process of this target. In this way, two tasks have been discerned:

- The first concerns the fabrication of the MgO matrix with internal tailored porosity. The University Laboratory “Ecole des Mines de Saint Etienne” (EMSE) and CEA Cadarache, has been involved in this duty.
- The second is related to the pyrochlore compound synthesis by CEA Cadarache.

In order to fabricate a dense MgO matrix with tailored porosity, two processes have been explored in a non-nuclear facility of EMSE:

- By sintering MgO green pellets at high temperature and under pressure of nitrogen. A suitable microstructure is obtained by this process, with porosity mainly located at the grain boundaries, as can be seen in Figure 2.

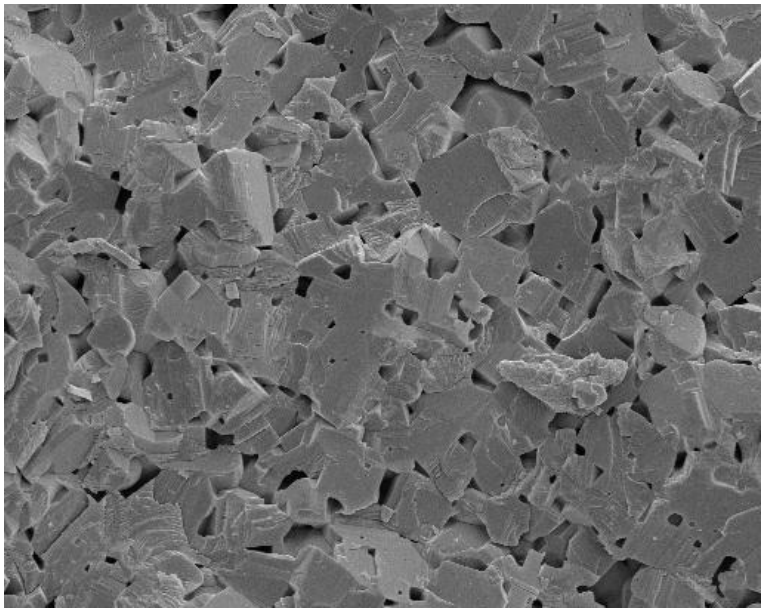
- A double pressing of MgO powder before sintering at high temperature. A large network of open porosity can be obtained by this method.

The characterisation indicates an open porosity close to 50% of the total porosity in the pellets after sintering. Re-sintering tests performed at 1 600°C show a high stability of the porosity.

With regard to the pyrochlore compound synthesis, fabrication tests have been performed with CeO_2 and ZrO_2 powders, to form $Ce_2Zr_2O_7$. A pure pyrochlore structure has been fabricated using a dry route process, by mixing the powders and heating at high temperature under reducing conditions. After this step, the same process has been used successfully to synthesize $Pu_2Zr_2O_7$ and $(Pu,Ce)_2Zr_2O_7$ pyrochlore compounds. X-ray diffraction examination shows that these pyrochlore compounds reveal no secondary phases after synthesis.

The next step will concern the fabrication of composite targets: MgO (porous) + $Am_2Zr_2O_7$ in the ATALANTE facility during 2003.

Figure 2. **Microstructure of MgO pellet after sintering under pressure of nitrogen**



Targets 2 and 3

Targets 2, 3 and 4 will be prepared in the Minor Actinide laboratory of ITU. For targets 2 and 3 the infiltration method will be used. [6] Polydisperse yttria-stabilised zirconium oxide (YSZ) spheres, produced by sol-gel technique using a rotating cup atomiser, will be infiltrated with actinide solution to achieve the required actinide content. The resulting beads are then compacted into pellets using a biaxial press, and sintered in Argon. Pre-tests with cerium as stand in for americium, and with plutonium [6,7] have shown that this process is feasible for the actinide concentrations of T5. Pellets with densities of around 90% of the theoretical one can be obtained, which are single phased.

Target 4

For target 4, nearly monodisperse YSZ spheres, fabricated in the same way and selected by sieving the 80-100 µm fraction, are infiltrated in multiple steps. The powder is then mixed with a metal matrix and blended. Next, the mixture is compacted to pellets with a bi-axial press and sintered in Ar. Pretests, again with cerium and plutonium, have shown that the process is feasible and gives pellets with a very good homogeneous distribution of the spheres in the matrix. [8] In case of cerium, the ceramic phase is a mixture of the pyrochlore and fluorite phases, which will probably also be the case of americium. The visual aspect and density of the pellets are good as long as the fraction of the ceramic phase is smaller than 40 vol%.

