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Short-term irradiation behavior of minor actinide doped uranium plutonium mixed oxide fuels irradiated in an experimental fast reactor

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ABSTRACT

A fuel irradiation program is being conducted using the experimental fast reactor 'Joyo'. Two short-term irradiation tests in the program were completed in 2006 using a uranium and plutonium mixed oxide fuel which contains minor actinides (MA-MOX fuel). The objective of the tests is the investigation of early thermal behavior of MA-MOX fuel such as fuel restructuring and redistribution of minor actinides. Three fuel pins which contained MA-MOX: 2% neptunium and 2% americium doped uranium plutonium mixed oxide $(Am,Pu,Np,U)O_{2-x}$ fuel were supplied for testing. The first test was conducted with high-linear heating rate of approximately 430 W cm^{-1} for only 10 min. After the first test, one fuel pin was removed for examinations. Then the second test was conducted with the remaining two pins at nearly the same linear power for 24 h. In these tests, two oxygen-to-metal molar ratios were used for fuel pellets as a test parameter. Non-destructive and destructive post-irradiation examinations results are discussed with early on the behavior of the fuel during irradiation.

1. Introduction

In the present once through nuclear fuel cycle, minor actinides (MAs) which have been generated in light water reactor (LWR) fuels remain in the high-level radioactive waste. The long-term geological disposal of the MAs of this high-level waste may affect the environment. In order to reduce this environmental concern, their separation by reprocessing and their transmutation in reactors should be technically established. One of the options transmutation is to irradiate uranium and plutonium mixed oxide (MOX) fuel containing some MAs (MA-MOX fuel) in fast reactors (FRs). One nuclear fuel cycle strategy proposes that the MAs such as Np and Am produced from LWRs are burnt as a 5% MA-MOX fuel in FRs [1]. Fabrication techniques for small amounts of MA-MOX fuel on a laboratory scale have already been developed [2]. However, the design of MA-MOX fuel still involves large uncertainties due to the very limited information on their thermal and mechanical properties and of insufficient data on their irradiation behavior [3]. To fill these knowledge gaps, a MA-MOX fuel irradiation program (Am-1) [4] is being conducted in the experimental fast reactor Joyo. This irradiation program is divided into a short-term phase to investigate the early thermal behavior of MA-MOX fuel and a long-term phase to determine the burn-up dependant irradiation behavior. In this irradiation program, $(Am,Pu,Np,U)O_{2-x}$ fuel and $(Am,Pu,U)O_{2-x}$ fuel as representative MA-MOX fuel were fabricated. This paper describes the results of post-irradiation examination (PIE) for $(Am,Pu,Np,U)O_{2-x}$ fuel used in the short irradiation tests.

2. Experimental

2.1. Short irradiation tests

The Am-1 program consists of short irradiation tests and long steady-state irradiation tests. Two short irradiation tests were conducted in the fast experimental reactor Joyo in 2006. In one test, irradiation of fuel pins was carried out for 10 min to check mainly whether or not fuel melting had occurred. Fuel melting was not actually expected in the test, but due to the large safety margins derived from the limited physical property data in the thermal design of the fuel pins, the maximum linear heating rate for the tests was determined as 430 W cm^{-1}, which corresponded to the limitation of maximum fuel melt fraction of 20%. One $(Am,Pu,Np,U)O_{2-x}$ fuel pin was removed from an irradiation vehicle [4] for PIE. The remaining two fuel pins were irradiated again in Joyo for 24 h to investigate the radial redistribution behavior of MAs.

2.2. Fuel fabrication

The nominal compositions of the $(Am,Pu,Np,U)O_{2-x}$ fuel and the reference MOX fuel were $(Am_{0.02},Pu_{0.29},Np_{0.02},U_{0.67})O_{2-x}$ and

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$(U_{0.7},Pu_{0.3})O_{2-x}$, respectively. Two O/M molar ratios were chosen at 1.95 (low) and 1.98 (high). The fuel pellet diameter was 5.42 mm and pellet height was 8 mm. The fuel relative density was 93% of the theoretical density (TD). Enrichment of ^{235}U and Pu were 8.3 and 30 wt%, respectively. A large amount of Si was contained in the $(Am,Pu,Np,U)O_{2-x}$ fuel pellet as a metallic impurity, but the concentration was less than 1100 ppm which satisfied the fuel specification (<1400 ppm). It is believed that the Si impurity was due to the silicon-lined ball mill uses to crush the raw powder.

