ECRIX-H experiment: Post-irradiation examinations and simulations

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Abstract

ECRIX-H experiment is devoted to study of the behaviour under irradiation of magnesia-based inert matrix fuels and targets for minor actinide (MA) transmutation in accelerator-driven systems (ADS) or in fast neutron reactors (FR) according to the heterogeneous route. A MgO-AmO$_{1.62}$ composite ceramic target was irradiated during 318 EFPD in the Phénix sodium-cooled fast reactor (SFR) in a specific moderator carrier subassembly. Neutron moderation makes it possible to achieve high rate in minor actinide transmutation, combining a high neutron flux and the increased cross-sections due to the slowed neutrons.

This article first recalls the ECRIX-H design and fabrication. Secondly, irradiation conditions are described and a summary of the results of post-irradiation examinations is presented. Then, post-irradiation thermal simulations are detailed together with an analysis of the behaviour of MgO-AmO$_{1.62}$ targets under irradiation. Results show that magnesia-based inert matrix targets display a satisfactory thermo-chemical-mechanical behaviour and moderated swelling (6.7 vol.%) under irradiation at about 700-800°C, even for significant quantities of helium produced and burn-up. On this basis, the preliminary design of transmutation fuel pins could be optimised so as to increase their performance level (initial MA content, burn-up…).

The achieved measured fission rate (25 at.%) is found to be lower than the one expected from neutronic simulations. The deviation could be due to the possible evolution of the physical and chemical properties CaH$_x$ under irradiation. It could also be due to neutronic simulation inaccuracies considering the neutronic modelling complexity and the uncertainties on nuclear data related to moderated neutron spectrum.

In addition, the major part of initial Am being transmuted into Pu under irradiation, a PuO$_x$-type phase is created within the initial AmO$_{1.62}$ particles, leading to an incomplete dissolution of irradiated targets in standard reprocessing conditions. This issue will have to be considered when dealing with transmutation fuels and targets devoted to multi-recycling of MA.

Introduction

This paper summarises the post-irradiation examinations and simulations related to the ECRIX-H experiment. The irradiation of the MgO-$AmO_{1.62}$ composite under a locally moderated neutron flux inside the Phénix reactor is part of studies conducted on the behaviour of magnesia-based inert matrix fuels and targets for minor actinide transmutation under irradiation in accelerator-driven systems (ADS) or in fast neutron reactors (FR) based on a heterogeneous mode [1,2].

Based on stability criteria under irradiation, thermal and chemical properties, neutron transparency, manufacturing and reprocessing possibilities, magnesia was initially identified as one of the best options for inert matrix transmutation targets from among several other possibilities ($MgAl_2O_4$, Al_2O_3, $Y_3Al_5O_{12}$, CeO_2, Y_2O_3, TiN, Cr, V, W, etc.) [1]. Preliminary studies conducted on the design of target fuel pins for the transmutation of minor actinides (americium and curium) in an MgO matrix have furthermore showed that performance levels would greatly depend on the swelling of irradiated targets (mainly due to the significant production of helium) [3,4]. More specifically, the swelling of target pins in an ADS reactor must not exceed about 12% (for an initial americium oxide content of about 3 g/cm^3 of fuel and a minor actinide fission rate of around 16 at.% at the end of the cycle) [5,6]. Higher swelling rates would initially require providing a large fuel-cladding gap to prevent any mechanical interaction between the fuel and cladding during irradiation. A large gap could generate an indentation risk for the cladding due to the possible displacement of fragmented fuel into the gas gap and due to the excessive temperatures reached during the irradiation phase.

