Temperature-dependent chemical changes of metallic fuel

Young-Sang Youn[a*], Jeongmook Lee[a], Jong Hwan Kim[b], Hoon Song[b], and Jong-Yun Kim[a]
[a]*Nuclear Chemistry Research Division, Korea Atomic Energy Research Institute, 111, Deadeok-daero 989beon-gil, Yuseong-gu, Daejeon 34057, Republic of Korea*
[b]*Next Generation Fuel Development Division, Korea Atomic Energy Research Institute, 111, Deadeok-daero 989beon-gil, Yuseong-gu, Daejeon 34057, Republic of Korea*
[*]*Corresponding author: ysyoun@kaeri.re.kr*

1. Introduction

Studies on metallic fuel slug of U-Zr alloys are performed to obtain the information on their physical and chemical properties because they are the candidates for a sodium-cooled fast reactor (SFR) [1-4]. Although many investigations for metallic fuel slug are executed in several decades, the chemical change of its surface depending on the temperature is rarely reported. Herein, we observed the temperature-dependent variations of U-Zr alloy using surface analysis techniques such as X-ray photoelectron spectroscopy (XPS), Raman spectroscopy, X-ray diffraction (XRD), and scanning electron microscope (SEM) equipped with energy-dispersive X-ray spectroscope (EDS).

2. Experimental Details

U-10wt%Zr alloy was manufactured by the injection-casting system with the casting temperature of 1600 °C under dynamic vacuum condition [5-6]. The heating of U-10Zr samples at 610 and 1130 °C for 5 min was performed using PBN heater in high vacuum chamber at a base pressure below 2.0×10^{-7} Torr. Temperature of the samples during the heating was measured by an infrared pyrometer (IMPAC IS 8 plus; LumaSense Technologies GmbH, Frankfurt, Germany). After the annealing of samples, they are transferred into the chamber for XPS measurement a base pressure of which is below 5.0×10^{-9} Torr.

XPS results were obtained in a high-vacuum chamber equipped with a VG Scientific ESCALAB 220i-XL system using an Al Kα X-ray source (1486.6 eV). XPS spectrum for survey scan was recorded with the pass energy of 100 eV and energy step of 0.5 eV. The binding energy of the spectrum was calibrated relative to that of adventitious C 1*s* (284.6 eV).

To minimize the exposure to air, the samples, which were heated in high vacuum system, were quickly transferred into Raman spectroscopy, XRD, and SEM installed with EDS system for measurement.

The Raman spectrum was measured by a SR303i Raman spectrometer with a 632.8 nm excitation wavelength helium-neon laser operating at 8 mW. The spectrum was acquired using an exposure of 300 s with a wavenumber range of 230 to 1050 cm^{-1}. The laser was focused onto a sample using 50× uncoated-objective lenses.

XRD patterns were obtained in the range of 20 to 120° with a scanning step of 0.02° for 0.2 s by BRUKER-AXS D8 ADVANCE. The Cu Kα filtered through Ni foil (beam current 40 mA at 40 kV) was used as X-ray source.

An SEM experiment was conducted without any sample treatment using a JEOL JSM-6610LV with an Oxford Instruments EDS.

3. Results

Figure 1 shows optical images of U-10Zr samples before and after heating at 610 and 1130 °C. The color of U-10Zr alloys is changed from gold before annealing (Figure 1a) to silver after heating at 1130 °C (Figure 1c).

Fig. 1. Optical images of U-10Zr samples (a) before and after the heating at (b) 610 and (c) 1130 °C.

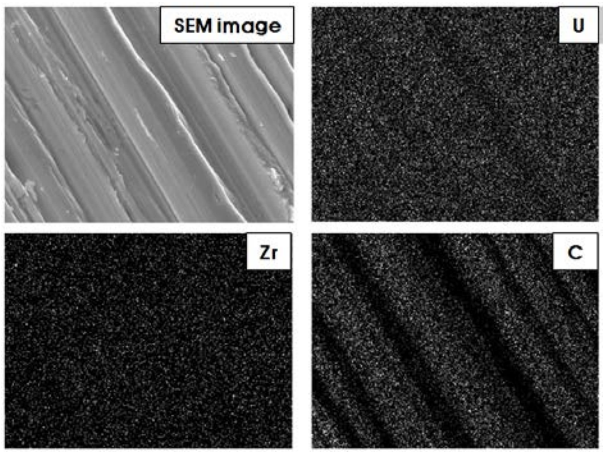

Fig. 2. SEM image (×3,000) and EDS mapping data of uranium, zirconium, and carbon elements obtained from U-10Zr sample before the heating.

