Application of FE-SEM to the measurement of U, Pu, Am in the irradiated MA-MOX fuel

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Abstract

It is important to make observations and elemental analyses of irradiated minor actinide-containing mixed oxide fuel (MA-MOX fuel) for the irradiation behavior investigation of MA-MOX fuel. However, irradiated MA-MOX specimens have high radioactivity and emit alpha-particles. In order to make detailed observations of microstructure and elemental analyses of irradiated MA-MOX fuel, a field emission scanning electron microscope (FE-SEM) equipped with a wavelength-dispersive X-ray spectrometer (WDX) was modified as follows.

1) To prevent leakage of radioactive materials, the instrument is attached to a remotely controlled air-tight specimen transfer unit between a shielded hot cell and the instrument.

2) To protect operators and the instruments from radiation, the instrument is installed in a lead shield box and the control unit is separately located outside the box.

By using the modified FE-SEM/WDX, MA-MOX specimens irradiated in Joyo were made observations and elemental analyses. As a result of observation by FE-SEM, microstructural changes were observed in irradiated MA-MOX specimens. As a result of elemental analysis, the characteristic X-rays peaks (U, Pu, Am) were detected by WDX successfully. By measuring the intensities of the characteristic X-rays, a quantitative analysis of U, Pu and Am along the radial direction of the irradiated MA-MOX specimens. Thereby, it was able to grasp the changes of microstructure and the change of U, Pu and Am radial distribution of irradiated MA-MOX fuel.

The technique has the great advantage of being able to evaluate the changes of microstructures and the changes of element distributions in MA-MOX fuel due to irradiation.

1. Introduction

In order to transmute minor actinide nuclides (MAs), the mixed oxide fuel doped with MAs (MA-MOX fuel) is irradiated in a fast reactor [1][2]. Therefore, it is important to measure the americium content in MA-MOX [3][4]. A field emission scanning electron microscope (FE-SEM), which uses a Schottky field emission electron gun, can obtain the image on the surface of specimen by higher magnification in the comparison with a traditional SEM [5][6]. In addition, FE-SEM has an electron beam current of high stability which can analyze the elements on surface.

In this study, the FE-SEM which can shield the radiation and prevent leakage of radioactive materials was developed and then the radial distribution of U, Pu, and Am was measured in the irradiated MA-MOX fuel specimens.

2. Instrument

The FE-SEM (JEOL JSM-7001F) is modified and installed in a shield box. In addition, the FE-SEM is equipped with wavelength-dispersive X-ray spectrometer (WDX) for the analysis of

elements on surface.

Figure 1 shows a schematic diagram of the modified FE-SEM, which was attached to the wall of the hot cell. In order to protect operators from radiation exposure and prevent leakage of radioactive materials, the following provisions are added, as shown in Fig. 1.

The FE-SEM is installed in a lead shield box. The control unit of the FE-SEM is separately installed outside the shield box, which is attached to a hot cell

The specimen transfer unit is located in the hot cell and an irradiated specimen is inserted in this unit and is supplied to FE-SEM through a port of hot cell by remote control. Maximum size of the specimen is 20 mm in diameter and 10 mm in height. The air tightness of the boundary between the operation room and the shielded hot cell is kept by an O-ring seal.

