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Microstructure and elemental distribution of americium-containing uranium plutonium mixed oxide fuel under a short-term irradiation test in a fast reactor

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ABSTRACT

In order to investigate the effect of americium addition in MOX fuel on the irradiation behavior, the 'Am-1' program is being conducted in the experimental fast reactor Joyo. The Am-1 program consists of two short-term irradiation tests of 10 min and 24 h irradiations and a steady-state irradiation test. The short-term irradiation tests were successfully completed and the post irradiation examinations (PIEs) are in progress. This paper reports on the results of PIEs for Am-containing MOX fuel irradiated for 10 min. MOX fuel pellets containing 3% or 5% Am were fabricated in a shielded air-tight hot cell using a remote handling technique. The oxygen to metal ratio (O/M) of these fuel pellets was 1.98. They were irradiated at peak linear heating rate of about 43 kW m^{-1}. Focus was being placed on migration behavior of Am during the irradiation. The ceramography results showed that structural changes such as lenticular pores and a central void occurred early, within the brief 10 min of irradiation. The results of electron probe micro-analysis revealed that the concentration of Am increased in the vicinity of the central void.

1. Introduction

A closed nuclear cycle based on the fast reactor is one of the most promising ways to achieve a sustainable energy supply [1]. In this cycle management of minor actinides (MA) such as americium (Am), neptunium (Np) and curium (Cm) is of crucial importance from the viewpoint of reduction of environmental burden, effective use of natural resources and enhancement of nuclear non-proliferation. MA-containing uranium (U) and plutonium (Pu) mixed oxide (MOX) fuel, in which several percent of MA and fission products are contained, is a promising candidate for such a fuel cycle. As a part of the research and development of MA-containing MOX (MA-MOX) fuel, an irradiation program called 'Am-1', is being conducted in the experimental fast reactor Joyo of the Japan Atomic Energy Agency (JAEA). The Am-1 program consists of two short-term irradiation tests of 10 min and 24 h irradiations and a steady-state irradiation test. The objectives of the short-term irradiation tests are to confirm whether or not fuel melting occurs at a high linear heating rate and to evaluate the redistribution behavior during the initial burn-up. The steady-state irradiation test is expected to evaluate the behavior of the fuel cladding chemical interaction (FCCI) and helium (He) release behavior up to medium or high burn-up. The couple of short-term irradiation tests were successfully completed and the post irradiation examinations (PIEs) have been conducted.

Two kinds of fuels were subjected to the irradiation tests of the Am-1 program. One was MOX fuel containing 3% or 5% Am (Am-MOX) and the other was MOX fuel containing 2% Np and 2% Am ((Am, Np)-MOX). Am-MOX fuels were fabricated using a remote handling technique in the shielded air-tight hot cell of the Alpha–Gamma Facility (AGF) at JAEA's Oarai Research and Development Center. (Am, Np)-MOX fuels were fabricated in a glove box unit of JAEA's Plutonium Fuel Development Facility (PFDF) at Tokai Research and Development Center. Two values of the oxygen to metal molar fraction ratio (O/M) of the fuel pellets were used as a test parameter in the Am-1 program.

The two short-term irradiation tests are outlined as follows. Three Am-MOX fuel pins and three (Am, Np)-MOX fuel pins were loaded into the B11(1) fuel subassembly by remote manipulation. This was done inside the hot cell of JAEA's Fuel Monitoring Facility (FMF) at the Oarai Research and Development Center. The first irradiation test was conducted at peak linear heating rate of about 43 kW m^{-1} for 10 min. Following the irradiation, one Am-MOX fuel pin and one (Am, Np)-MOX fuel pin were removed from the fuel assembly and were supplied for PIEs. The remaining four fuel pins were re-installed into another fuel subassembly, the B11(2), and then irradiated for 24 h at almost the same peak linear heating rate as that of the first irradiation test. In the short-term irradiation tests, focus is placed on elucidating the microstructural evolution and migration behavior of Am on the MA-MOX fuels at the initial

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Table 1
Nominal specifications of Am-MOX fuel pin.

