TRANSMUTATION OF AMERICIUM AND TECHNETIUM: RECENT RESULTS OF EFTTRA

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Abstract

The results of irradiation experiments of the transmutation of technetium and americium, performed by the EFTTRA group, are described. First the results of the recent tests, T1 and T2, for technetium in the HFR are discussed. In two subsequent experiments targets of metallic technetium were transmuted to an extent of about 6% and 16%. Post-irradiation examination of the targets revealed an excellent in-pile behaviour. Secondly, the results of the T4 test, the transmutation of americium in a spinel-based fuel (micro-dispersed), are presented. It is shown that extensive swelling of the fuel took place, probably due to the damage of the matrix by fission product and/or accumulation of helium as a result of alpha decay. A more promising concept for an inert matrix fuel for transmutation of americium, based on a macrodispersion of a host phase in the inert matrix is discussed.

Introduction

EFTTRA, which is the acronym for Experimental Feasibility of Targets for Transmutation, is a network of research organisations in France, Germany and the Netherlands that was formed in 1992 [1]. The goal of EFTTRA is the study of transmutation of americium as well as of the long-lived fission products technetium (^{99}Tc) and iodine (^{129}I). The work of the partners of the EFTTRA group is focused on the development and testing of targets and fuels, taking into account the scenario's developed in Europe for possible P&T strategies. To that purpose fabrication routes are being investigated, irradiation tests are performed, and post-irradiation examinations are made in the various facilities of the EFTTRA partners. In Table 1 an overview is given of the EFTTRA irradiation experiments that are completed or ongoing at present.

In the present paper the results of the EFTTRA irradiations test on the transmutation of technetium and americium performed in the High Flux Reactor at Petten are described. A more detailed version of this paper has recently been presented at the OECD/NEA Workshop on Advanced Reactors with Innovative Fuels [3].

Table 1. **EFTTRA irradiation experiments**

Name[a]	Reactor	Description	State of the art	Refs
T1	HFR	Technetium and iodine	completed	3-6
T2	HFR	Technetium	completed	7
		Neutron damage in inert matrices	completed	8
T2bis	HFR	Neutron damage in inert matrices	completed	
T3	HFR	Neutron damage in inert matrices	PIE to be started	
		Dispersion inert matrix fuel using enriched UO_2	PIE to be started	
T4	HFR	Americium in spinel	PIE ongoing	9,10
T4bis	HFR	Americium in spinel	irradiation ongoing	
T4ter	HFR	Central fuel temperature of spinel/UO_2	irradiation ongoing	11
F1	Phénix	Neutron damage in inert matrices	continued	12
		Dispersion inert matrix fuel using enriched UO_2	PIE ongoing	12
F1A	Phénix	Neutron damage in inert matrices	irradiation ongoing	
		Dispersion inert matrix fuel using enriched UO_2	irradiation ongoing	
F2	Phénix	Technetium and iodine	start pending	

[a] Some of the experiments are also known under different names such as RAS or MATINA.

Transmutation of Technetium

The study of the transmutation of the technetium was started in 1992 when irradiation tests of targets of ^{99}Tc metal were planned for the High Flux Reactor (HFR) at Petten as well as the Phénix fast reactor at Cadarache (see Table 1). The fabrication of the targets for these tests was studied at ITU where a technique was developed for the fabrication of cylinders/pellets by casting of liquid technetium metal in a water-cooled copper mould. The targets that were fabricated for the irradiation tests each contained two of such cylinders (4.8 mm diameter, 25 mm length).

The results of the T1 test, which was completed in 1995, have been reported extensively [3-6]. They showed good behaviour of the technetium metal at an extent of transmutation of about 6%. To study the behaviour at an higher extent of transmutation, one of the targets (2 cylinders) of the T1 test was re-encapsulated and irradiated again in the HFR. The T2 test has been completed successfully in 1997 and the results of the post-irradiation examinations have become available recently and will be summarised shortly below.

