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The EFTTRA experiment on irradiation of Am targets

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Abstract

The EFTTRA European collaboration (Experimental Feasibility of Targets for Transmutation), started in 1992, analyzes material problems related to the possibility of transmuting long-lived radioactive nuclides. One of the EFTTRA activities concerns the development of targets for the transmutation of americium in a matrix (heterogeneous cycle). The irradiation in the HFR of a sample of americium oxide embedded in a spinel matrix, the EFTTRA-T4 experiment, containing 11.2 wt.% of Am-241, was started in September 1996. © 1998 Elsevier Science S.A.

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1. Introduction

Americium is the product of the activation of uranium-238 in fuels of nuclear fission reactors and is, after plutonium, the most important contributor to the radiotoxicity of the nuclear waste. Currently, the transmutation of americium in nuclear reactors, in combination with its partitioning from the fuel, is being studied as a promising option to deal with this problem.

For the transmutation of americium new fuels need to be developed. One of the promising fuel types for transmutation is the so-called uranium-free fuel which contains a support material that is relatively inert with respect to neutron activation, the inert matrix. Several materials have been proposed as inert matrix, for example magnesia (MgO) or magnesium aluminate spinel ($MgAl_2O_4$), but the fabrication and the irradiation behaviour of such fuels are poorly known.

The activities of the EFTTRA group (Experimental Feasibility of Targets for Transmutation), which is a collaboration between EDF, CEA-Cadarache (France), JRC-ITU, FZK (Germany), JRC-IAM, and ECN (Netherlands) [1], are presently focused on the development of materials for the transmutation of technetium and of americium; the study of uranium-free americium fuel is one of their goals. An irradiation experiment for americium oxide supported by a $MgAl_2O_4$ matrix has been defined. The work is divided in three phases: sample fabrication, irradiation in the High Flux Reactor in Petten, and post-irradiation examination. This paper describes the present state of the experiment, i.e. the first two phases of the project.

2. Fuel design

The design of the pins is shown in Fig. 1. The pin consists of a fuel stack of seven pellets of magnesium aluminate spinel containing 11.2 wt.% ^{241}Am. The pellet diameter is 5.4 mm, the stack length is 70.6 mm. The pellets are enclosed in a 15/15 Ti stainless steel cladding. A pure spinel pellet and a hafnium oxide pellet are placed at the top and bottom of the fuel stack, for obtaining a homogeneous flux on the samples. In the top of the capsule a spring is placed to keep the pellets in their position.

3. Fabrication method

The fuel pins were fabricated at ITU. For the fabrication of the fuel pellets a method developed in the frame of a