Neutronics aspects

In order to make a preliminary design of the T5 experiment, the Pu and Am power are estimated (Figure 3), using the results from detailed computations on previous HFR irradiation experiments EFTTRA-T4-bis [4] and OTTO. [9]

Figure 3. **Estimated power for different Am and fissile plutonium (Pufis) concentrations** (the irradiation position is C7 and the pellet diameter is 5.45 mm)

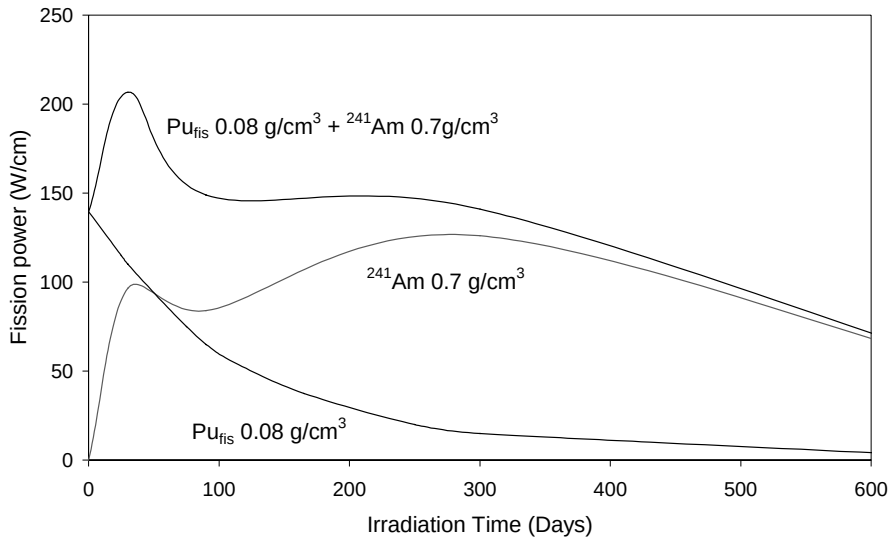


Figure 3 shows that ^{241}Am do not generate power at the start of the irradiation. The first maximum in the power generated in americium is due to fission of ^{242m}Am, while the second maximum is due to fission of ^{239}Pu. The power in the fissile plutonium decreases rapidly, while that in the americium remains relatively constant for a long period, reflecting the slow burn-up of the actinides present in the americium pin. From the computations can be concluded that:

- The maximum power generated per gram of Pu_{fis} is much higher (about 10 times) than that per gram of ^{241}Am.
- The power decrease and burn-up of Pu_{fis} are much faster than that of ^{241}Am.
- Fuel containing only 0.7 g Am/cm^3 and no plutonium generates low power (maximum power <150W/cm.

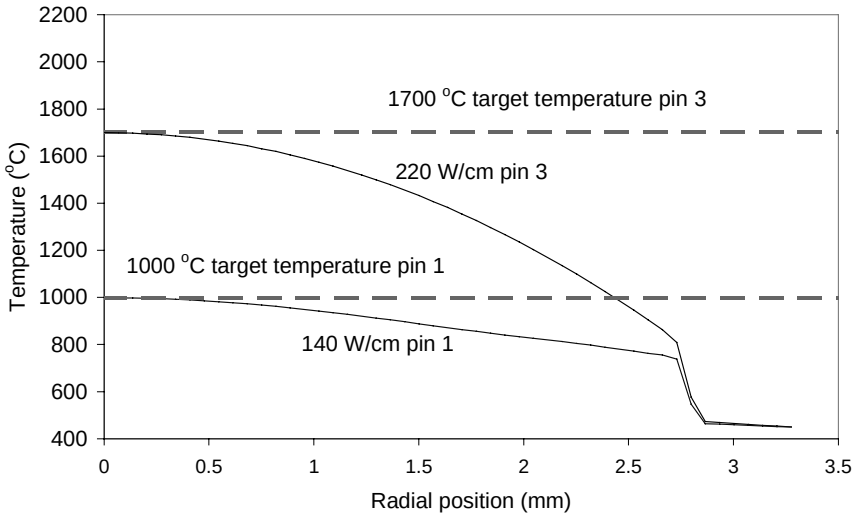
- The combination of ^{241}Am (0.7 g/cm^3) and Pu_{fis} (0.08 g/cm^3) shows a more or less gradual power decrease, which is typical for irradiation under FBR and LWR conditions.