The fuel pellets were inserted into the cladding tubes of 20% cold worked PNC-316 along with insulator pellets, a spring, etc. The cladding tube diameter and its wall thickness were 6.5 and 0.47 mm, respectively. The length of the fissile column was 200 mm and is shorter than the Joyo driver fuel (500 mm). The $(Am,Pu,Np,U)O_{2-x}$ fuel pellets were loaded into the middle part of the fuel column and MOX fuel pellets as a reference were loaded into the rest of the fuel column. The $(Am,Pu,Np,U)O_{2-x}$ fuel column length differed in the first and second test, being 40 mm (z/L: 0.4–0.6) and 72 mm (z/L: 0.32–0.68), respectively. Here, z and L stand for the distance from the bottom of the fissile column and the fissile column length, respectively.

The pellets were fabricated in a normal alpha particle-tight glove box facility at Nuclear Fuel Cycle Engineering Research Institute. The grid spaced fuel pins were separately loaded in the irradiation vehicle at Oarai Research and Development Center.

2.3. Irradiation conditions

For the first test, three fuel pins were loaded in the irradiation vehicle for continuous irradiations in the reactor. The reactor thermal power was continuously raised at 12 MW h^{-1} (10% power thermal per hour) from 60 to 120 MW. At the maximum power level, the reactor power was held for 10 min, then the reactor was shutdown rapidly by a manual scram in order to preserve the fuel microstructure and to prevent additional fuel restructuring. After the first test, one fuel pin was removed for PIE. The other two fuel pins in the irradiation vehicle were re-loaded in the reactor. In the second test, the reactor power was raised just as in the first test, and then the reactor was held at the maximum power level for 24 h, after which the shutdown by normal means was carried out. In both short irradiation tests, the maximum linear heating rate and local burn-up of the $(Am,Pu,Np,U)O_{2-x}$ fuel were approximately 430 W cm^{-1} and 0.0082 at.%, respectively.

2.4. Post-irradiation examinations

After each irradiation test in Joyo, the irradiation vehicle was transferred to the hot laboratory and disassembled. Non-destructive examinations (NDE) included visual inspection, cladding outer diameter profilometry, and gamma scanning, weight measurements, X-ray radiography and X-ray computer tomography for all test fuel pins. Hereby, the proper performance of the irradiation vehicle was ensured. After the NDE, the test fuel pins were punctured and sectioned into samples for optical microscopy, electron probe microanalysis (EPMA), and burn-up measurements by destructive examination (DE).

3. Results and discussion

3.1. Destructive examination for the first test samples

There were no indications of fuel melting in the X-ray computer tomography and radiography. The fuel pin was then sectioned for ceramography. Samples were removed from the $(Am,Pu,Np,U)O_{2-x}$ fuel and the MOX fuel as a reference above and below the $(Am,Pu,Np,U)O_{2-x}$ fuel column. Axial positions of samples removed from the fuel pin are expressed using such terms as z/L.

Fig. 1 shows ceramographs obtained for the fuel pin removed after the first test. Fuel restructuring had already started within the brief 10 min irradiation, but no apparent fuel melting was observed. In the case of the $(Am,Pu,Np,U)O_{2-x}$ fuel ($z/L = 0.50$), lenticular voids, nucleated from pre-existing sintering pores were not observed. Instead of lenticular voids, a number of spherical and tubular pores nucleated in the columnar grains of the $(Am,Pu,Np,U)O_{2-x}$ fuel were observed. Thus, the typical columnar