Figure 1 **Microscopic cross section of irradiated Tc target from the T2 experiment showing oriented crystal growth (diameter 4.8 mm)**

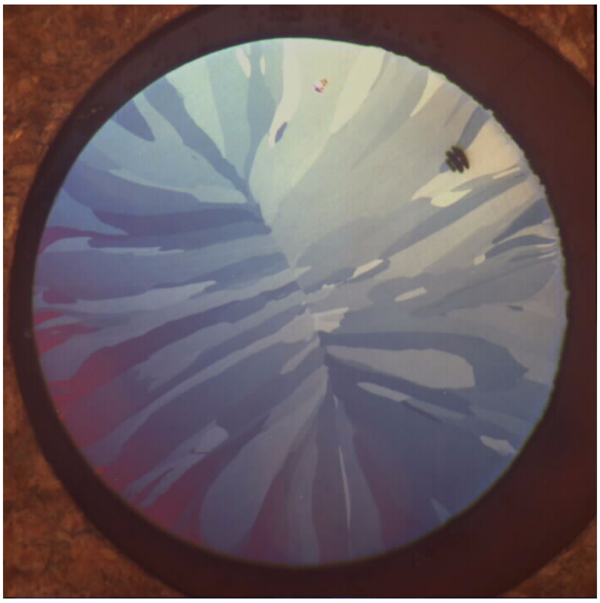
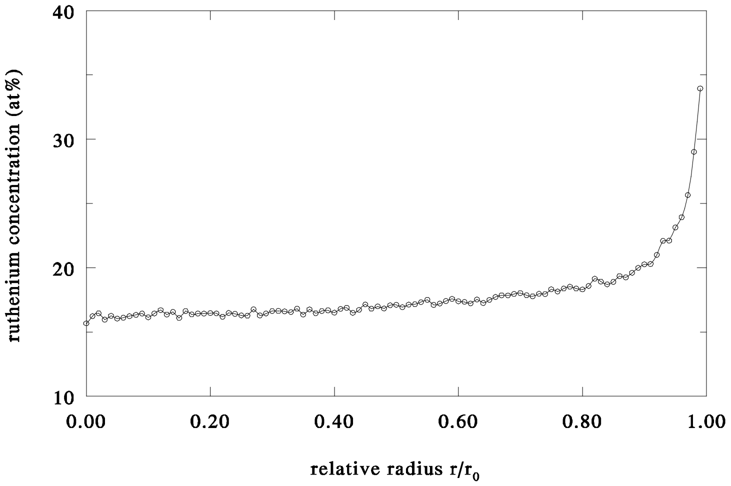


Figure 2 **A representative EPMA curve of the radial ruthenium concentration in technetium from the T2 experiment**



The cumulative irradiation time for the T2 experiments was 24 reactor cycles (579.3 full power days) during which the total neutron fluency accumulated to about 5.8×10^{26} m^{-2}. The post-irradiation examination (PIE) revealed an excellent in-pile behaviour of the metallic technetium rods. Metallographic examinations showed no changes in the microstructure compared to the unirradiated material. An example of a typical micrograph is given in Figure 1. Electron probe microanalysis (EPMA) of the radial distribution of ruthenium, the product of the transmutation process, showed an increase from about 15-16 % in the centre to 30-40 % near the rim of the pellets, giving a pellet-average Ru concentration of about 18 % (Figure 2). Mass spectrometric analysis of the pellet-average Ru concentration showed that the extent of transmutation is about 16 %.

From the results of the T2 test it is concluded that there are no technical limitations to the use of metallic technetium as a target for transmutation. No further EFTTRA irradiation experiments in the HFR are foreseen, but an irradiation test of the same material in the Phénix reactor (F2) is in preparation, as mentioned before.

Transmutation of Americium in an Inert Matrix

In 1995 the T4 experiment was initiated by the EFTTRA partners to study the feasibility of americium transmutation in a so-called once-through mode. In this scenario the extent of fissioning should be very high (>90%).The T4 experiment was performed as a shared cost action project in the cluster "P&T strategy studies and transmutation experiments" of the Fourth Framework Programme of the CEC (Contrat No. FI4I-CT95-0007)

For the T4 experiment pellets of magnesium aluminate spinel ($MgAl_2O_4$) containing about 10-12 ^{w}% ^{241}Am were fabricated by the newly developed infiltration method (INRAM) at ITU [9,13]. This methods yields a relatively fine distribution of the americium in the pellet [9,13]. However, it was observed that this distribution is not uniform and that the americium, intended to be present as an oxide, formed a compound during sintering, probably $AmAlO_3$. Two irradiation capsules were prepared.