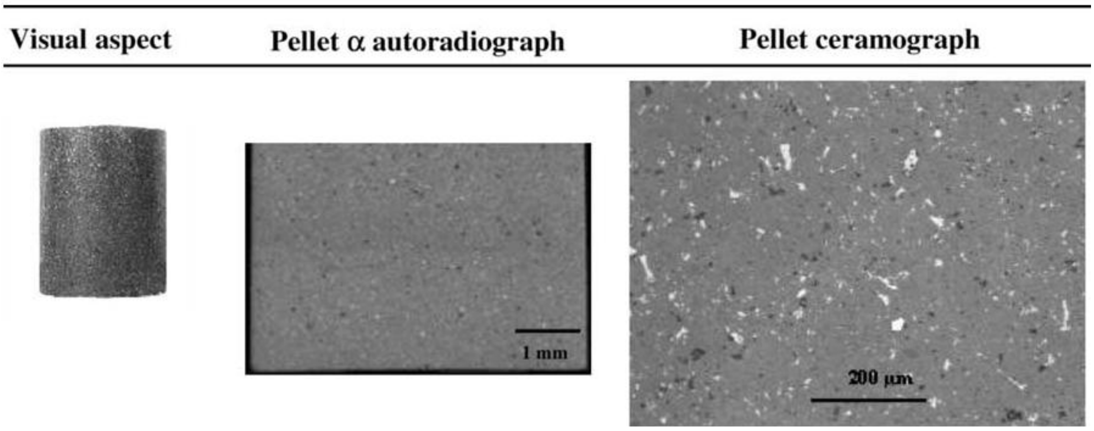
A complete set of experiments (EFTTRA T3, MATINA1, MATINA 1A, BORA-BORA, ECRIX-H, ECRIX-B, MATINA2-3, COCHIX, HELIOS, FUTURIX-FTA), which have either been completed or are still under way, will help to accurately assess the capacity of magnesia to provide an efficient support matrix for transmutation targets and fuels [1,7]. The purpose of the EFTTRA T3, MATINA 1 and 1A, BORA-BORA and MATINA2-3 irradiation experiments was to study the behaviour of MgO and particularly its swelling under a neutron flux, as well as under a neutron flux associated with damage generated by fission products from uranium or plutonium dioxide particles dispersed inside the matrix with different burn-ups and various microstructures. In addition, the helium release/swelling issue is addressed in the case of the ECRIX-H, ECRIX-B, CAMIX-COCHIX and FUTURIX-FTA irradiation experiments due to the presence of ^{241}Am in the targets and in the experimental fuels.

The results of the post-irradiation examinations EFTTRA T3, MATINA 1 and 1A and BORA-BORA have confirmed that magnesia remains stable and behaves satisfactorily under irradiation [7-10]. Results concerning the BORA-BORA PuO_2-MgO irradiation experiment have shown that, when no helium is produced due to ^{241}Am transmutation, the swelling of the composite targets with an MgO matrix was compatible with the preliminary design of the fuel pins and the transmutation assemblies, even for a high burn-up of about 278 GWd/m^3. The ECRIX-H irradiation experiment will first provide data on helium swelling for magnesia-based inert matrix targets containing significant amounts of ^{241}Am. This paper first recalls the ECRIX-H design and fabrication. Secondly, the irradiation conditions are described. A synthesis of post-irradiation examinations (PIE) is presented. The post-irradiation thermal simulations are then detailed together with an analysis of the behaviour of MgO-$AmO_{1.62}$ targets under irradiation.