Thermal aspects

An axi-symmetrical Finite Element Method (FEM) temperature model has been used for these exploratory computations. Effects such as pellet fracture, pellet relocation and radial power distribution have been ignored. The thermal computations are only done for the start of the irradiation, since it is impossible to give an accurate prediction of the temperature during irradiation due to the absence of data on properties such as swelling and gas release. The radial temperature profile of the pins 1 and 3 are shown from the centre of the pellet to the outside of the cladding.

These thermal computations show that the cladding temperature of the experiment is about 450°C and the outside temperature of the pellets is about 800°C, depending on the fuel power. Figure 4 shows that pin 3 requires 220 W/cm to reach a central temperature of 1 700°C. Figure 3 shows that a maximum power of 220 W/cm requires 0.7g/cm^3 ^{241}Am and about 0.08 g/cm^3 Pu_{fis}, while a maximum power of 140 W/cm can be reached using only 0.7g/cm^3 ^{241}Am.

Figure 4. **Radial temperature distribution in pin 1 (140W/cm) and pin 3 (220W/cm)**



During the course of the irradiation the following phenomena are expected to occur:

- Swelling of the fuel pellets, which decreases the pellet-cladding gap.
- Helium release into the plenum.
- Degradation of the thermal conductivity of the pellets, due to the presence of fission products and additional porosity.

The combined effect of these phenomena will probably be that the temperature gradient over the pellet increases, while the temperature gradient over the gas gap decreases during irradiation.

Several studies have been performed on the helium release from inert matrices using ion implantation. [10,11] For $MgAl_2O_4$ it has been found that helium release starts at about 500°C and is

nearly complete at 1 300°C. [10] For $Zr_{0.9}Y_{0.1}O_{1.95}$ similar behaviour and a similar temperature range are observed. [11] Figure 4 shows for pin 3 that the pellet central temperature of 1 700°C is clearly above the helium release temperature. The pellet outside temperature of pin 3 is about 800°C and is expected to decrease below 800°C during irradiation. Completely different helium release behaviour is expected for pin 2, which contains no plutonium, and therefore has a lower BOI temperature.

In order to study the consequences of a temperature difference in pin 2 and pin 3, the temperature of pin 2 should remain below the temperature of pin 3 during the complete irradiation. The power versus time curves in Figure 3 show that after 150-200 days of irradiation the power of pin 2 and pin 3 become similar and that therefore the temperatures are expected to become similar, which would complicate the observation of the temperature induced helium release. Due to this the irradiation will be stopped after 150-200 days.

From Table 1 it is concluded that in order to achieve a significant burn-up of the ^{241}Am and the actinides formed from the ^{241}Am a much longer irradiation period (up to 650 full power days) is required in a high flux position, which is confirmed by the estimates reflected in Figure 3. The goal of the present experiment is the study of the volume swelling, which is known to occur already at moderate burn-up values, rather than acquiring a very high burn-up.

Schedule of the experiment

The fabrication of the targets (CEA and ITU) and the design of the irradiation (JRC-IE and NRG) are expected to start in 2003. The irradiation in the High Flux Reactor is planned for 2004. The experiment will be proposed for the 6th FWP of the EU.

Conclusions

One of the main problems in the transmutation of americium is the swelling of the fuel pellets induced by the helium produced during irradiation. The EFTTRA-T5 experiment studies the possible enhancement of the helium release from the fuel pellets by increasing the irradiation temperature or by open porosity. Such an enhanced helium release is expected to limit the fuel swelling. Four pin concepts have been discussed that could be used to study the fuel swelling. Preliminary neutronics and thermal computations suggest that power and temperature variations during the complete EFTTRA-T5 irradiation are rather large. The design parameters (Pu content, porosity, gap width etc.) are chosen such that distinct differences for the Helium release behaviour, and consequently the swelling behaviour, is to be expected for the different targets. This EFTTRA T5 experiment should result in a better understanding and an optimised design of Americium-containing inert matrix targets.

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