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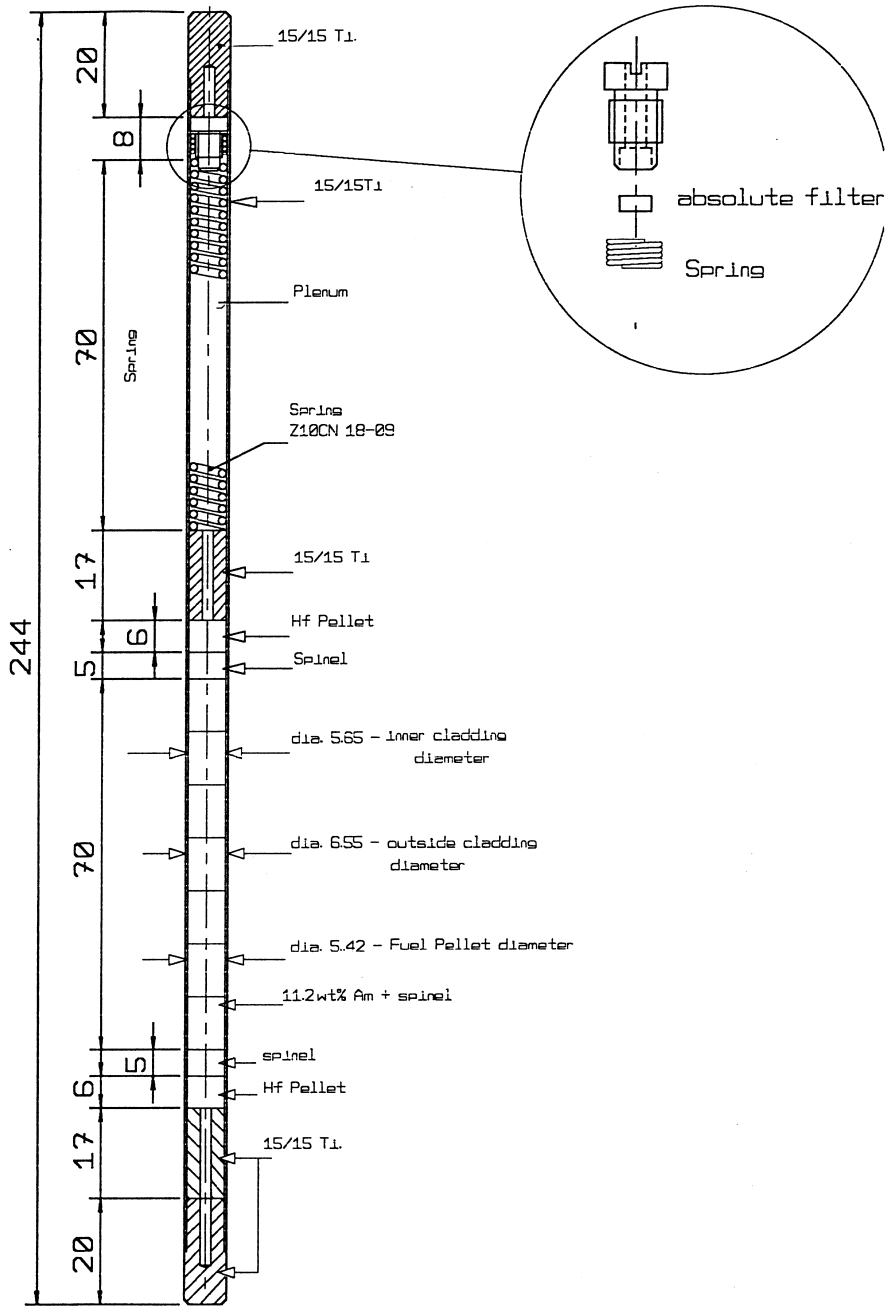

Fig. 1. Schematic drawing of the target capsule.

research programme of the Institute on innovative fabrication techniques for the fuels of tomorrow was used. The procedure, based on infiltration of aqueous actinide solutions into porous pellets, is called INRAM (Infiltration of Radioactive Materials) [2,3].

The green pellets have been fabricated from a commercial spinel powder. For infiltration, these pellets have been immersed in a nitric acid solution with an Am concentration of about 400 g/l. Following their removal from solution, the infiltrated americium nitrate was converted to the oxide by a thermal treatment and then sintered; the density of the pellets was 96–97% of the estimated theoretical value. Gravimetric analysis showed that they contained (11.2±0.3) wt.% americium. This value of Am content was confirmed by gamma spectroscopy.

The distribution of americium within the targets was determined by autoradiography measurements taken from polished faces of sectioned pellets. The measurements indicate that there is a roughly cylindrically symmetric shell, 200 μm thick, in the pellet, within which the americium content is higher than in the surrounding region. Similar features are also observed in X-ray radiographs of the samples. These results have been further quantified using EMPA: within the americium-rich shell,

the americium concentration is about 14 wt.%; in the regions both inside and outside of this shell, it is about 9 wt.%. The presence of a cylindrical shell within the pellet is also observed in the optical micrographs of the sectioned pellets; this effect is most likely due to a porosity change in this region. Moreover, the micrographs indicate that the americium-containing particles within the spinel matrix have a diameter of less than 2–3 μm. This is significantly smaller than that obtained in other samples prepared by pressing mixtures of powders.