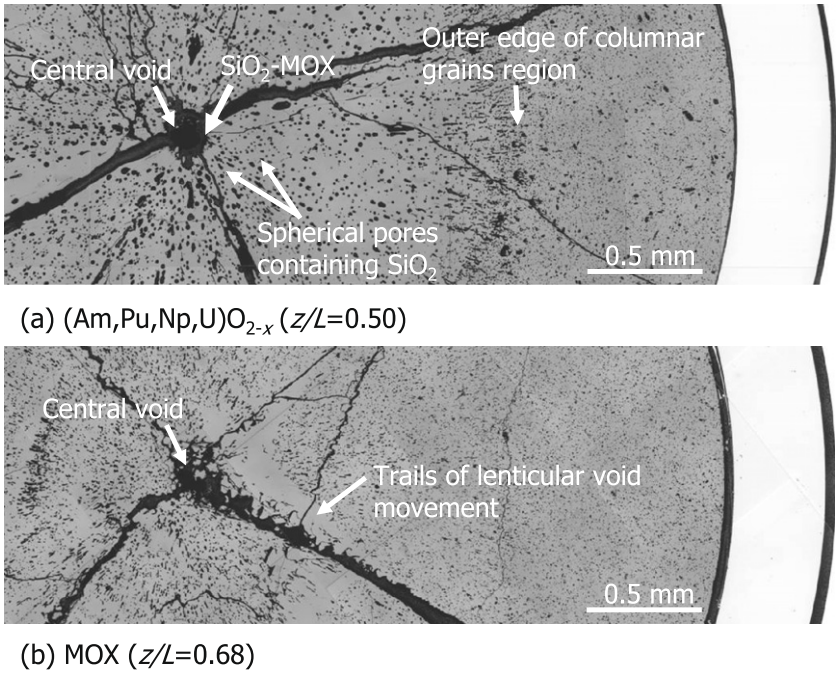

Fig. 1. Comparison of fuel microstructure between the $(Am,Pu,Np,U)O_{2-x}$ fuel ($z/L = 0.50$, 427 W cm^{-1}) and the reference MOX fuel ($z/L = 0.68$, 416 W cm^{-1}) in the first test (O/M molar ratio 1.98).

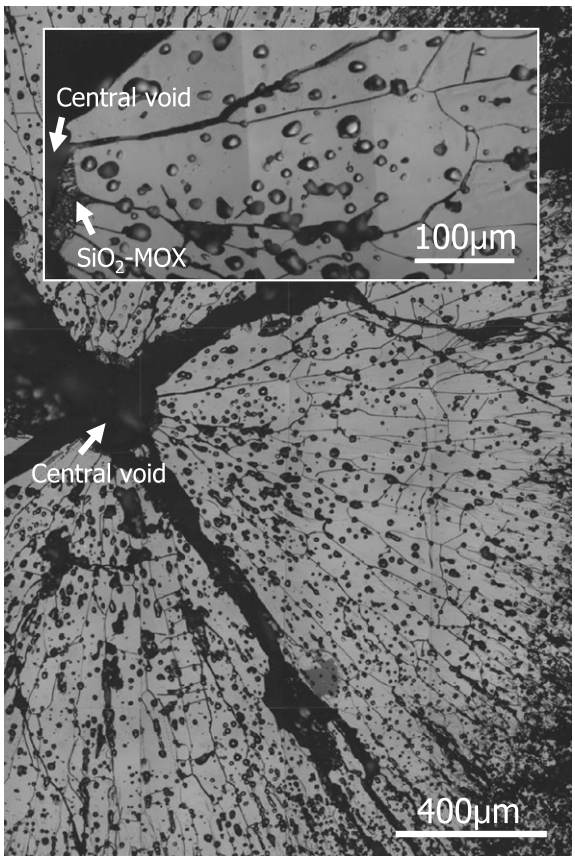

Fig. 2. Fuel microstructure of the chemically etched $(Am,Pu,Np,U)O_{2-x}$ (first test, $z/L = 0.50$, O/M molar ratio 1.98).

grain structure was generated despite the absence of lenticular voids. The degree of the relocation was not significant because gap closure occurred easily due to the small as-fabricated gap which was 140 μm. It should be considered, however, that a certain contribution to the central void volume by fuel relocation was included. In the case of the reference MOX fuel ($z/L = 0.68$), generation of a small central void and relocation of the fuel pellet by fuel cracking were also seen. In addition some trails of lenticular void movement were developed from the fuel cracks. From the image analysis, the radial position of the nucleated spherical pores and lenticular voids were approximately the same for the samples. This suggests that the restructuring of $(Am,Pu,Np,U)O_{2-x}$ fuel occurred faster than the reference MOX fuel due to the movement of spherical and tubular pores.