Fuel pellet	
Diameter (mm)	6.52
Height (mm)	8
O/M molar fraction ratio	1.98
Relative density (%)	93
Pu content : Pu/(U + Pu + Am) (wt%)	29
Am content : Am/(U + Pu + Am) (wt%)	3 and 5
Fuel stack length (mm)	200
Cladding	
Material	15 Cr–20 Ni stainless steel
Outer diameter (mm)	7.5
Thickness (mm)	0.4

stage of irradiation. A detailed description concerning the Am-1 program, including fuel design and irradiation conditions, can be found in the literature [2].

This paper reports on the results of ceramography and electron probe microanalysis (EPMA) for Am-MOX fuel pellets irradiated for 10 min.

2. Experimental

2.1. Specifications of the fuel pellets

Table 1 lists specifications of the Am-MOX fuel pin. Its fuel pellets containing 3% or 5% Am were fabricated using the remote handling system [3] in the AGF. The AGF was originally constructed as the PIE facility for irradiated fuels [4]. It was later equipped with a small-scale fuel fabrication unit in the hot cell for the purpose of developing a remote handling fuel fabrication technology for MA-MOX fuel. The conventional powder metallurgy method was adopted for the fabrication of Am-MOX fuel pellets. Fig. 1 shows a flow sheet of the fuel pin fabrication process.

Powders of UO_2 and two kinds of Am-containing PuO_2 powders were used as raw materials. Detailed characterization of the raw powders has been reported in the literature [3]. Before the sintering step, these three powders were annealed in a furnace with a cantal heater to remove their absorbed moisture. After heat treatment, these powders were weighed by using an electronic balance in order to adjust the amount to the predetermined weight ratio. This was followed by mixing in a ball mill for 5 h. Zinc stearate was added as binder and the powder was further mixed for 30 min before cold-pressing on green pellets under a pressure of 3.84 t cm^{-2}. The green pellets were pre-sintered at 1073 K for 2.5 h in the furnace with the cantal heater to remove the binder and were sintered at 1973 K for 3 h in a furnace with a tungsten mesh heater. The pellets were then heat treated to adjust O/M ratio to 1.98. The O/M ratio of the pellets was confirmed gravimetrically at room temperature. All of the heat treatments and sintering processes were carried out under the flowing gas atmosphere of Ar-5% H_2 or Ar-0.05% H_2 by adding an appropriate amount of moisture.

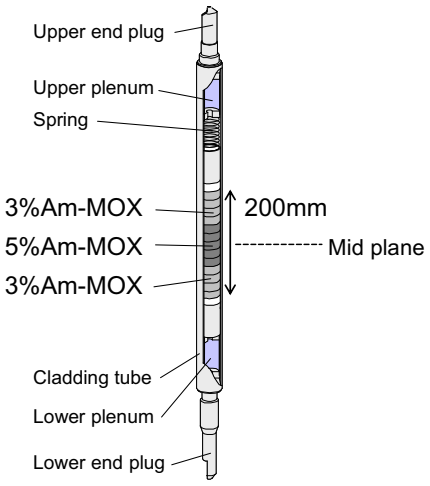

Fig. 2. Configuration of the Am-MOX fuel pin.

The Am-MOX fuel pellets were inserted into a cladding tube made of austenitic stainless steel (Table 1), together with thermal insulator pellets of UO_2 and reflector components. The Am-MOX fuel pellets containing 5% Am were loaded at the middle of the fuel column and 3% Am-containing MOX fuel pellets were placed at the lower and upper ends of the fuel column. The fuel pins were sealed by TIG welding and then inspected using a He-leak detection system for confirmation of their air-tightness/soundness. Fig. 2 shows a schematic drawing of the Am-MOX fuel pin. Manufactured fuel pins were then assembled into the fuel subassembly B11(1).

2.2. Irradiation conditions

After the reactor power was raised continuously to the targeted peak linear heating rate of 43 kW m^{-1} and kept there for 10 min, the power was manually shut down. The fuel subassembly B11(1) was irradiated up to around 0.0057% FIMA in the core center location [000] of the operational cycle 3-1 of the Joyo MK-III