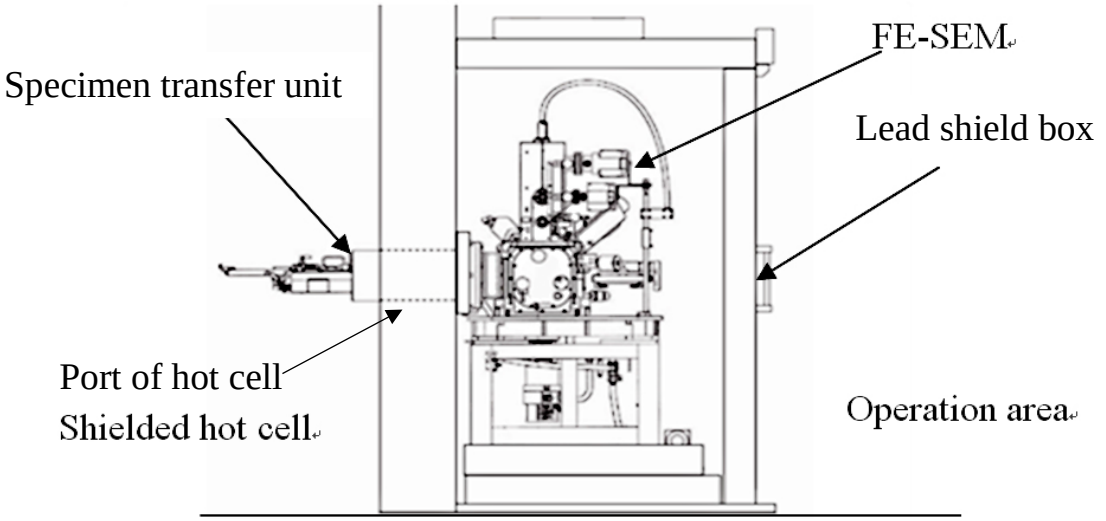

Figure 1. Schematic diagram of the modified FE-SEM.

3. Experimental

The MA-MOX fuel specimens were irradiated for short times in Joyo. These specimens were examined by above FE-SEM. Their specifications and irradiation conditions of MA-MOX fuel specimens are shown in Table 1.

Table 1. The specifications of MA-MOX fuel specimens and irradiation conditions

specimen	O/M(-)	Gap (dia.) (μm)	Liner heat rate (kW/m)	Burn up (MWd/t)
Q801531	1.98	210	48.5	450
Q80162	1.98	210	42.9	444
Q803532	1.98	160	47.7	449
Q803612	1.98	160	43	450
Q804532	1.96	160	47.4	448
Q80462	1.96	160	42.9	437
Q806531	2	210	47.6	451
Q80662	2	210	43	445

These specimens were cut into about 5 mm in height and mounted on holder (Φ20mm×10mm) by using epoxy resin. After mounting, the specimens were polished by emery papers and diamond pastes with kerosene, and subsequently were ultrasonically rinsed in a mixed solution of kerosene and ethanol. Finally, the specimens were coated with carbon. The maximum radiation dose rate of these specimens was 16.1 mSv/h. After preparation, the

specimens were transferred from the shielded hot cell to the FE-SEM.

4. Results and discussion

The specimens were observed and analyzed by the FE-SEM. After observations and elemental analyses, the specimens were transferred from the FE-SEM to the shielded hot cell. During their transfer, the radiation dose rate outside of the shield box is under 1μSv/h (the radiation dose limit of the operation area is 20μSv/h). The air-tightness of the shielded hot cell and the FE-SEM are maintained. There are no leaks of radioactive materials from the shielded hot cell and the FE-SEM.

Figure 2 shows secondary electron images of the fuel specimen surfaces. These images of the fuel specimen surface were taken at high magnification (2000X, 50000X). It is possible to identify the grain boundaries and cracks in detail (bright and dark areas correspond to the grain boundaries and cracks, respectively). The acceleration voltage, magnification, astigmatism, and specimen stage position can be well conditioned by remote control during the observation of fuel specimen.

