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Radial Distribution of U, Pu in a 55,000 MWd/MtU Spent Nuclear Fuel

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1. Introduction

At a high burn-up, nuclear fuel produces fission products proportional to a burn-up, and it creates highly porous and fine grain size micro structure at the pellet periphery by higher fission gas release. Among the burn-up dependant properties, the radial distribution of the fission products from core to rim of a spent fuel was our main concern. Currently, electron microprobe analysis (EPMA) is used for the spatial analysis of highly radioactive solid samples.[1] This method has advantages in high spatial resolution, but does not give isotopic information. In this work, ICP-MS was used as the tool for determining isotopic distribution of U, Pu in a spent fuel with burn-up of 55,600 MWd/MtU originating from the Yeonggwang-4 PWR reactor. The spatial information would be lost when a sampling method is based on the dissolution of a fuel. Thus, for the direct analysis of the isotopes and their radial distribution in a spent fuel, we developed a radiation shielded laser ablation system in conjunction with the ICP-MS system.[2-4] The analysis of isotopic ratio in a nuclear fuel was carried out by this radiation shielded LA-ICP-MS system.

2. Experimental

2.1 Instrumentation

The laser ablation system consisted of a Q-switched Nd:YAG laser, image analyzer, XYZ translator with motion controller, ablation chamber, and various optics.

This system was gamma shielded by a lead shield glove-box for the analysis of the radioactive material in a spent nuclear fuel. The ablation chamber, image analyzer, XYZ translator and optics are installed inside the shielded glove box while the laser system and electronics for remote control are installed outside. The laser beam is sent into the cell through a UV window on the rear door of the shielded glove box. The Laser ablation system was coupled to ICP-MS (Element, Finnigan) by PVC tubing with an argon flow of 1 L/min. Sampling was carried out by a Q-switched Nd:YAG laser at 266 nm. The ICP-MS system was optimized at maximum ion intensity for ^{238}U. The laser was operated at 45 % laser power (4mJ at 100%), 10Hz repetition rate and 2s sampling time. The ablated particles were carried to the plasma of the ICP-MS system through a PVC tube.

2.2 Preparation of sample specimen

For the analysis, a high burn-up spent nuclear fuel (J502-A14, 55,600 MWd/MtU) from the Yeonggwang-4 nuclear reactor was chosen as the test specimen. It was cut along the diameter of the pellet including the cladding by 3(w)x1(t) mm and embedded in epoxy resin and then polished..

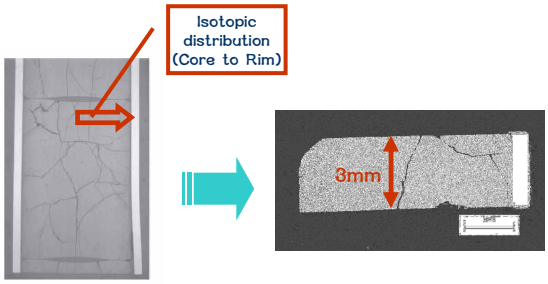

Fig. 1. Photos of prepared spent nuclear fuel specimen. (J502-A14, 55,600 MWd/MtU)

3. Results and Discussions

3.1 Distribution of Fission Products in Spent Fuel

Among the fission products, isotopic distribution of ^{236}U, ^{239}Pu, ^{240}Pu and ^{241}Pu in the spent fuel was measured by the LA-ICP-MS system. For the analysis of the spatial distribution of the U, Pu isotopes in spent fuel, 12 points from core to rim of the specimen were measured. Sampling was performed from core to rim of the pellet with intervals of 500 μm around core, reduced to 300 μm, 200 μm in the middle, and then 100 μm near the rim. The formation of ^{239}Pu is known to be preferential at the pellet periphery due to the epi-thermal neutron by ^{238}U. The higher Pu isotopes are produced as a result of successive neutron capture reactions. The increase of the Pu concentration in the rim zone was observed.

Fig.2 shows the isotopic ratios with respect to the ^{235}U peak as a function of the distance to the pellet rim. As can be observed, the ratio of ^{236}U/^{235}U remains almost constant. The ratios of Pu in the center are almost constant and the ratios of the Pu isotopes increased largely near the pellet periphery due to the rim effect. The mass of 241 corresponds both to ^{241}Pu and its beta decay product, ^{241}Am. As can be seen in our results, the ratio of Pu seems to increase at 400 μm

from the pellet rim. In this study, sampling was performed by 100 μm intervals near the rim zone.

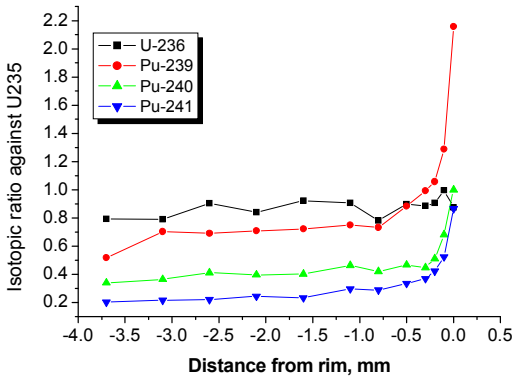

Fig. 2. The distribution of U, Pu from core to rim of spent fuel.

3. Conclusion

The radiation shielded LA-ICP-MS was applied to study the isotopic distribution of U, Pu in a spent nuclear fuel. It was observed that the isotopic ratio of ^{236}U with respect to ^{235}U is almost constant and that of Pu increased significantly near the pellet periphery due to the epi-thermal neutron by ^{238}U (so-called rim effect). Changes in the isotopic distribution of Pu in the high burn-up spent nuclear fuel were observed for the first time in Korea. The obtained results agreed well with those obtained from ITU.[5] This system can be applicable for the analysis of the spatial distribution of isotopes in an irradiated fuel from a research reactor as well as a high burn-up spent nuclear fuel.

Acknowledgements

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