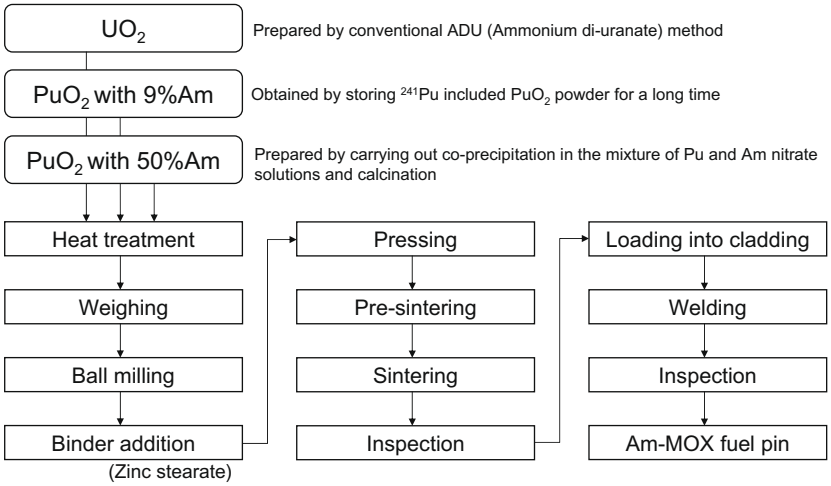

Fig. 1. Fabrication flow of Am-MOX fuel pin.

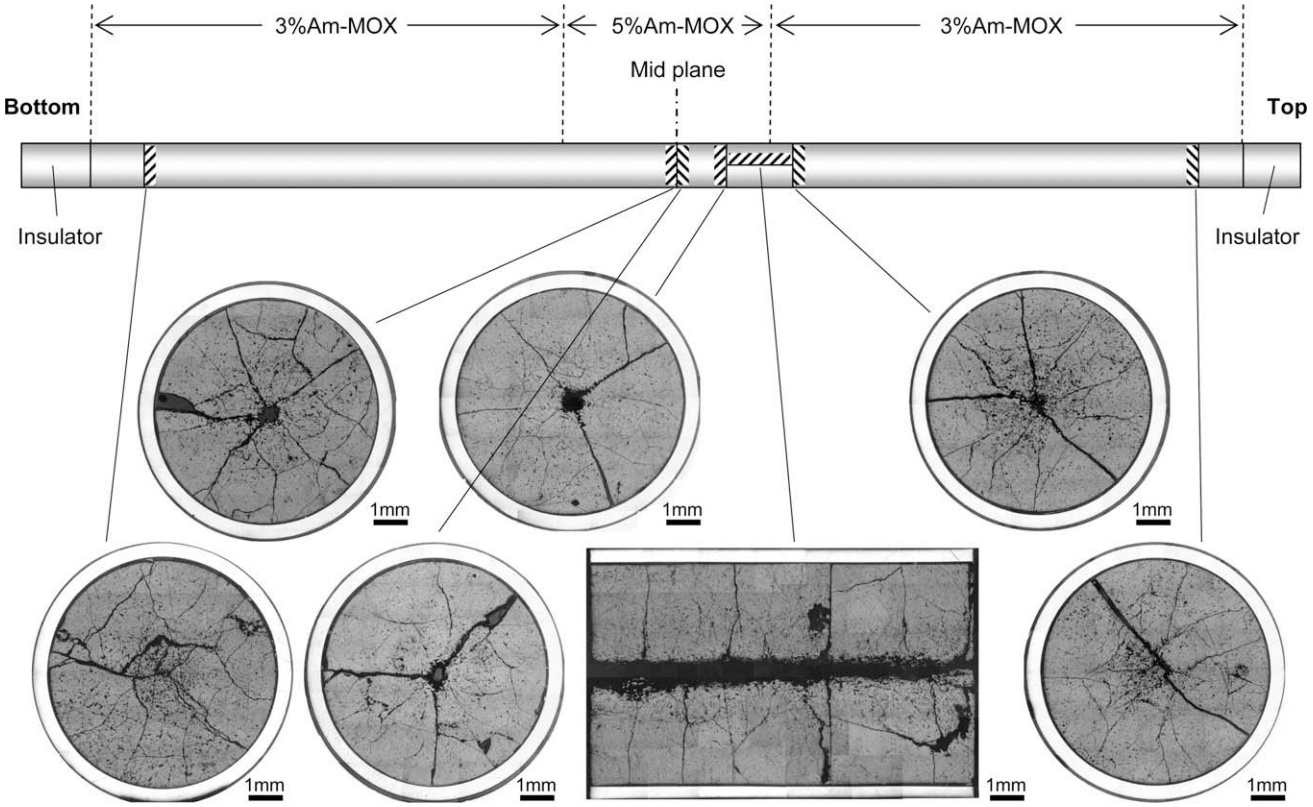

Fig. 3. Ceramographs of Am-MOX fuel pellets.

core. The burn-ups for two samples of 3% Am-MOX and one sample of 5% Am-MOX were determined by chemical analysis based on the isotope dilution method [5] in the AGF. Fixed linear heating rate was evaluated using fission rate derived from chemical analysis and fission energy of each nuclide [6]. The determined linear heating rate agreed well with the calculated value [7] within an experimental error of 3%.