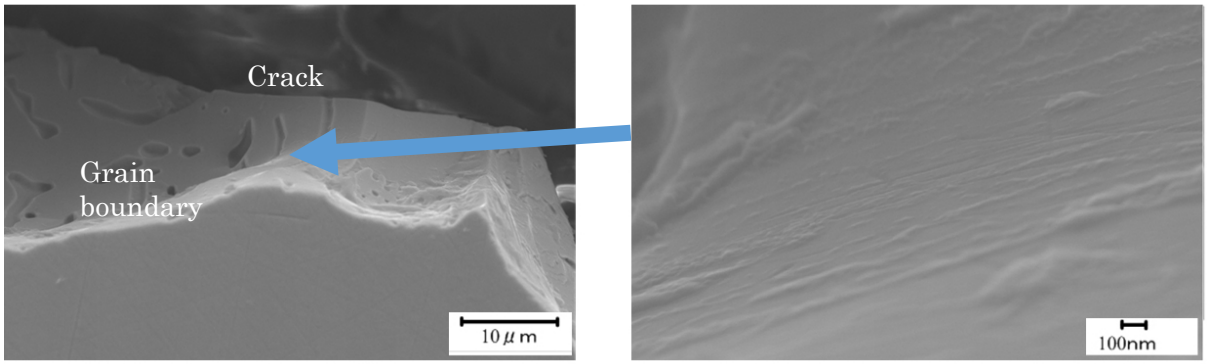

Figure 2. Secondary electron images of the MA-MOX fuel specimen surface (Accelerating voltage, 15kV, left-side image at 2000X and right-side Image at 50000X).

Figure 3 shows the results of elemental analyses of U, Pu and Am in an irradiated fuel specimen obtained by WDX. The characteristic X-ray peaks of U, Pu and Am are successfully detected.

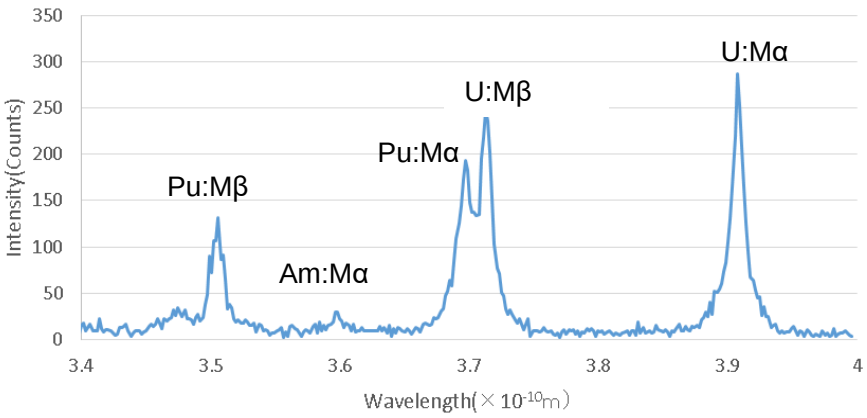

Figure 3. The characteristic X-ray peaks of U, Pu and Am(Accelerating voltage, 20kV).

The quantitative analyses of U, Pu and Am were obtained by measuring the intensities of the characteristic X-rays at many positions along the radial direction of the irradiated MA-MOX

specimens. The method of quantitative analysis is as follows.

1) The intensities of the characteristic X-ray are measured in non-irradiated MA-MOX fuel specimens which have low radioactivity and different concentration of U, Pu and Am.
2) The calibration curve is made from these measurements.
3) The intensities of the characteristic X-rays are measured along two diagonal lines in the irradiated MA-MOX fuel specimens.
4) The quantitative analyses are carried out using the calibration curve.

Figures 4-1 and 4-2 show the radial distributions of U, Pu, Am in irradiated MA-MOX specimens.