ECRIX-H targets, pin and carrier design and fabrication

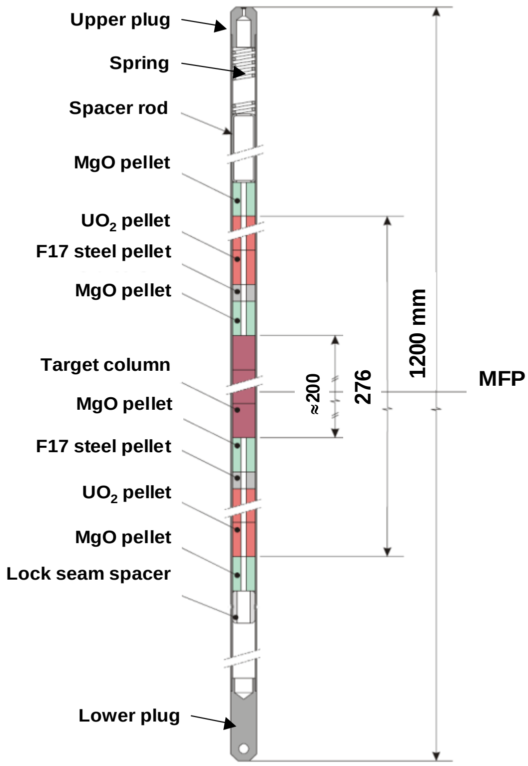
The ECRIX targets were manufactured in the Atalante laboratories at the CEA-Marcoule nuclear facility [11] according to a classical powder metallurgy method. The sintered pellets contained 16.65 wt.% of Am microdispersed in MgO, with the ^{243}Am/(^{241}Am+^{243}Am) ratio being equal to 5.45%. The average diameter and height of the pellets were equal to 5.14 and 6.30 mm, respectively. The O/Am ratio and density were equal to 1.62 and 3.98 g/cm^3 respectively, thus representing 97% of the theoretical mass-volume density. The overall porosity was equivalent to 3%, of which 0.15% was open porosity. Figure 1 shows an α autoradiograph and a ceramographic observation of an ECRIX-H pellet.

Figure 1: Visual aspect, α autoradiograph and ceramographic examination of an ECRIX-H pellet [11]



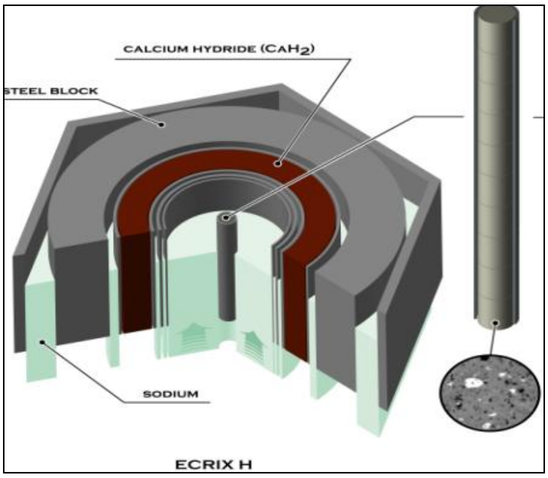
The fuel pin design is detailed in Figure 2. The pin included 32 composite cercer targets. The americium-based target column measured 201.6 mm and contained a total of 2.765 g of americium. Three UO_2 pellets with 4% ^{235}U enrichment were positioned on either side of the target column to detect cladding failure via the delayed neutron detection system in the Phénix reactor. Thermal and chemical insulating MgO and F17 steel pellets surrounded the different fuel columns. The cladding used in this experiment was made of cold-worked AIM1. The filling atmosphere initially contained almost pure helium at a pressure of around 0.1 MPa (at 20°C).

Figure 2: Illustration of the ECRIX-H pin



The ECRIX-H pin is placed in a specific carrier subassembly equipped with annular blocks of CaHx which act as a moderator. When combined with a high neutron flux in an FR, the increased cross-sections due to the slowed neutrons makes it possible to achieve high rate in minor actinide transmutation [12], while limiting damage to the cladding. A diagram representing the pin positioned inside the moderated carrier is shown in Figure 3 [13]. The calcium hydride rings contained inside a leak-tight metal container (which is surrounded by a steel shell) are about 9 mm thick, with an average H/Ca atomic ratio of 1.92 and a total hydrogen mass of about 40 g.

Figure 3: Illustration of the specific carrier sub-assembly (DMC-2) used in the ECRIX-H experiment



Irradiation conditions

ECRIX-H neutronic simulations were performed by using the GEPHIX and CESAR calculation codes [14] devoted to the management of the Phénix plant. Complementary simulations were also performed using the ERANOS and DARWIN codes [15] to obtain a more detailed description of fuel evolution during irradiation, including its composition in terms of helium and fission products. The ECRIX-H pin was irradiated for 318 EFPD in the Phénix reactor and integrated an overall fluence of 6.12 10^{26} $n.m^{-2}$, including 29% of fast fluence ($E > 0.1$ MeV). The fluences calculated at the bottom and top of the americium-based target column amounted to 95% and 93% of the maximum value respectively. The integrated dose at the maximum flux plane was equal to 8.78 dpa NRT Fe.