Table 2 **Dimensional changes of the pellets of the T4 test derived indirectly from non-destructive examinations**

	Length (%)	Diameter (%)
Neutron radiography (mol[a])	+5.2	
Gamma spectrometry	+4.5	+6.7
Gamma tomography		+6.7

[a]Mol = mid-of-life (231.4 full power days)

The irradiation of the T4 target in the HFR at Petten has been performed from August 1996 to January 1998 (358.4 full power days), during which a burnup of 32% FIMA (value obtained from post-test burnup calculation) has been achieved. During the irradiation a neutron radiograph was made. After the irradiation the fuel was examined non-destructively in the hot-cell laboratories in Petten. The following results are now available [10]:

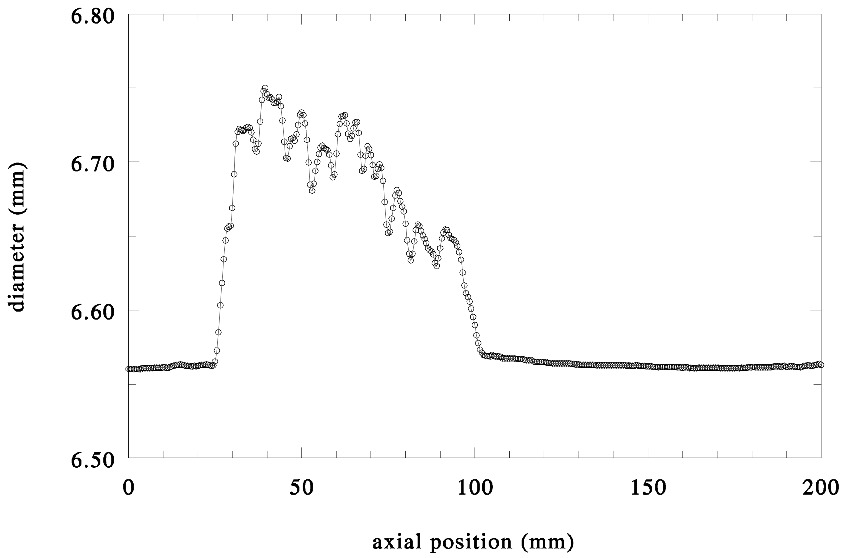
- Neutron radiography of the sample holder at mid-of-life shows no visible damage of the fuel pin and an increase of the length of the pellet stack (Table 2).

– Gamma-spectrometry and gamma-tomography of the fuel pin shows that the distribution of the fission products is correlated to the initial americium concentration of the pellets.

– Gamma-spectrometry and gamma-tomography of the fuel pin show an increase in length of the pellet stack and in the diameter of the pellets (Table 2).

Profilometry of the fuel pin showed an increase of the diameter of the cladding from 6.55 mm originally to 6.75 mm at maximum after irradiation (Figure 3).

Figure 3 .**The diameter of the cladding of the irradiated T4 rod (original diameter 6.55 mm)**



From the results it is clear that considerable swelling of the fuel occurred. From the data in Table 2 is can be calculated that the increase in volume is about 18%. This swelling can be caused by two different processes: (i) the damage of the spinel matrix by fission fragments, which can be extensive in this type of fuel due to the relatively small size of the americium dispersions, and (ii) the formation of gas bubbles containing helium that was produced by the alpha decay of ^{242}Cm, one of the isotopes in the transmutation chain of ^{241}Am. Destructive examinations to be performed at ITU have to confirm this.

The results of the T4 test, combined with results of theoretical studies and separate effect experiments, are being used to modify the design of as reported in a separate study [14]. It is proposed to use a so-called “hybrid” fuel, a dispersion of spherical inclusions (between 50 and 300 :m) of a host phase, which contains the americium, in a dense inert matrix. For the host phase the cubic solid solution $(Zr,Am)O_2$, possibly stabilised by yttrium, and for the inert matrix MgO (fast reactors) and $MgAl_2O_4$ (thermal reactors) are the most likely candidates.

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