2.3. Post irradiation examinations

After the Joyo irradiation, the fuel subassembly B11(1) was transported to the FMF adjacent to Joyo to carry out non-destructive PIEs including X-ray CT scans [8], X-ray radiography, weight measurements, visual inspections, cladding outer diameter profilometry, and gamma scanning. The results of non-destructive PIEs showed that the Am-MOX fuel pin was irradiated without any failure as expected. After the non-destructive PIEs, the fuel pin was punctured and sectioned to cut off the upper and lower ends of the fuel pin which were then transported to the AGF for destructive PIEs.

The Am-MOX fuel pin was cut at different axial positions into several segments. Six cross-sectional specimens were prepared by the following processes; each segment (about 20 mm in length) was impregnated with epoxy resin in vacuum and cut transversely

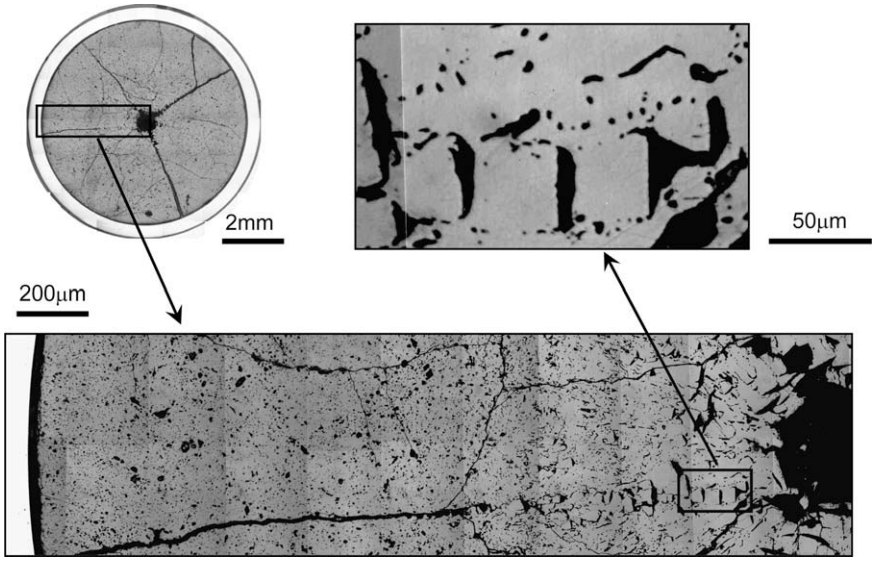

Fig. 4. Enlarged ceramographs of Am-MOX fuel pellets.

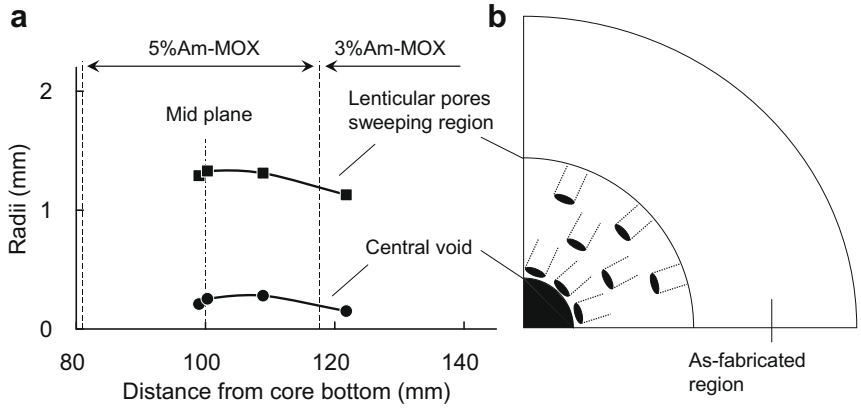

Fig. 5. Restructuring evolution along the axial direction.

into discs of about 5 mm in thickness. These discs were mounted into holders using epoxy resin, and then were ground and mirror-polished with anhydrous lubricant. One longitudinal specimen was taken from the axial region; it included a boundary between a 5% Am-MOX fuel pellet and an upper part of a 3% Am-MOX fuel pellet and it was also prepared by the processes mentioned above. The microstructure was observed with an optical microscope (TELATOM, Reichert). In order to investigate development of the restructuring region quantitatively, image analysis was carried out on optical micrographs.