We carried out the SEM experiment of U-10Zr alloy at room temperature. Based on the analysis of SEM image and EDS mapping data, we found that the surface constitution of U-10Zr sample uniformly exists as shown in Figure 2.

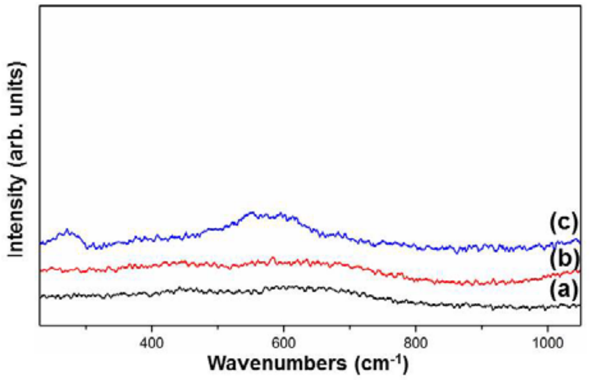

Fig. 3. Raman spectrum of U-10Zr samples obtained in range of 230 to 1050 cm^{-1} (a) before and after the heating at (b) 610 and (c) 1130 °C.

Figure 3 displays the Raman spectra of U-10Zr samples observed from a range of 230 to 1050 cm^{-1} (a) before and after heating at (b) 610 and (c) 1130 °C. There are no peak of Raman spectra for U-10Zr alloys at room temperature and 610 °C shown in Figure 3a and 3b, respectively. As shown in Figure 3c, the peaks around 274, 554, and 597 cm^{-1} are observed, which is resulted from the formation of zirconium carbide in U-10Zr sample after heating at 1130 °C. We also found similar phenomenon through the analysis of the XRD data, which are not shown here.

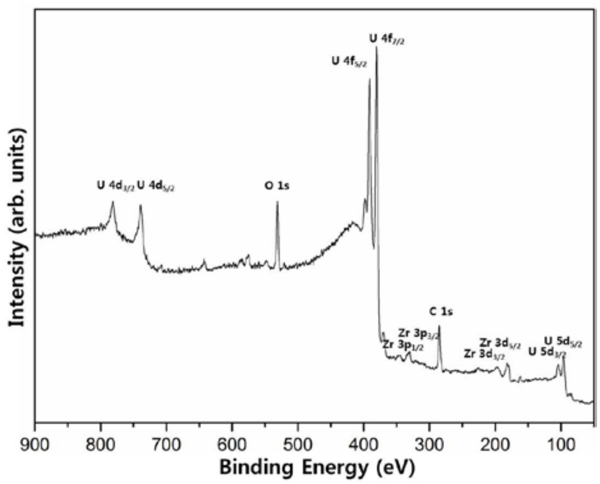

Fig. 4. XPS survey spectra of U-10Zr sample after the heating at 1130 °C.

XPS survey spectra of U-10Zr alloy after the heating at 1130 °C is exhibited in Figure 4. There are several peaks related to uranium, zirconium, oxygen, and carbon. Through the analysis of XPS data, we checked that the valence state of uranium on U-10Zr surface is gradually changed from U^{4+} at room temperature to U^{0} after heating at 1130 °C.

4. Conclusions

In this work, we exhibited the results of XPS, Raman, XRD, and SEM-EDS for U-10wt%Zr alloy at room temperature, 610 and 1130 °C. In SEM-EDS data, we observed that uranium and zirconium elements uniformly exist. After the annealing of U-10Zr sample at 1130 °C, the formation of zirconium carbide is verified through Raman spectroscopy and XRD results. Additionally, the change of valence state for uranium element is also confirmed by XPS analysis.

We anticipate that the study on the chemical change of U-10Zr alloys depending on the temperature will contribute to an improvement of knowledge in metallic fuel slug of U-10Zr alloys.

ACKNOWLEDGEMENT

This work was supported by the National Research Foundation of Korea (NRF) grant funded by the Korean government (MSIP) (No.2012M2A8A5025925)

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] T. Ogata, M. Akabori, A. Itoh, T. Ogawa, Interdiffusion in uranium-zirconium solid solutions, Journal of Nuclear Materials, Vol.232, p. 125, 1996.
[4] J. T. McKeown, S. Irukuvarghula, S. Ahn, M. A. Wall, L. L. Hsiung, S. McDeavitt, P. E. A. Turchi, Coexistence of the α and δ phases in an as-cast uranium-rich U-Zr alloy, Journal of Nuclear Materials, Vol.436, p. 100, 2003.
[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