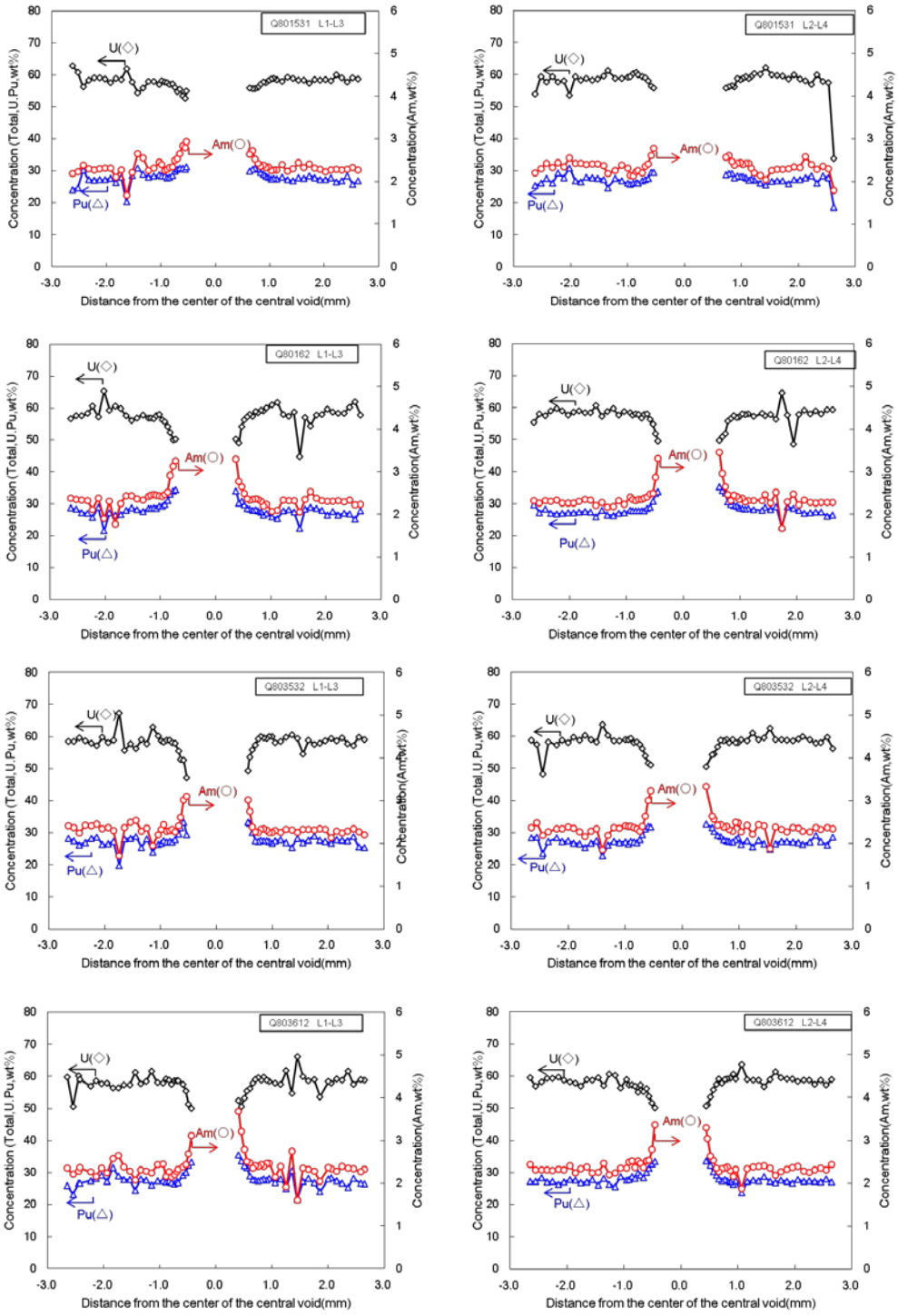

Figure 4-1. The radial distributions of U, Pu and Am along the radial direction of irradiated MA-MOX specimens (Q801531, Q80162, Q803532, Q803612).

