

Performance Test of Radiation Shielded Laser Ablation ICP-MS on SRM613 and SIMFUEL

Sun-Ho Han, Yeong-Keong Ha, 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

Burn up measurement by analysis of fission monitors in a spent fuel and the radial distribution of fission product is essential for the operation of a reactor. Concerning the radial distribution of the fission products from core to rim in a spent fuel, the analytical method based on the dissolution is inadequate.[1,2] The laser ablation coupled to inductively coupled plasma mass spectrometry (ICP-MS) could be applied to the direct analysis of solid sample. It also provides a route for studying the spatial distribution of elements in the solid sample by probing the sample with a laser beam. Thus, we developed the lead shielded laser ablation system for the direct analysis of the isotopes and their radial distribution in a spent fuel.[3-5] In this work, the analysis of isotopic ratio in a standard reference material (SRM 613) and a simulated nuclear fuel (SIMFUEL) was carried out by this system in conjunction with ICP-MS system.

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 (Fig. 1). The image analyzer, XYZ translator, ablation chamber and various optics were installed inside the glove box, 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 (Fig. 1)

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

The SRM 613 and SIMFUEL were used as sample specimen for the performance test of shielded LA-ICP-MS system. The SRM 613 was supplied by NIST and the SIMFUEL was prepared from $(U_{1-y}Gd_y)O_2$ powder with nine different Gd compositions ($y = 0\sim 0.28$). The pellet was prepared by compacting the powder layer by layer followed by sintering in hydrogen atmosphere at 1700 °C. Sintered pellet was embedded in epoxy resin and then polished.



Fig. 1. Radiation shielded glove box and cask adapter for laser ablation system

3. Results and Discussions

3.1. Reproducibility

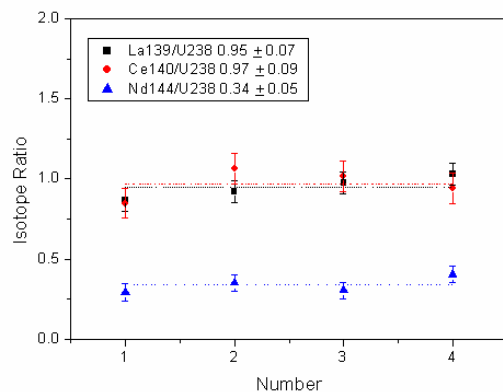


Fig. 2. Isotopic ratio of SRM613 by shielded LA-ICP-MS system

The isotopic ratios of ^{139}La , ^{140}Ce , ^{144}Nd against ^{238}U in the SRM 613 were measured by LA-ICP-MS system. The standard deviation of four measurements was less than 10 %, which satisfy the goal of this system.

3.2 Isotopic ratio of ^{158}Gd against ^{235}U

The layered SIMFUEL was piled up in the order of $y=0, 0.023, 0.045, 0.067, 0.088, 0.156, 0.277, 0.222$ and 0.123 . For an analysis, sampling was performed along the layer by $300\ \mu\text{m}$ intervals, so eight measurements were done for spatial analysis.

Considering the possibility of intermixture between layers, the obtained ^{158}Gd isotopic ratios were agreed comparatively well with expected value.

3. Conclusion

The radiation shielded LA-ICP-MS was applied to study isotopic distribution in the layered SIMFUEL. The results satisfied the requirements of this system. Thus, we concluded that 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.

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