Transactions of the Korean Nuclear Society Spring Meeting
Jeju, Korea, May 10-11, 2007

Application of Radiation Shielded Laser Ablation ICP-MS on Spent Nuclear Fuel

Yeong-Keong Ha, Sun-Ho Han, Hyun-Gyum Kim, Kwang Yong Jee
Korea Atomic Energy Research Institute, 150-1, Duckjin-dong, Yuseong-gu, Daejeon, Korea, 305-353 , nykha@kaeri.re.kr

1. Introduction

In relation to high burn up and extended fuel cycle for the fuel cycle efficiency, we need to take chemical analysis of spent nuclear fuel for the integrity of nuclear fuel at high burn up. The analysis based on the dissolution of the fuel lost information about volatile fission products and spatial information. Currently, Electron Microprobe Analysis (EPMA) has been used for highly radioactive materials. The laser ablation system coupled to inductively coupled plasma mass spectrometry (ICP-MS) could be applied to the direct analysis of solid sample. It provides a route for studying the spatial distribution of elements in the solid sample by probing the sample with a laser beam. To measure the direct analysis of the isotopes and their radial distribution in a high burn-up spent nuclear fuel, radiation shielded laser ablation system was designed and fabricated.[1-3] In this work, this system in conjunction with ICP-MS system was applied to the direct analysis of fission products in a spent nuclear fuel.

2. Experimental

2.1 Instrumentation

The laser ablation system was developed for the direct analysis of the fission products and their radial distribution in a spent fuel. It 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 lead shield glove-box for the analysis of radioactive material in a spent nuclear fuel The front panel consisted of a lead glass window, a couple of manipulator, and a pair of glove port. The rear panel has maintenance door and fused silica window for the UV laser entrance. The floor panel has three utility line holes for the translators, carrier gas and the image system. A lighting system was installed on the roof panel. The left side panel has cask adapter and specimen entrance hole. The image analyzer, XYZ translator, ablation chamber and various optics were installed inside the glove box (Fig. 1), while Q-switched Nd:YAG laser, optics, motion controller and associated electronics were outside.

The specimen cask was also fabricated to transport highly radioactive material.

The Laser ablation system was coupled to ICP-MS (Element, Finnigan) by PVC tubing with an argon flow of 1 L/min.

2.2 Preparation of sample specimen

For the analysis, a spent nuclear fuel (J502-A14, 55,600 MWd/MtU) was chosen as 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. Sampling was performed from core to rim of the pellet with changing intervals by 500 μm around core, reduced to 300 μm, 200 μm in the middle, and then 100 μm near the rim.

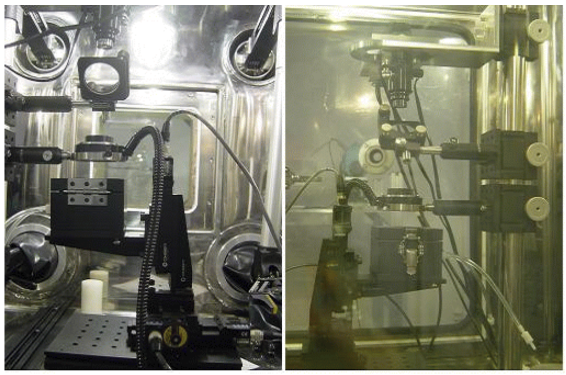

Fig. 1. Laser ablation system in a glove box (left) rear view, (right) front view through lead glass window

3. Results and Discussions

3.1 Distribution of Fission Products in Spent Fuel

Among the fission products, isotopic distribution of ^{100}Mo, ^{137}Cs, ^{144}Nd in the spent fuel was measured by the LA-ICP-MS system. From core to rim of the specimen, 12 points were measured. The increase of 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 in the center is almost constant and it increased near the pellet periphery due to the rim effect. It was reported that the width of rim zone was to be only 100 to 200 μm by EPMA, depending on the burn-up.[6] As can be seen in our results, it seems to increase at 400 μm from the pellet rim. In this study, sampling was performed by 100 μm intervals, but details of rim zone can be determined more precisely due to our smaller crater diameter (< 50 μm).

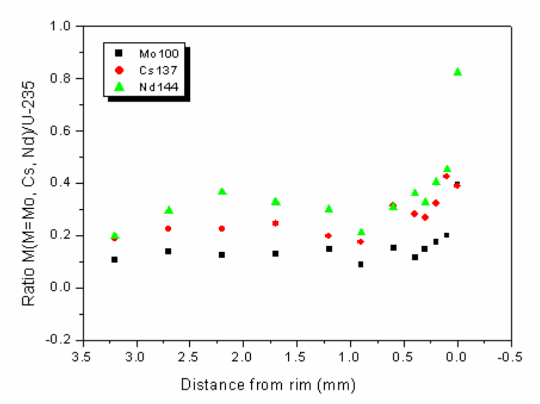

Fig. 2. The distribution of isotopic ratio from core to rim of spent fuel.

3. Conclusion

The radiation shielded LA-ICP-MS was applied to study isotopic distribution of fission products in spent nuclear fuel. It was observed that the isotopic ratio in the center is almost constant and it increased near the pellet periphery due to the rim effect. Details of the rim zone are the subject for a further study.

Changes in isotopic distribution of spent nuclear fuel were observed for the first time in Korea. This system can be used for the analysis of isotopic distribution from core to rim of a spent nuclear fuel and/or an irradiated fuel from research reactor.

Acknowledgements

We acknowledge the financial support of Nuclear Development Fund from Ministry of Science and Technology.

REFERENCES

[1] Y-K. Ha, S-H. Han, K.C. Han, K.Y. Jee, W.H. Kim, KRS:ISSN 1738-1142, Vol 2(2), p.184, 2004
[2] S-H. Han, Y-K. Ha, K.C. Han, Y.S. Park, K.Y. Jee, W.H. Kim, J. Korean Radioactive Waste Society, V.3(1), p.17, 2005
[3] Y-K. Ha, S.H. Han, Y.S. Park, S.D. Park, K.Y. Jee, W.H. Kim, KAERI/TR-3248/2006, 2006
[4] Hj. Matzke, J. Nucl. Mater., V.189, p.141 (1992)