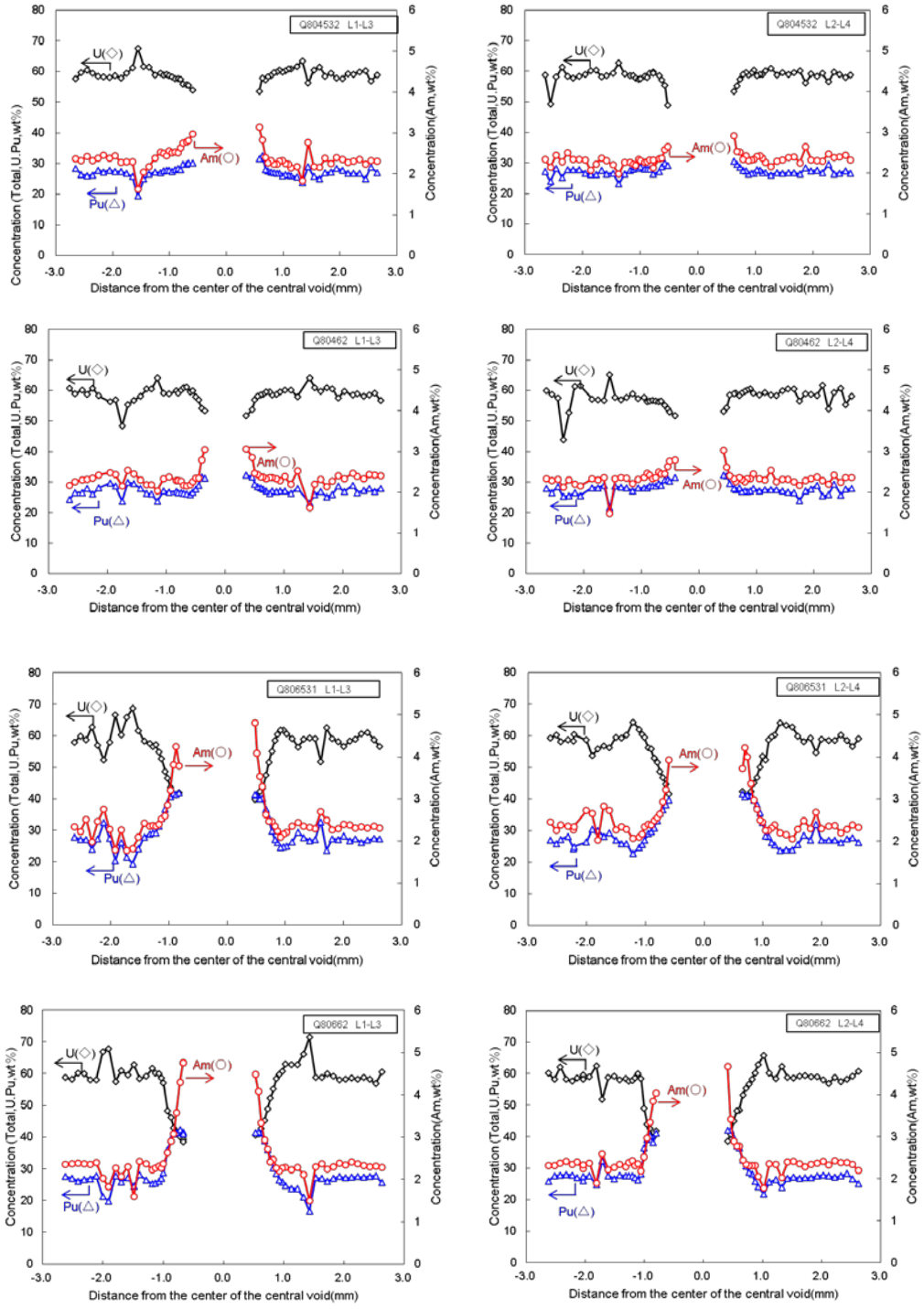

Figure 4-2. The radial distributions of U, Pu and Am along the radial Direction of irradiated MA-MOX specimens (Q804352, Q80462, Q806531, Q80662).

The concentrations of Pu and Am is higher in the region near the central void are higher than in the region near the cladding tube. The maximum concentrations are approximately 40 wt %(Pu) and 5wt%(Am). On the other hand, the concentration of U is lower in the region near the central void than in the region near the cladding tube area. The distribution changes of Pu and Am in high O/M specimens (Q806531, Q80662) are larger in the comparison with other specimens. Because pores in the fuel pellets migrate towards the fuel center by a steep temperature gradient during irradiation, the microstructural changes of fuel pellets and the distribution changes of fuel elements are occurred in the irradiation pellets by this steep

temperature gradient [7]-[11].

5. Conclusion

The modified FE-SEM was installed in the shield box and attached to the wall of a hot cell. This FE-SEM was applied to both observations of the microstructures and elemental analyses of irradiated MA-MOX fuel specimens. Thereby, it is able to grasp the changes of the microstructures and the quantitative distribution changes of U, Pu, Am along the radial direction. This technique has the great advantage of being able to evaluate irradiated MA-MOX fuel specimens in detail.

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