Reactivity Evaluation of U-Zr Melt Residue after Melting in the Graphite Crucible

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1. Introduction

U-Zr alloy is a candidate of metallic fuel for sodiumcooled fast reactors (SFR) [1]. In the fabrication process of U-Zr metallic fuel, the graphite crucible is commonly used for melting and casting and Y_2O_3 is coated on the inner surface of the graphite crucible to prevent the reaction with melted U-Zr alloy [2]. It is crucial to prevent the reaction between the melt and crucible for controlling the loss in the metallic fuel fabrication.

In this study, we investigated the reactivity between uranium, zirconium and graphite crucible. The uncoated graphite crucible was used to clarify how the graphite react with melt uranium and zirconium. Microstructures in the surface region of the melt-residue of U-Zr alloy were characterized using Scanning electron microscopy (SEM) equipped with Energydispersive X-ray spectroscopy (EDS).

2. Methods and Results

A depleted uranium metal and zirconium sponges were prepared for the alloy. U-10wt.%Zr alloy was melted using injection casting apparatus which is an induction furnace. Raw materials consisting of zirconium sponge and uranium metal were charged in the un-coated graphite crucible in the order named. The materials was superheated at 1,600°C for alloying and it was cooled in the furnace after alloying. The surfaces of the U-10wt.%Zr alloy on the side, bottom and top regions were investigated to verify the reaction characteristics of the molten alloy and the graphite crucible using SEM-EDS.

Although the un-coated graphite crucible was used for melting uranium and zirconium, the U-10wt.%Zr alloy was fabricated without alloying between uranium and the graphite crucible. The melt residue was easily separated from the graphite crucible. The cross-section of the surface region of U-10wt.%Zr alloy is shown in Fig. 1. As the results indicate, on the whole, the reaction layer was formed on the surface. In contrast, the dark layer mostly consisted of C and Zr, and bright matrix consisted of U and Zr. The thickness of reaction layer was about 5 µm at side region and 10 µm at bottom region. The bottom region had the thickest reaction layer among whole surface because first of all, Zr was reacted with graphite crucible at the bottom region when the charged raw materials were heated. The thickness of the reaction layer was assumed as the depth penetrated into the U-Zr matrix from the graphite crucible depending on the volume of the zirconium, melting temperature and time.

The chemical composition of the reaction layer for the surface of the U-10wt.%Zr alloy are presented using EDS analysis in Fig. 2. The result indicates the inside carbonization of Zr in U-10wt.%Zr alloy from the graphite crucible during melting at high temperature The Zr and C contents of the reaction layer varied and the reaction layer had a tendency to be inversely proportional between Zr and C contents.





Fig. 1. Scanning electron micrographs of U-10wt.%Zr alloy fabricated using un-coated graphite crucible; (a) side region, (b) bottom region.



(b)

Fig. 2. Chemical compositions of the reaction layer of U-10wt.%Zr melt residue measured using EDS; (a) scanning electron micrograph, (b) U, Zr and C contents of the surface in melt residue.

The surface of U-10wt.%Zr alloy at the top region is shown in Fig. 3. As the result indicates, the surface of melt residue was roughly solidified including Zr-rich layer like the reaction layer. It was difficult to demonstrate the direct cause in this paper, but the formation of Zr on the surface of melt residue was assumed as impurities contaminated from graphite crucible, rigid felt, raw materials and low vacuum degree during melting and this layer was formed during melting and cooling because of density difference with U-Zr alloy. In addition, unlike the reaction layer directly formed from graphite crucible, the surface layer of melt residue had a high oxygen content. The thickness of surface layer was about 3 µm.



Fig. 3. Scanning electron micrographs cross-section images of the surface of U-10wt.%Zr melt residue fabricated using un-

coated graphite crucible.

3. Conclusions

In this study, U-10wt.%Zr alloy was fabricated in the optimizing manufacturing process to examine the reactivity between uranium, zirconium and un-coated graphite crucible. The Zr-rich layer was formed on the whole surface of the melt residue. Zr was commonly reacted with C at the contact region with graphite crucible. Zr-rich layer was formed by the contamination with C and O at the top surface region. The thickness of Zr-rich layers was found to increase in the order of bottom, side and top region.

REFERENCES

[1] G. L. Hofman, L. C. Walters, Metallic fast reactor fuels, in: R. W. Cahn, P. Hassen, E. J. Kramer (Eds.), Material Science and Technology, Nuclear Materials, Part I, vol. 10A, VCH Publishers Inc., 1994, p. 3.

[2] J. H. Kim, H. Song, K. H. Kim, and C. B. Lee, Protective of yttira coatings of melting crucible for metallic fuel slugs, Surface and Interface Analysis, Vol. 47, no. 3, 2015, pp. 301-307.