The Am transmutation rate determined from neutronic simulations was equal to 93 at.%; this value is consistent with the one measured from post-irradiation destructive examinations equal to 94 at.% (as explained later). On the contrary, on the basis of these examinations, the measured fission rate (25 at.%) was found to be lower than that predicted by neutronic simulations (34 at.%). This deviation could be due to the possible variation in the physical and chemical properties of CaHx under irradiation not taken into account in the simulation. It could also be due to neutronic simulation inaccuracies considering the complexity of neutron modelling and the uncertainties on nuclear data related to moderated neutron spectrums. Figure 4 represents the linear power and burn-up factor variations for americium-based targets at the maximum flux plane as a function of the irradiation time. At end of the irradiation phase, the burn-up reaches 154 GW.d per initial m^3 of target or 39 GW.d per tonne of target at the maximum flux plane. The He production amounts to around 5.7 mg of He per cm^3 of initial target.

Non-destructive post-irradiation examinations

The examinations performed in the CEI facility of the Phénix reactor and in the LECA-STAR facility at the Cadarache centre included visual inspections, metrology, gamma spectrometry and Eddy current testing. No abnormalities were observed during the visual inspection of the entire length of the capsule and fuel pin. No post-irradiation elongation was detected in the pin.

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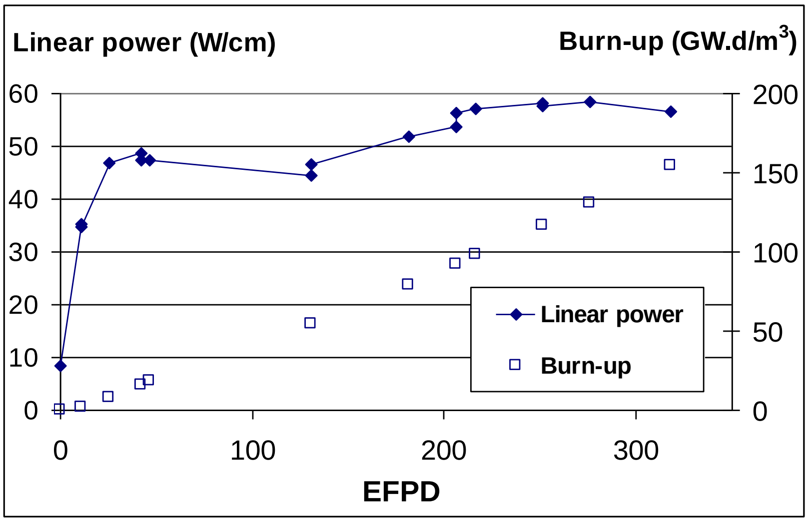
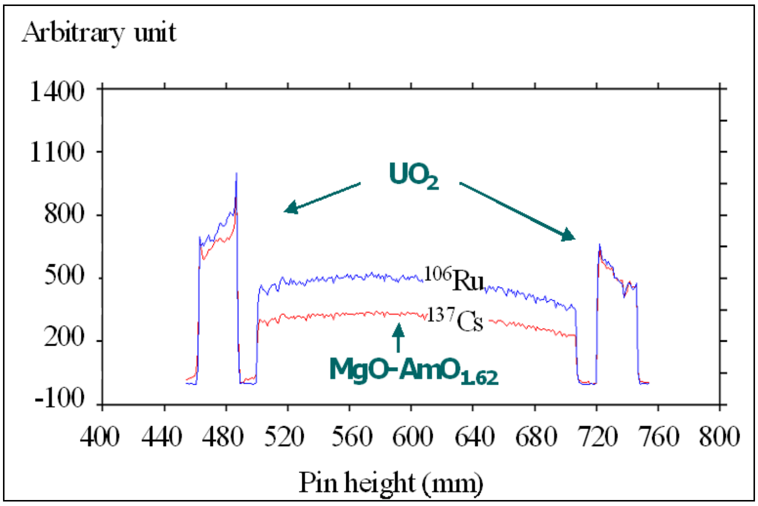
Figure 4: Power and burn-up of targets at the maximum flux plane as a function of irradiation time