EPMA of U, Pu and Am was done using the SX-100R model (Cameca) which was specially shielded with lead and tungsten to permit the analysis of irradiated nuclear fuels. An electron acceleration potential of 25 kV and a beam current of 50 nA were selected for the analysis. Three wavelength dispersive spectrometers with pentaerythritol (PET) diffracting crystals were used for the analysis simultaneously. Selected characteristic X-rays were M_α lines for U and Am, and the M_β line for Pu. Qualitative image mapping of X-ray intensities and quantitative point analysis were conducted on polished specimens. Calibration data of U, Pu and Am were obtained using a non-irradiated Am-MOX fuel pellet containing 3% Am and the raw X-ray data were converted to wt% using the Cameca matrix correction program based on the PAP model [9]. The quantitative measurements were made away from pores and cracks. The secondary electron image was used to obtain information about the fuel microstructure at the locations selected for analysis and to position the electron beam. Immediately prior to EPMA of the samples a thin film of carbon was applied to their surface by vacuum evaporation to avoid electron charging effects.

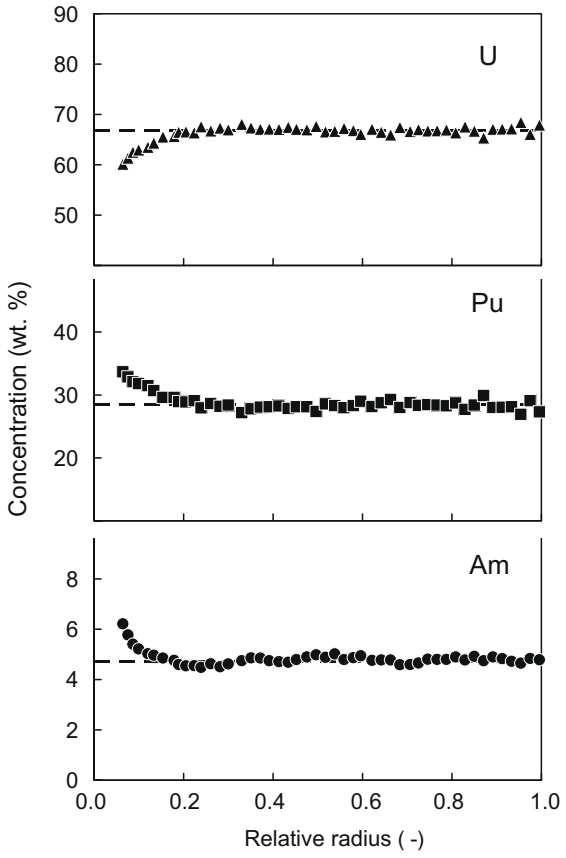

Fig. 6. Radial distributions of U, Pu and Am.

3. Results and discussion

3.1. Microstructures

Fig. 3 shows the as-polished macroscopic ceramographs of Am-MOX fuel pellets. Some cracks were observed along the radial and circumferential directions. A central void formed on the specimens taken around the axial mid position of the fuel pin. Fig. 4 gives enlarged views of the microstructure focusing on the features of the fuel restructuring behavior. Plano-convex-shape lenticular pores were observed around the central void. Lenticular pores were also found in the vicinity of cracks which developed in the fuel due to thermal stresses. Crack healings occurred in some parts of the fuel pellets after the lenticular pores moved to the fuel center along the temperature gradient. Closer inspection of the trails behind the lenticular pores in Fig. 4 showed that many of them consisted of a string of small spheres. The configuration of the lenticular pores was similar to that of conventional oxide fuels irradiated for a short-term at the high linear heating rate condition [10–12]. No significant difference in the restructuring feature was observed between the 5% Am-containing and the 3% Am-containing MOX fuel pellets. The radial cut specimens revealed no central void for upper and lower end parts of the fuel pin. Although no restructuring was found on the lower part of the fuel pin, the formation of a small number of lenticular pores was observed in the vicinity of the cracks on the upper part of the fuel pin. As shown in Figs. 3 and 4, fuel restructuring had started in the Am-MOX fuel pellets even in the brief irradiation time of 10 min.