After the controls as required by the specifications, the pellets were loaded and welded into two target capsules. All the filling operations were performed in glove boxes under a controlled inert atmosphere. The capsules were sent to Petten for irradiation: the first capsule was loaded in HFR (EFTTRA-T4 experiment), and the second one kept for the EFTTRA-T4bis experiment, to be started later.

4. Irradiation facility/irradiation conditions

The target capsule has been placed in a central hole of the sample holder, which is instrumented with thermocouples and monitor sets for neutron metrology. The irradiation is carried out in one leg of a TRIO irradiation facility, which is a standard in-core device [4]. The in-pile part of the irradiation facility consists of two independently controlled containments which enclose the target capsule.

The irradiation in the HFR started on 19 September 1996; meanwhile nine irradiation cycles have been completed and the cumulative irradiation time is 231 full power days. The neutron fluence ($E>0.1$ MeV) is about 1×10^{26} m^{-2}. The temperature of the cladding, which is adjusted by the control system, varies between 390 and 450°C.

In July 1997 the irradiation was interrupted for a neutron radiograph. The results show that the fuel pin and fuel stack are still intact. No indication for significant swelling was obtained since the gap between pellets and cladding could still be observed.

The irradiation continued in August 1997 and will last until January 1998, after which the cumulative irradiation time will be about 360 full power days.

5. Characteristics of the irradiation

Pre-test calculations were made for a pin containing 10 wt.% AmO_2 in spinel for up to 400 days of irradiation in the HFR using the WIMS6 nuclear code package.

Fig. 2 shows the calculated fission power as a function of the irradiation time. The first maximum after 50 days, for which the fission power is about 185 W cm^{-3}, is due to the fission of the transmutation product ^{242m}Am; the maximum after 375 days, for which the fission power is about 270 W cm^{-3}, is due to fission of ^{239}Pu, which is formed by subsequent activation and decay processes ($^{241}\text{Am}\rightarrow{}^{242}\text{Cm}\rightarrow{}^{238}\text{Pu}\rightarrow{}^{239}\text{Pu}$).

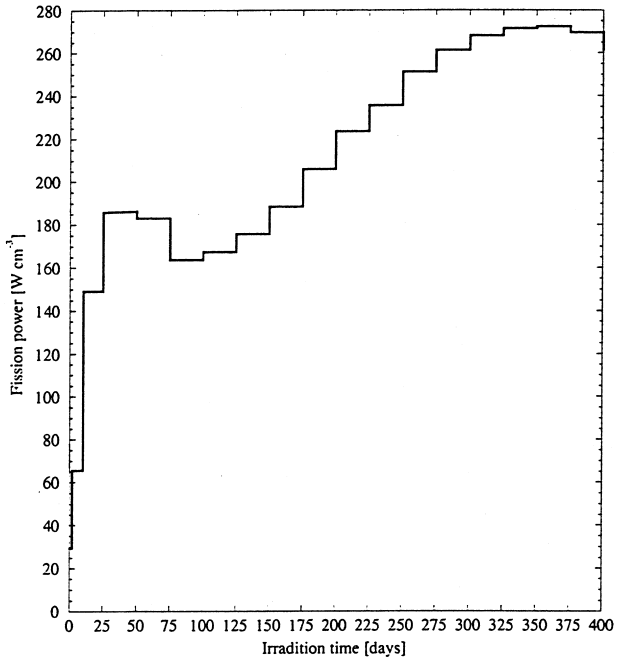

Fig. 2. Fission power in the Am target as a function of irradiation time.

The burn-up calculations show that Am will be almost completely transmuted during the irradiation period (Fig. 3): the residual ^{241}Am fraction is e.g. 0.4% after 400 days. However, since Am is transmuted to other actinides, the total actinide reduction should be about 35%, according to the calculation.