A diametral metrology analysis of the pin by contact made it possible to detect a slight diameter increase, which was observed in the americium-based target column (around 6 μm). This deformation was probably due to the thermal expansion of the cladding caused by the decay heat from the targets after irradiation (around 5 W for all 32 targets when profilometry measurements were carried out). No major defects (cracks, internal corrosion, etc.) were detected by Eddy current testing on the ECRIX-H pin.

Figure 5 represents ^{106}Ru and ^{137}Cs gamma spectrometry profiles. ^{106}Ru is a non-migrating fission product that provides information on the height of the target column. The elongation of the target column was around 5.5 mm, i.e. around 2.7% of its relative length. The inter-pellets of the central column were not particularly opened and were mostly visible at the end sections. Inter-pellets were also visible on UO_2 pellet columns. The ^{137}Cs profile does not indicate any significant migration of this isotope. Moderate irradiation temperatures (see later) explain why only a slight migration has occurred under irradiation.

Figure 5: ^{106}Ru and ^{137}Cs gamma spectrometry profiles



In summary, the non-destructive examinations carried out in this experiment have confirmed the satisfactory behaviour of the fuel pin: no longitudinal elongation of the fuel pin, very limited diametral deformation, no sign of alteration in the cladding material, no significant migration, and no axial redistribution of the fission products.

Destructive post-irradiation examinations

The destructive operations and measurements were performed in the LECA-STAR facility at CEA-Cadarache, and in the Atalante facility at CEA-Marcoule. This section briefly describes the following examinations: pin puncturing, gas analysis, pellet recovery and geometrical and hydrostat density measurements of the targets. The main results of ceramographic and scanning electron microscopy (SEM) observations of an irradiated target are also summarised, together with the results obtained by means of electron probe microanalysis (EPMA) and X-ray diffraction (XRD). Chemical and isotopic analyses performed on americium-based irradiated pellets are then presented. The above-mentioned destructive examinations together with secondary ion mass spectroscopy (SIMS) measurements, SEM observations performed on a fractographic surface, and annealing tests are described elsewhere in detail [16-18].

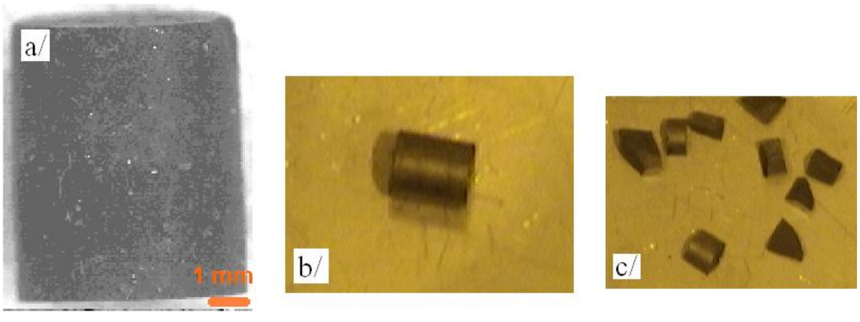
Pin gas analysis and target density measurements

The ECRIX-H pin was punctured and the volumes of helium, xenon and krypton in the filling gas were measured. Considering the total helium and fission gas production assessment, the following fractions were released:

- around 23% for He (as the production is mostly due to Am, this release rate corresponds to that of americium-based targets, excluding of course initial He filling);
- around 4% for Xe and Kr (as the production is due to Am and U in similar proportions, this release rate represents an average value for all americium-based targets and UO_2 pellets).