Fig. 5(a) shows the restructuring evolution behavior of the cross-sectional specimens. The radii were converted from the area

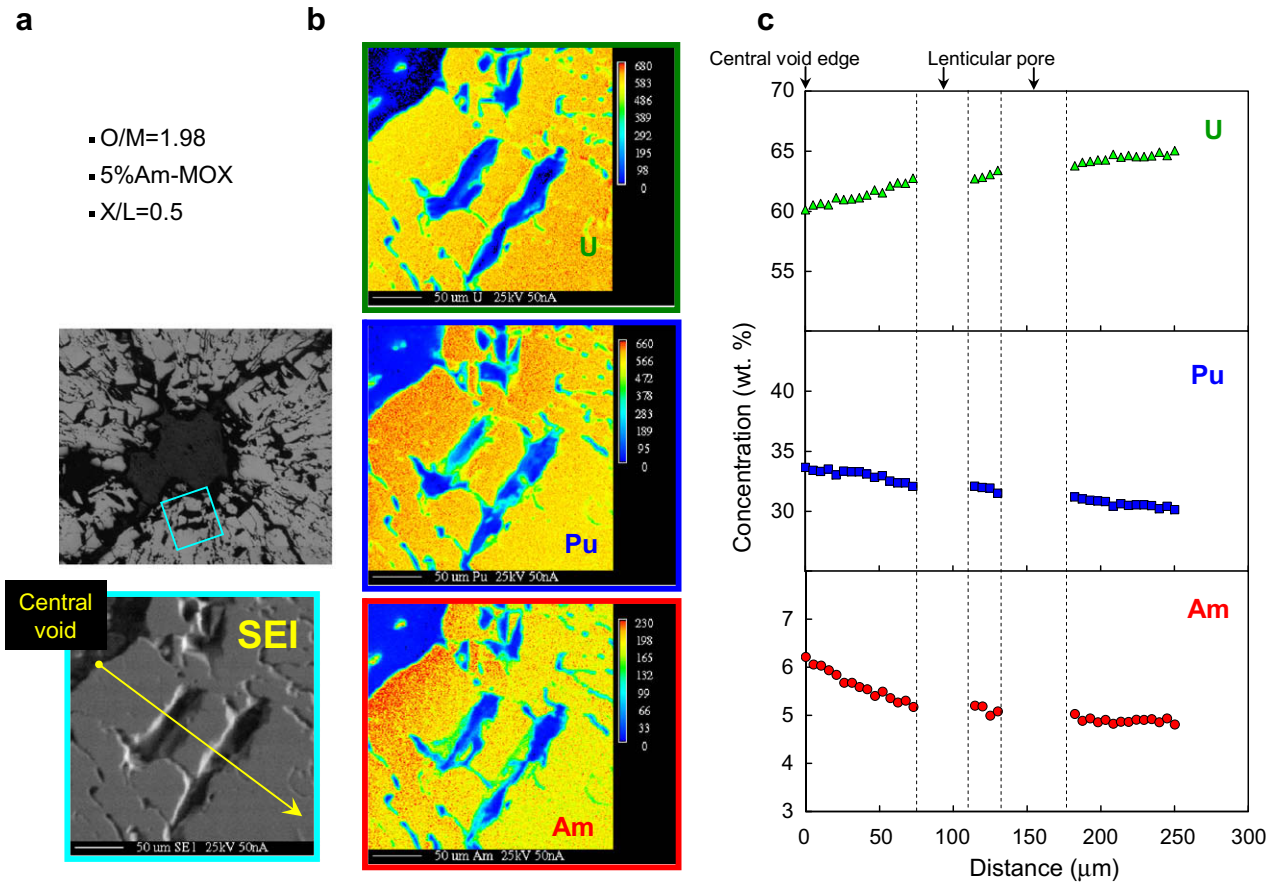

Fig. 7. Radial distributions of U, Pu and Am in the vicinity of the central void.

of the central void and lenticular pore sweeping region shown in the Fig. 5(b). The differences observed in the size of the central void and lenticular pore sweeping region can be explained by the axial distribution of linear heating rate.