As a result of the alpha decay of ^{242}Cm into ^{238}Pu, a significant amount of helium gas is produced in the target. The amount of helium produced as a function of the

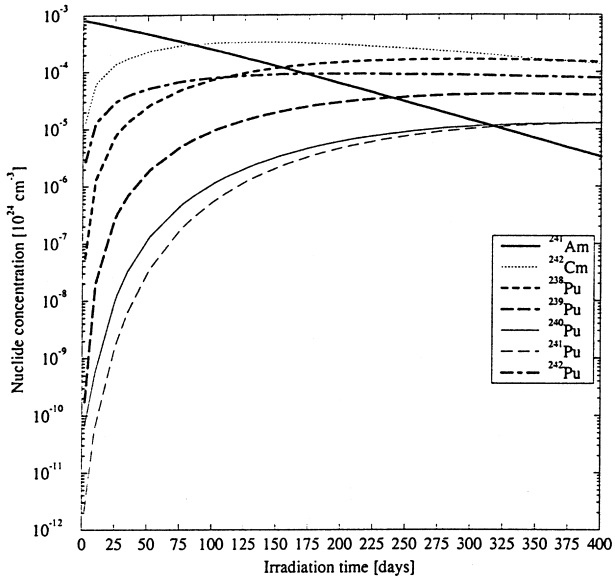

Fig. 3. Main actinides concentration during irradiation in the HFR Petten.

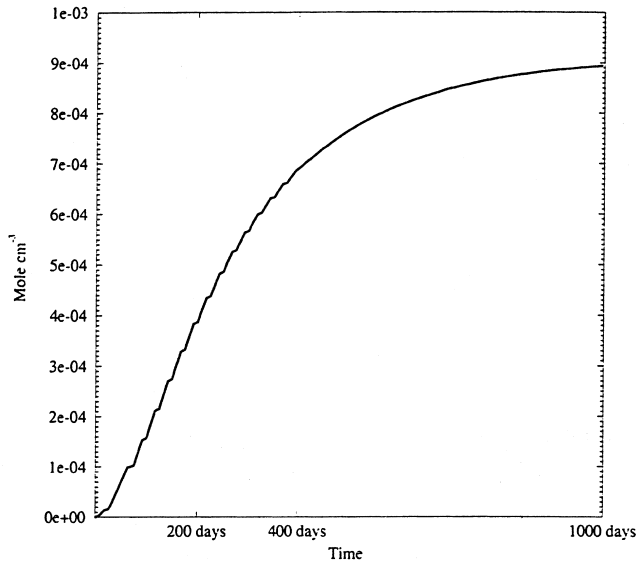

Fig. 4. Amount of helium produced per cm^3 of sample during and after irradiation (end of irradiation after 400 days).

irradiation and cooling time is shown in Fig. 4. At the end of the irradiation the amount of helium is about 10 times larger than the amount of noble gases produced by fission.

6. Future plans

After completion of the irradiation, in the beginning of 1998, the EFFTRA-T4 samples will be sent to ITU for examination. EFTTRA-T4 will be followed by two further americium target irradiation experiments, EFTTRA-T4bis and ECRIN. The first one is an EFTTRA experiment, consisting of the irradiation in HFR of a similar target to EFTTRA-T4, but for a longer irradiation time (625 full power days), and was started in August 1997. The samples were produced by ITU, using the same impregnation technique. The ECRIN experiment, consisting in the irradiation of americium targets in the Superphenix reactor, is not an EFTTRA experiment, but a CEA experiment, with the participation of ITU: valuable information is expected from the comparison of the results of EFTTRA-T4bis and ECRIN, i.e. of the irradiation of an americium target in a thermal flux and in a fast neutron flux. The EFTTRA-T4 and T4bis Am target irradiation experiments are part of the EFTTRA experimental programme, aiming at contributing to the evaluation of the possible future solutions for the treatment of nuclear wastes.

Acknowledgements

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