Fig. 2 shows the microstructure of the $(Am,Pu,Np,U)O_{2-x}$ fuel ($z/L = 0.50$) which was chemically etched in order to investigate the grain boundaries. It was found that the some grains were surrounded by grayish inclusions which were identified as a SiO_2-rich compound by EPMA. The spherical and tubular pores were coalesced and elongated towards the fuel center on the grain boundaries. Also some small pores were seen in the large grains. The spherical pores tended to migrate through grain boundaries, while the tubular pores were formed by pore interlinking during the migration. Such behavior has been reported before [5].

3.2. Destructive examination for the second test samples

The same NDEs were carried out as for the first test samples. Noting unexpected that could have influenced the fuel behavior was recognized. Then, both fuel pins were punctured and sectioned for ceramography.

The ceramographs of samples removed from the fuel pin with O/M molar ratio of 1.98, are shown in Fig. 3. In the restructured region of the $(Am,Pu,Np,U)O_{2-x}$ fuel ($z/L = 0.50$), columnar grains were developed completely. The central voids, about 1 mm in diameter, were formed. Neither the spherical nor the tubular pores were observed around the central void and in the restructured region. At the site of the crack opening, grayish inclusions of a SiO_2-rich compound, along with a shallow attack of the inner wall of the

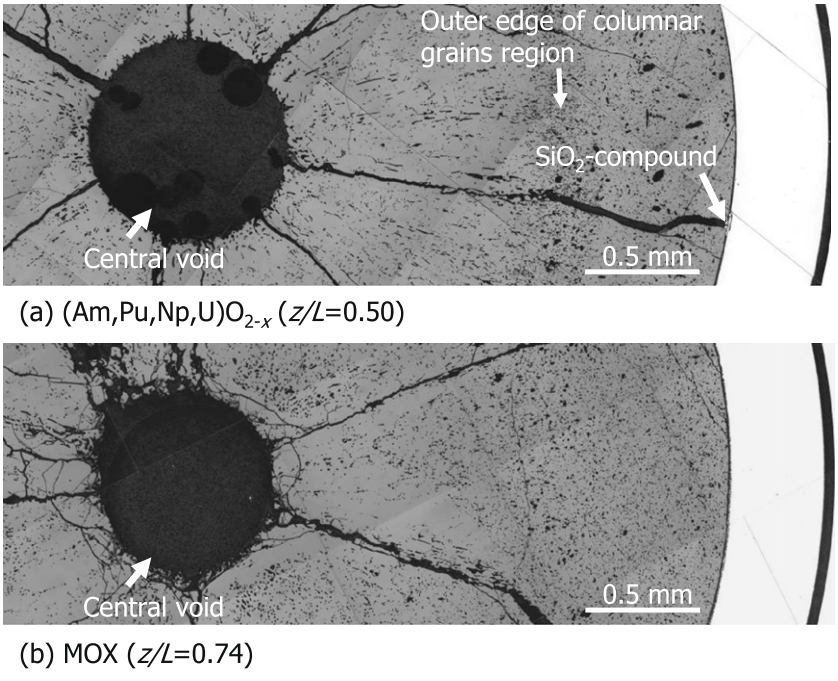

Fig. 3. Comparison of fuel microstructure between the $(Am,Pu,Np,U)O_{2-x}$ fuel ($z/L = 0.50$, 432 W cm^{-1}) and the reference MOX fuel ($z/L = 0.74$, 420 W cm^{-1}) in the second test (O/M molar ratio 1.98).