Initially, all objects were to be recovered by cutting the pin at the level of the spacer tube and the crimped spacer. The objects were to be recovered one by one, pushing them out using a metal rod adapted to the internal pin diameter. In fact, most targets were extracted in relatively large fragments (see illustration in Figure 6) and up to ten cuts were required to extract the objects. The target fragments obviously moved and came in contact with the cladding, thus preventing the target column from moving easily when tilting the pin or using the metal rod. Two Am-based targets were recovered whole (Figure 6). From macroscopic examinations of the whole pellet, practically no open crack was observed on the targets at this observation scale. Some chips were found on the pellet surface. A decrease of around 6.7% was found in the geometrical density of the two whole pellets. This is consistent with the target column elongation as measured from the ^{106}Ru gamma spectrometry profile (2.7%). Geometrical measurements indicated isotropic swelling for the two targets recovered whole.

Figure 6: Macroscope (a) and periscope (b,c) photographs of irradiated fragmented (c) and whole (a,b) targets



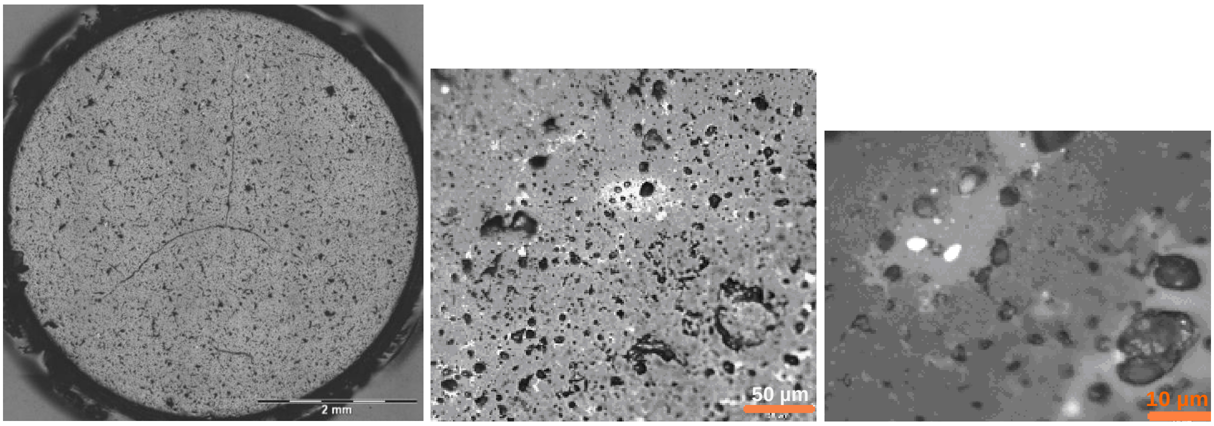
Ceramographic, SEM, chemical, EPMA, SIMS and XRD analyses of irradiated targets

Ceramographic and SEM examinations make it possible to observe long distance axial and radial thermal cracks having led to the fragmentation of targets. Many porosities or bubbles were found within or on the surface of the ex-$AmO_{1.62}$ particles in the targets (see Figure 7). As indicated by means of ceramographic image analysis, about 6.4% of porosity was created during irradiation. The 6.7 vol.% macroscopic swelling of targets is thus mainly due to the creation of these porosities probably generated by fission gas and/or helium precipitation [18]. This is confirmed by the fact that no amorphisation and almost no chemical variation in the host MgO matrix were detected after irradiation, as seen by means of XRD and EPMA [17].

Figure 7: Ceramographic observations of Am-based targets

(a) and (b) – microdispersed ex-$AmO_{1.62}$ particles appear in white, the MgO matrix in gray, and porosities and cracks in black
(c) – fission product precipitates appear in white within the light grey ex-$AmO_{1.62}$ particles

(a) (b) (c)



Small discontinuous cracks were observed between two areas with a high ex-$AmO_{1.62}$ particle density and between ex-$AmO_{1.62}$ particles which were larger than average. These cracks were probably generated by the swelling of ex-$AmO_{1.62}$ particles resulting from the formation of porosities in these particles (see Figure 8 for illustration). Cracking between ex-$AmO_{1.62}$ particles could partly explain the gaseous release measured during pin puncturing (see previous section).