In order to confirm whether or not fuel melting occurred at the high linear heating rate, careful observations were carried out. It has been reported that when the fuel melted at the initial stage of irradiation, a pore-free structure was observed in the center of the fuel pellet or around the central void [13,14]. No sign of fuel melting was found in any of the specimens taken from the Am-MOX fuel pin irradiated for 10 min. This implied that the thermal design for this test had sufficient safety margin. No sign of FCCI was observed either.

3.2. Radial distributions of U, Pu and Am

Fig. 6 shows the radial distributions of U, Pu and Am on the specimen taken from the axial mid position of the fuel pin. The concentration of these elements from the middle to outer part of the fuel pellets was almost the same as the chemically analyzed value at the fabrication stage. In the vicinity of the central void the concentrations of Pu and Am increased whereas that of U decreased. The radial distributions of U, Pu and Am on the specimens taken from upper and lower end parts of the fuel pin were quite flat and no apparent redistribution was found. The tendency of appearance or non-appearance of redistribution agreed with the fuel restructuring behavior which could be explained by the axial distribution of linear heating rate.

Fig. 7(a) shows the optical and secondary electron images indicating the position measured by EPMA for acquiring a detailed elemental distribution. The radial position which was subjected to EPMA was adjacent to the central void and included some lenticular pores. Fig. 7(b) shows the qualitative characteristic X-ray mapping of U, Pu and Am. As shown in this figure, the concentrations of Pu and Am around the central void were relatively higher than in the outer part of the pellet. It was obvious from Fig. 7(c) that the concentrations of Pu and Am gradually increased on approaching the edge of the central void (the line for analysis corresponds to the arrow indicated in the secondary electron image shown in Fig. 7(a)). A slight increase in Am concentration at the center of the fuel pellet has been reported in the Am-MOX fuel in the SUPER-FACT program [15].

A quick comparison between Pu and Am distribution profiles shown in Figs. 6 and 7 indicated that both profiles were similar. Generally, ^{241}Am accumulates in Pu-containing fuels by the beta decay of ^{241}Pu. Therefore, careful attention should be paid for evaluating the amount of Am concentration in MOX fuels. According to calculation results using the ORIGEN2 code, the change in concentrations of Am and Pu was confirmed to be negligible during the short-term irradiation and the storage period up to the time of the EPMA analysis. That is to say, Am behaved like Pu during the initial stage of irradiation. As shown in Figs. 3 and 4, initial fuel restructuring was not complete in this fuel pin. It was considered that the increase in concentrations of Pu and Am in the center of the fuel pellet could be attributed to vapor transport of U during the evaporation–condensation mechanism of the pores.

At the center of the pellets the concentration of Pu reached about 34 wt%; an increase of almost 18% over the as-fabricated Pu concentration of 29 wt%. And 6.2 wt% Am was measured at the center of the pellet, which compared with the as-fabricated Am concentration of 4.7 wt% and corresponded to an increase of about 32%. This implied that Am had a tendency to accumulate rapidly at the center

of the fuel pellet compared to Pu. The concentration gradient of Am along the radial direction of the pellet was steeper than that of Pu. It is well-known that the local concentration variations of Pu due to migration affect the fuel thermal properties in MOX fuel. It has been reported that the melting temperature is decreased by the addition of a large amount of Am [16,17], which leads to a smaller margin for fuel melting. The present EPMA results indicated that careful consideration must be given to the redistribution behavior of Am as well as that of Pu for evaluating the impact on the thermal performance of Am-MOX fuels.

4. Summary

The Am-MOX fuel pins containing 3% or 5% Am were irradiated at about 43 kW m^{-1} for 10 min in the experimental fast reactor Joyo. Subsequently, one of them was subjected to PIEs and the microstructural evolutions in the fuel pellets were observed by optical microscopy and redistribution behavior of constituent elements was determined by EPMA. The ceramography results showed that structural changes such as formation of the lenticular pores and the central void occurred early, within the brief 10 min of irradiation. No signs of fuel melting were found in any of the fuel pellet specimens. The EPMA results revealed that the concentrations of Pu and Am increased whereas that of U decreased in the vicinity of the central void.

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