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Preliminary Application of Laser Ablation Inductively Coupled Plasma Mass Spectrometry for Characterization of U-Mo Dispersion Fuel

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1. Introduction

Dispersion fuels have been developed in order to replace highly enriched uranium fuels in research reactors with low enriched uranium fuels for nuclear nonproliferation [1–4]. Among several current dispersion fuels, U-Mo particles dispersed in an Al base matrix (U-Mo/Al) are considered as a potential fuel because they have an excellent irradiation performance with good stability [5–7]. But irradiated U-Mo/Al fuel has some problems such as fuel particle swelling and interaction layer (IL) growth [8].

Laser ablation inductively coupled plasma mass spectrometry (LA-ICP-MS) has been successfully applied to determine trace elements and isotope ratios in U-10Mo fuel [9] and irradiated nuclear fuel [10-12]. LA-ICP-MS offers accurate, quick, and quantitative measurement with simple sample preparation and high sensitive chemical analysis. It is expected that LA-ICP-MS would be very useful to analyze irradiated U-Mo/Al dispersion fuel about the distribution of isotopes and the dispersed domain of each isotope. Those are very helpful to investigate the irradiation performance of U-Mo/Al dispersion fuel. Before applying LA-ICP-MS to directly characterize irradiated U-Mo/Al dispersion fuel, however, preliminary its application for unirradiated U-Mo/Al fuel sample is required to confirm the capabilities of this technique.

In this study, it was confirmed that LA-ICP-MS has the potential and the feasibility for the characterization of U-7Mo/Al-5Si dispersion fuel.

2. Materials and methods

U-7Mo/Al-5Si dispersion fuel was manufactured through a centrifugal atomized method at the Korea Atomic Energy Research Institute. The fuel plate with Al cladding was cut to a length of 28 mm and thickness of 1.4 mm. The U-7Mo particles were spherical within the Al-5Si matrix (Fig. 1). A NIST-SRM (Standard Reference Materials) 612 glass was also used to evaluate the capability of our LA-ICP-MS setup. ^{98}Mo and ^{238}U, the main isotopes in U-7Mo particles, were measured to confirm the reproducibility and the uniformity of the Mo/U element ratio.

LA-ICP-MS system consisted of ICP-MS (Element, Thermo Scientific) and a laser ablation system (LSX-213, Teledyne CETAC Technologies). The ablated materials by 213 nm laser source were introduced into the ICP torch with helium carrier gas and argon sampling gas. The ablated materials were ionized through the plasma and entered the mass detector. Spot analysis and line scanning were used for LA-ICP-MS measurement.

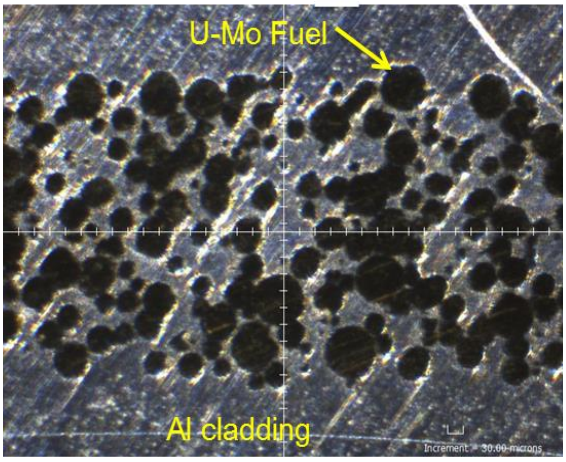

Figure 1. Cross section of U-7Mo/Al-5Si dispersion fuel. U-7Mo particles are dispersed in Al-5Si matrix. One interval in guide lines is 30 μm.

3. Results & Discussion

From a spot analysis for 25 sampling points of U-7Mo particles, the measured ^{98}Mo/^{238}U ratios show good reproducibility with 5.0% RSD. For reference, the spot analysis of the NIST-SRM 612 glass which has a homogeneous elemental distribution shows 3.4% RSD from same analysis method. From other result, femtosecond LA-ICP-MS analysis of ^{235}U/^{238}U ratio in U-10Mo sample showed 3% RSD [9]. These results of the measured data by LA-ICP-MS support that our instrumentation can provide precise isotope ratio measurements for fuel samples.