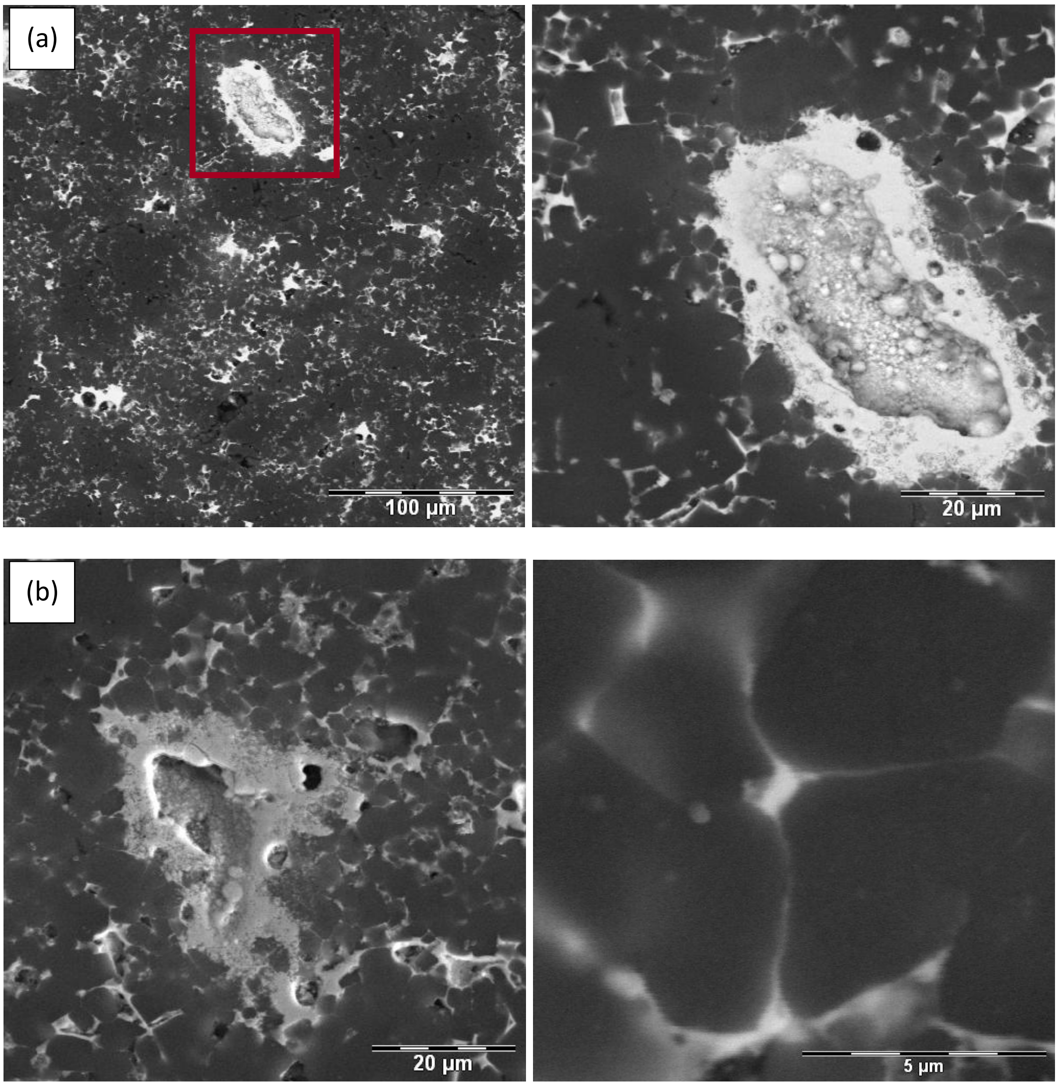
A big size porosity in an ex-$AmO_{1.62}$ particle can be observed on SEM images presented in Figure 8(a). Small size porosities (around 0.2 μm) were also found within such particles. Almost no porosity was encountered within the MgO matrix. At the periphery of the target, porosities up to 5 μm size were also detected at the interface between the matrix and some ex-$AmO_{1.62}$ particles. No chemical interaction could be observed between the matrix and the particles.

