

Study on chemical variations of interface between U-Zr alloy and Fe through annealing

Young-Sang Youn^{a*}, Jeongmook Lee^a, Jong Hwan Kim^b, Hoon Song^b, Jeong-Yong Park^b, Jong-Yun Kim^{a,c}

^aNuclear Chemistry Research Division, Korea Atomic Energy Research Institute, 111, Deadeok-daero 989beon-gil, Yuseong-gu, Daejeon 34057, Republic of Korea

^bNext Generation Fuel Development Division, Korea Atomic Energy Research Institute, 111, Deadeok-daero 989beon-gil, Yuseong-gu, Daejeon 34057, Republic of Korea

^cRadiochemistry & Nuclear Nonproliferation, University of Science & Technology, Gajeong-ro 217, Yuseong-gu, Daejeon, 34113, Republic of Korea

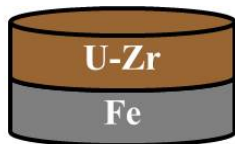
*Corresponding author: ysyoun@kaeri.re.kr

1. Introduction

Metallic fuel can be used for one of the fuels available in sodium-cooled fast reactors (SFRs) because it has advantages for thermal conductivity, inherent passive safety, and compatibility with sodium coolant [1-4]. Because the interfacial products between metallic fuel and cladding under high-temperature can affect the properties of metallic fuel, the effect of annealing on the interface between metallic fuel and cladding should be researched to understand the interfacial phenomenon. Therefore, herein we demonstrate the chemical changes at the interface between U-Zr alloy and Fe before and after annealing at 600 °C using X-ray diffraction (XRD), X-ray photoelectron spectroscopy (XPS), and Raman spectroscopy.

2. Experimental Details

The metallic fuel slug of U-10wt %Zr (U-Zr) was manufactured using injection casing method [5-6]. U-Zr samples 1 mm thick were made by cutting a metallic fuel slug. Fe was utilized as a simulated cladding because the main component of cladding for SFR is Fe. The U-Zr/Fe system in which a U-Zr specimen was placed on a Fe sample (Figure 1) was annealed at 600 °C for 1 h with a heating rate of 5 °C/min in an alumina tube furnace (Ajeon Heating Industrial) under an Ar atmosphere. After the annealing, the bottom of the U-Zr and the top of the Fe in the U-Zr/Fe system were measured using surface analysis tools.



U-Zr/Fe system

Fig. 1. Schematic U-Zr/Fe system consisting of U-Zr and Fe samples.

XRD data within the range of 20 to 120° were gained with a scanning step of 0.02° for 0.2 s using D8

ADVANCE (Bruker). The Cu K α was used as the X-ray source (beam current 40 mA at 40 kV). XPS spectra were obtained using a VG Scientific ESCALAB 220i-XL system with a Al K α X-ray source (1486.6 eV) at a base pressure below 5.0×10^{-9} Torr. XPS spectra were accumulated with an energy step of 0.1 eV and pass energy of 20 eV. The binding energies were calibrated using adventitious C 1s at 284.6 eV. The Raman results were obtained for 300 s using a SR500i Raman spectrometer (ANDOR) with a 632.8 nm helium-neon laser. The laser was focused on samples with 50 \times uncoated-objective lenses.

3. Results

All diffraction patterns of U-Zr and Fe samples before annealing correspond to those of orthorhombic α -uranium and cubic iron, respectively. After annealing of the U-Zr/Fe system at 600 °C, XRD data show diffraction peaks related to uranium dioxide in the U-Zr sample and iron with a low amount of iron oxide in the Fe specimen. In addition, through the XPS results before annealing, we confirmed that uranium and zirconium elements on the surface of the U-Zr sample were mostly oxidized into hyperstoichiometric UO $_{2+x}$ and Zr $_2$ O $_3$ compositions. In the Raman spectra, while a distinct peak of the U-Zr sample before annealing was not observed, after annealing of the U-Zr/Fe system the signals in the U-Zr sample appeared at 445, 575, 640, and 1150 cm $^{-1}$ for UO $_2$ as well as D and G bands at 1330 and 1590 cm $^{-1}$ for carbon. Raman spectra of the Fe samples before and after annealing exhibited a small peak at 668 cm $^{-1}$ for iron oxide and signals at 313, 538, 668 cm $^{-1}$ for iron oxide with a shallow D band at 1330 cm $^{-1}$ and G band at 1590 cm $^{-1}$ for carbon.

4. Conclusions

The chemical changes in the interface between U-Zr and Fe after annealing at 600 °C were investigated using XRD, XPS, and Raman spectroscopy. We observed that the surface of U-Zr sample at room temperature was mainly oxidized into hyperstoichiometric UO $_{2+x}$ and Zr $_2$ O $_3$ constitutions. On the basis of XRD and Raman data, the compositions of the U-Zr and Fe samples were

converted in UO_2 with carbon and iron oxide with a small amount of carbon by annealing of the U-Zr/Fe system, respectively. We anticipate that this work will contribute to fundamental information on the interfacial phenomenon between metallic fuel and cladding in a fuel rod.

ACKNOWLEDGEMENT

This work was supported by the National Research Foundation of Korea (NRF) funded by the Ministry of Science, ICT and Future Planning.

REFERENCES

- [1] G. L. Hofman, L. C. Walters, T. H. Bauer, Metallic fast reactor fuels, *Progress in Nuclear Energy*, Vol.31, p. 83, 1997.
- [2] T. Sofu, A review of inherent safety characteristics of metal alloy sodium-cooled fast reactor fuel against postulated accidents, *Nuclear Engineering and Technology*, Vol.47, p. 227, 2015.
- [3] A. Karahan, N. C. Andrews, Extended fuel swelling models and ultra high burn-up fuel behavior of U-Pu-Zr metallic fuel using FEAST-METAL, *Nuclear Engineering and Design*, Vol.258, p. 26, 2013.
- [4] Y.-S. Youn, J. Lee, J.-Y. Kim, J. H. Kim, H. Song, Change of Composition in Metallic Fuel Slug of U-Zr Alloy from High-Temperature Annealing, *Bulletin of the Korean Chemical Society*, Vol.37, p. 1492, 2016.
- [5] J. H. Kim, H. Song, H. T. Kim, K. H. Kim, C. B. Lee, R. S. Fielding, Development of a new casting method to fabricate U-Zr alloy containing minor actinides, *Journal of Radioanalytical and Nuclear Chemistry*, Vol.299, p. 103, 2014.
- [6] J. H. Kim, H. Song, K. H. Kim, C. B. Lee, Fabrication of uranium alloy fuel slug for sodium-cooled fast reactor by injection casting, *Journal of Radioanalytical and Nuclear Chemistry*, Vol.301, p. 797, 2014.