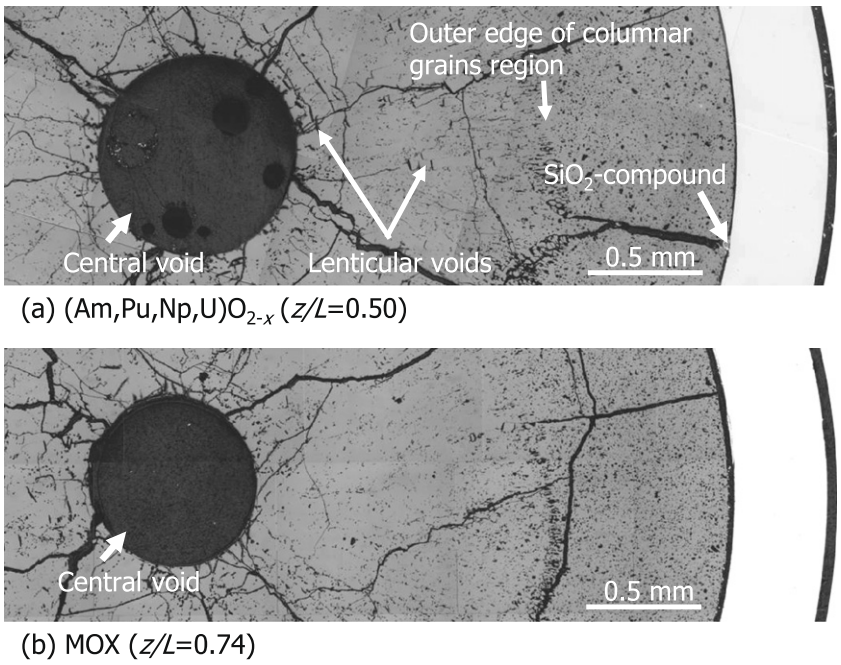

Fig. 4. Comparison of fuel microstructure between the $(Am,Pu,Np,U)O_{2-x}$ fuel (z/L = 0.50, 429 W cm^{-1}) and the reference MOX fuel (z/L = 0.74, 417 W cm^{-1}) in the second test (O/M molar ratio 1.95).

cladding were observed. In the second test, a SiO_2-rich compound was not seen in the restructured region, but a small amount deposited at the crack opening.

The ceramographs of samples removed from the fuel pin with an O/M molar ratio of 1.95, are shown in Fig. 4. In the restructured region of the $(Am,Pu,Np,U)O_{2-x}$ fuel (z/L = 0.50), neither spherical nor tubular pores were seen. Instead of the pores, some lenticular voids were still migrating towards the central region, and the columnar grains were not completely developed yet. At the site of the crack opening, grayish inclusions and a shallow attack of the inner wall of the cladding were seen just as for the fuel pin with an O/M molar ratio of 1.98.

The equivalent diameters of the restructured regions were determined by image analysis and are shown in Fig. 5 as a function of the distance from the bottom of the fissile column. Comparison of the $(Am,Pu,Np,U)O_{2-x}$ fuel samples which had fuel O/M molar ratios of 1.95 and 1.98, showed slight differences in the diameters of the columnar grains and the central void, respectively. This suggested that the fuel restructuring of the $(Am,Pu,Np,U)O_{2-x}$ fuel with an O/M molar ratio of 1.95 was almost finished, as the central void is generally formed by migration of pre-existing sintering pores up the thermal gradient. Thus the fuel O/M molar ratio had little influence on fuel restructuring behavior. Taking the axial distribution of linear heating rate into consideration, the rather pronounced difference in fuel restructuring between the $(Am,Pu,Np,U)O_{2-x}$ fuel and the reference MOX fuel was reasonably understood.

3.3. Radial redistributions of MAs and Pu

The radial intensity profiles of MAs and Pu for the $(Am,Pu,Np,U)O_{2-x}$ fuel (z/L = 0.50) of the first test are shown in Fig. 6. The Np profile was almost flat, but Pu and Am were slightly redistributed in the columnar grains region. Local depletions of Pu

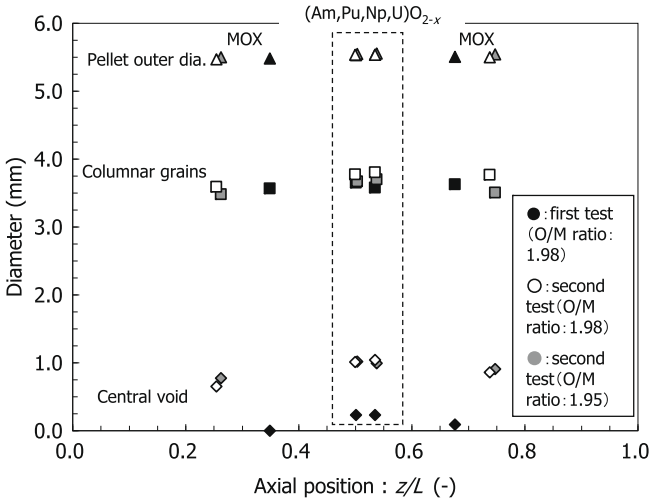

Fig. 5. Axial distributions of the restructured regions in the samples (two short tests, O/M molar ratio 1.98 and 1.95).