As indicated by EPMA, Pu, Am and Cm were found to be co-localised within the ex-$AmO_{1.62}$ particles. From XRD examination, these elements are situated into a crystallographic PuO_x-type phase. No migration of metal fission products was detected after their implantation into the MgO matrix (on about 10 μm in depth). On the contrary, precipitates of these fission products were found in the ex-$AmO_{1.62}$ particles with a diameter that could reach several microns [see Figure 7(c)]. From SIMS measurements, implanted Xe was found in the MgO matrix, mainly dissolved in it. In the particles, Xe was found to be mainly localised into bubbles. He release is found to occur preferentially at the core of the pellet.

For chemical analysis purpose, an irradiated Am-based target sample was dissolved in a boiling 4 M HNO_3 acid solution, in relation to the standard dissolution conditions for reprocessing of UO_2 and MOX fuels. As a result of the first dissolution, a significant quantity of non-dissolved matter could be visually observed. This fraction of non-dissolved sample probably corresponded to the previously-mentioned PuOx-type phase created within the ex-$AmO_{1.62}$ particles under

Figure 8: SEM observation of irradiated target

(a) Backscattered, (b) Secondary electrons



irradiation which cannot be dissolved in standard reprocessing conditions. Another sample was thus dissolved by using a boiling 4 M HNO_3 acid solution including 0.1 M HF in order to achieve full dissolution for chemical analysis. From these analyses, Am transmutation and fission rates could be determined: respectively 94 and 25 at.%. It should be noted that almost the same values were found by means of EPMA.

Post-irradiation thermal simulations

A 2-D axisymmetrical thermomechanical model with an (R,Z) representation was used to assess the behaviour of the americium-based targets under irradiation. Characteristics of the calculation performed with the CAST3M finite-element code developed at CEA are detailed elsewhere [16]. Compared to the calculation performed previously [16], an updated simulation including all the results presented in the present paper was achieved. Maximum temperatures at the core of the pellet under irradiation are finally found to be situated around 700 to 800°C, with a low thermal gradient in the pellets (<100°C).

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Conclusion

This document summarises the post-irradiation examinations and simulations performed for the ECRIX-H irradiation experiment. This experiment falls within the scope of studies on the behaviour of magnesia (MgO) when used as an inert matrix for transmutation fuels and targets in either accelerator-driven systems (ADS), or in fast neutron reactors (FR) using a heterogeneous recycling mode. The aim is to collect data on the behaviour of MgO and particularly on its swelling characteristics under the effect of damage generated by the neutron flux, the fission products, and the significant production of helium due to ^{241}Am transmutation.

The new results obtained within the scope of the ECRIX-H experiment supplement the results of other irradiation experiments (EFTTRA T3, MATINA 1 and 1A, BORA-BORA). These results show that magnesia-based inert matrix targets display a satisfactory thermo-chemical-mechanical behaviour and moderated swelling (6.7 vol.%) under irradiation at about 700-800°C, even for significant quantities of helium produced and burn-up. On this basis, the preliminary design of transmutation fuel pins could be optimised so as to increase their performance level (initial MA content, burn-up…). Moreover, the ECRIX-H experiment tends to prove that MgO is an efficient support matrix for the confinement of fission products resulting from fissile particles in the targets, for the temperature range studied within the framework of the irradiation (i.e. about 700 to 800°C).

The achieved measured fission rate (25 at.%) is found to be lower than the one expected from neutronic simulations. The deviation could be due to the possible evolution of the physical and chemical properties CaH_x under irradiation. It could also be due to neutronic simulation inaccuracies considering the neutronic modelling complexity and the uncertainties on nuclear data related to moderated neutron spectrum.

In addition, the major part of initial Am being transmuted into Pu under irradiation, a PuO_x-type phase is created within the initial $AmO_{1.62}$ particles, leading to an incomplete dissolution of irradiated targets in standard reprocessing conditions. This issue will have to be considered when dealing with transmutation fuels and targets devoted to multi-recycling of MA.

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