Line scanning is also performed to confirm the distinction between the U-7Mo fuel particles and the Al-5Si matrix along the scanned line. It is also performed to determine the size of the U-7Mo particles. The features of the ^{98}Mo and ^{238}U mass spectra from the line scans showed nearly identical responses and the U-7Mo fuel particles are clearly distinguished from the Al-5Si matrix. Average $^{98}Mo/^{238}U$ ratio for line scans is quite similar with that from spot analysis. The size of the U-7Mo fuel particle is estimated from the time width of the ^{98}Mo mass spectrum peak and the scan rate used for the line scan. The estimated value closely matches the optically measured diameter of the fuel particle. This measuring process fully demonstrates the capability of laser ablation inductively coupled plasma mass spectrometry for determining the dispersed domain of ^{98}Mo corresponding to the fuel particle size. Further optimized application should measure the change of fuel particle sizes caused by volume expansion by IL growth and swelling by fission products during irradiation of U-Mo/Al dispersion fuels.

4. Conclusions

The reproducibility of the measured data from spot analysis of U-7Mo fuel particles supports the capability of our instrumentation for precise isotope ratio measurements in fuel samples. The ability of this setup could distinguish the fuel particles from the Al-5Si matrix with line scanning method. The time width of peak in mass spectra corresponds to the size of fuel particle. This preliminary study clearly demonstrates that LA-ICP-MS has the potential to directly identify isotope ratios and dispersed domain of isotopes in U-Mo/Al dispersion fuels.

REFERENCES

[1] S. H. Lee, J. M. Park, and C. K. Kim, Thermophysical properties of U–Mo/Al alloy dispersion fuel meats, International Journal of Thermophysics, Vol.28, p.1578, 2007.
[2] H. J. Ryu, Y. S. Kim, J. M. Park, H. T. Chae, and C. K. Kim, Performance evaluation of U-Mo/Al dispersion fuel by considering a fuel-matrix interaction, Nuclear Engineering and Technology, Vol.40, p.409, 2008.
[3] Y. S. Kim and G. L. Hofman, Irradiation behavior of the interaction product of U-Mo fuel particle dispersion in an Al matrix, Journal of Nuclear Materials, vol.425, p.181, 2012.
[4] Y. S. Kim, B. J. Cho, D.-S. Sohn, and J. M. Park, Thermal conductivity modeling of U-Mo/Al dispersion fuel, Journal of Nuclear Materials, vol.466, p.576, 2015.
[5] J. L. Snelgrove, G. L. Hofman, M. K. Meyer, C. L. Trybus, and T. C. Wiencek, Development of very-high-density low-enriched-uranium fuels, Nuclear Engineering and Design, Vol.178, p.119, 1997.
[6] G. L. Hofman and Y. S. Kim , A classification of uniquely different types of nuclear fission gas behavior, Nuclear Engineering and Technology, Vol.37, p.299, 2005.
[7] C. K. Kim, J. M. Park, and H. J. Ryu, Use of a centrifugal atomization process in the development of research reactor fuel, Nuclear Engineering and Technology, Vol.39, p.617, 2007.
[8] H. J. Ryu and Y. S. Kim, Influence of fuel-matrix interaction on the breakaway swelling of U-Mo dispersion fuel in Al, Nuclear and Engineering and Technology, Vol.46, p.159, 2014.
[9] G. J. Havrilla and J. Gonzalez, Demonstration of femtosecond laser ablation inductively coupled plasma mass spectrometry for uranium isotopic measurements in U-10Mo nuclear fuel foils, Los Alamos National Laboratory, LA-UR-15-24319, 2015.
[10] Y.-K. Ha, S. H. Han, H. G. Kim, W. H. Kim, and K. W. Jee, Shielded laser ablation ICP-MS system for the characterization of high burnup fuel, Nuclear Engineering and Technology, Vol.40, p.311, 2008.
[11] Y.-K. Ha, J. Kim, Y. S. Jeon, S. H. Han, H. S. Seo, and K. Song, Local burnup characteristics of PWR spent nuclear fuels discharged from YEONGGWANG-2 nuclear power plant, Nuclear Engineering and Technology, Vol.42, p.79, 2010.
[12] Y.-K. Ha, J.-G. Kim, Y. S. Park, S. D. Park, and K. Song, Behaviors of molybdenum in UO_2 fuel matrix, Nuclear Engineering and Technology, Vol.43, p.309, 2011.