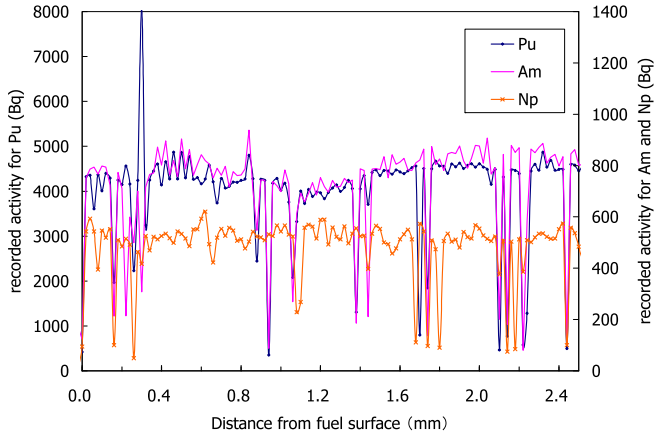

Fig. 6. Radial intensity profiles of Np, Am and Pu in the sample of the $(Am,Pu,Np,U)O_{2-x}$ fuel (first test, z/L = 0.50, O/M molar ratio 1.98).

and Am were identified in the vicinity of the outer edge of the columnar grains, and were attributed to vapor transport of uranium as UO_3 down the radial temperature gradient during the irradiation [6].

The radial intensity profiles of MAs and Pu for the samples with an O/M molar ratio of 1.98 of the second test are shown in Fig. 7. The profile of Np in the $(Am,Pu,Np,U)O_{2-x}$ fuel sample was remained constant after the fuel restructuring. In contrast, the Pu and Am concentration increased at the fuel center and were redistributed in the columnar grains region. The radial profiles of Pu and Am content in the $(Am,Pu,Np,U)O_{2-x}$ fuel was also depleted in the vicinity of the outer edge of the columnar grains. In addition, EPMA detected islands rich in Am in the region up to 0.88 mm from the surface, and were introduced in the fuel matrix during the fabrication. The redistribution of Pu was also observed in the reference MOX fuel sample, and was similar to Pu in the $(Am,Pu,Np,U)O_{2-x}$ fuel sample. In comparison with the $(Am,Pu,Np,U)O_{2-x}$ fuel sample of the first test, the depletions of Am and Pu content at the outer edge of the columnar gains region were pronounced, and Am and Pu content increased towards the central void. The Am and Pu content profiles were distributed similarly as a function of fuel radius. The redistribution behavior of Am and Pu from the onset through to the end of fuel restructuring seemed to be similar.

The radial intensity profiles of MAs and Pu for the samples with an O/M molar ratio of 1.95 of the second test are shown in Fig. 8. Profiles of Np, Am and Pu in the $(Am,Pu,Np,U)O_{2-x}$ fuel were similar in the fuel with an O/M molar ratio of 1.98, but the degree of the redistribution of Am and Pu was smaller than that of fuel with an O/M molar ratio of 1.98. This was attributed to a lower vapor transport of uranium as UO_3, a consequence of the low oxygen potential of the fuel with an O/M molar ratio of 1.95. In this low oxygen potential condition, the radial profiles of Am and Pu also exhibit a similar tendency to redistribute in the columnar grains region. The redistribution of Pu was also observed in the reference MOX fuel sample. The degree of redistribution of Pu in the reference sample was also similar with that of Pu in the $(Am,Pu,Np,U)O_{2-x}$ fuel sample.

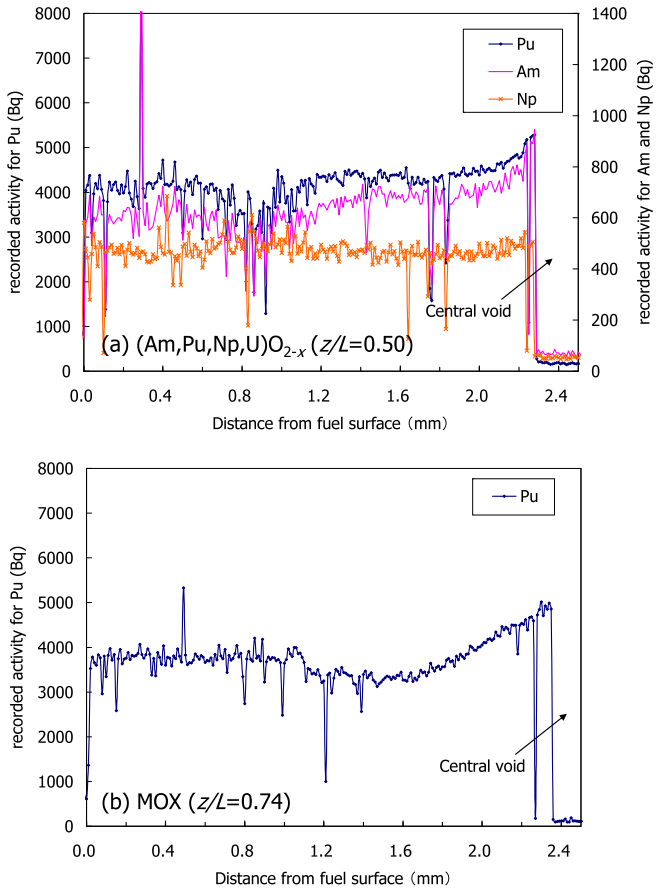

Fig. 7. Radial intensity profiles of Np, Am and Pu in the samples of the $(Am,Pu,Np,U)O_{2-x}$ fuel and the reference MOX fuel (second test, O/M molar ratio 1.98).

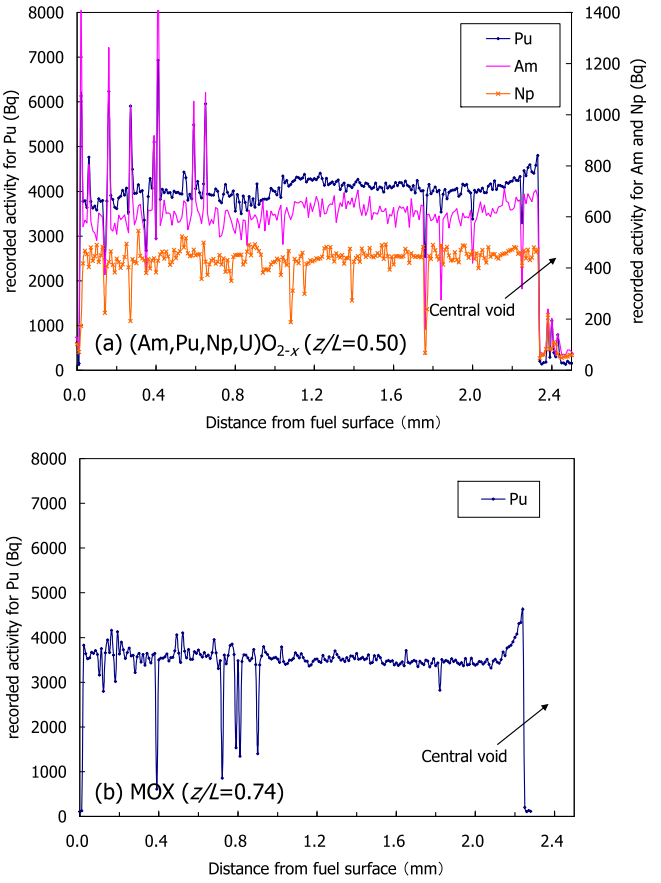

Fig. 8. Radial intensity profiles of Np, Am and Pu in the samples of the $(Am,Pu,Np,U)O_{2-x}$ fuel and the reference MOX fuel (second test, O/M molar ratio 1.95).

These results reveal that the radial profiles of Am and Pu show a similar tendency to redistribute in the columnar grain region, and that small addition of MAs did not alter the redistribution characteristics of U and Pu from those of the MOX fuel without MAs. The magnitude of fuel restructuring in the samples was dependent on the linear heating rate, but was independent of small MA additions. The radial redistribution of Am and Pu were, to a large extent, a direct consequence of the mechanisms generating the formation of the columnar grains and central void, in addition, a greater dependence of the fuel O/M molar ratio on the radial redistributions of Am and Pu was suggested.

Quantitative analysis of the samples of $(Am,Pu,Np,U)O_{2-x}$ fuel ($z/L = 0.50$) with the two fuel O/M molar ratios are shown in Fig. 9. Radial distributions of Am with O/M molar ratios of 1.98 and 1.95 were similar up to 1.8 mm from the fuel surface. The enrichment of Am near the central void, however, became abruptly larger in the sample with high-fuel O/M molar ratio. A similar dependence of the O/M molar ratio on radial distribution of Pu was seen in the samples, and is attributed to the O/M dependence on the vapor transport of uranium as UO_3 down the radial temperature gradient during the irradiation. The maximum enrichments

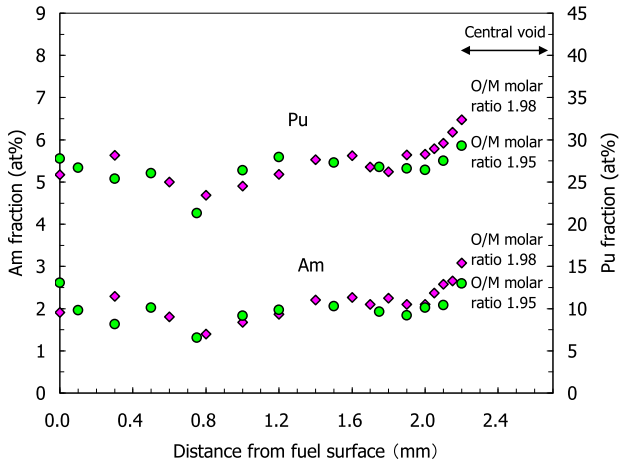

Fig. 9. Comparison of radial distributions of Am and Pu in the $(Am,Pu,Np,U)O_{2-x}$ fuel samples (second test, $z/L = 0.5$, O/M molar ratio 1.95 and 1.98).

of Am and Pu were 3.08 and 32.36 at.%, respectively, and are sufficiently low to suggest larger safety margins can be used in thermal design of the fuel pins.

4. Summary

PIE for two short irradiation tests of $(Am,Pu,Np,U)O_{2-x}$ fuel were successfully conducted. In the first test, $(Am,Pu,Np,U)O_{2-x}$ fuel showed no lenticular voids in the columnar grains region. This was due to the small as-fabricated fuel-cladding gap and possibly also due to the large amount of Si impurity which was retained as SiO_2 in the spherical and tubular pores during the fuel restructuring. This prevented the pores from developing lenticular voids.

In the second test, a fuel microstructure including SiO_2 as in the first test was not observed in the restructured region of the $(Am,Pu,Np,U)O_{2-x}$ fuel. Migration of lenticular voids was observed and the columnar grains were developed. In the $(Am,Pu,Np,U)O_{2-x}$ fuel, the Am and Pu increase towards the fuel center and indeed their entire radial intensity profiles were similar. Enrichment of Pu around the central void was almost the same in both the $(Am,Pu,Np,U)O_{2-x}$ fuel and the reference MOX case. The radial profiles of Am and Pu indicate that the elements had a similar tendency to redistribute in the columnar grain region, and that small addition of MAs did not alter the redistribution characteristics of U and Pu from the $(Am,Pu,Np,U)O_{2-x}$ fuel without MAs. Comparison of the $(Am,Pu,Np,U)O_{2-x}$ fuel and the reference MOX fuel showed that the degree of fuel restructuring had a greater dependency on the linear heating rate than on the small addition of MAs and the fuel O/M molar ratio. In contrast, the enrichment of Am and Pu around the central void showed a significant dependence on the O/M molar ratio.

Some of the results have indicated that the thermal design for the $(Am,Pu,Np,U)O_{2-x}$ fuel had more than sufficient safety margins. The fuel behavior at the high-linear heating rate obtained from the short irradiation tests will be used to verify the thermal performance of the $(Am,Pu,Np,U)O_{2-x}$ fuel modeled in fuel pin thermal analysis codes.

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