Characterization of Surface Reaction Layer for U-Zr- RE Metallic Fuel Slugs

Seung-Uk Mun^{a,b*}, Ki-Hwan Kim^b, Sun-Ig Hong^a

^aDepartment of Advanced Materials Engineering, Chungnam National University, Daejeon, Republic of Korea ^bSFR Nuclear Fuel Development Division, Korea Atomic Energy Research Instite, Daejeon, Republic of Korea ^{s*}Corresponding author: khkim@kaeri.re.kr

1. Introduction

As a part of a Sodium-cooled Fast Reactor (SFR) fuel development, metallic fuel slugs have been developed as a nuclear fuel of Next Generation Nuclear Plant(NGNP). A Metallic fuel is used for SFR, which is a representative model of Gen-IV. These metallic fuel slugs have been fabricated using an injection casting method. A modified injection casting has been used for production of metallic fuel slugs as an alternative fabrication method. The modified injection casting prevents the evaporation of volatile elements under pressurized Ar atmosphere during the melting process [1]. The uranium-zirconium-rare earth (U-10wt.%Zr-Xwt.% RE (X = 0, 5, 10)) alloys have a strong reactivity with quartz molds. The Si element in a quartz mold reacts and penetrates the U-Zr-RE (RE: 53%Nd, 25%Ce, 16%Pr, 6%La) metal fuel slug during injection casting. This injection casting process is described herein, and surface reaction layer of metallic fuel slugs were characterized using SEM/EDS.

2. Methods and Results

The quartz mold was preheated to $600\,^\circ$ C while the charged metal-fuel material was heated to $1470\,^\circ$ C. The quartz mold was immersed in the melted metal fuel material at $1470\,^\circ$ C, and Ar gas was infused to inject the molten metal into the quartz mold. The quartz tube mold was coated with using a slurry coating method Y₂O₃ which showed good performance for U-Zr-RE alloy [2]. The diameter of the cast U-10Zr-RE fuel slugs was 5mm, with a length of 250mm. The injection casting was carried out with a higher injection pressure of 2kgf/cm².

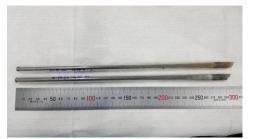


Fig. 1. Metallic fuel slugs fabricated using an injection casting method.

Fig.1 shows fabricated U-Zr-RE metallic fuel slugs fabricated with an injection casting system. Metallic fuel slugs were generally sound without crack or thin sections. All fuel slugs with a diameter of about 5.5mm were fabricated per batch.

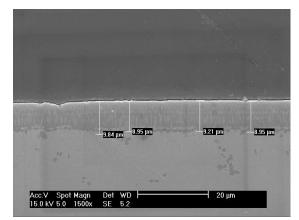


Fig. 2. SEM cross-section image of the U-10wt.%Zr metallic fuel slug surface reaction layer

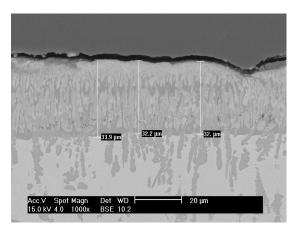


Fig. 3. SEM cross-section image of the U-10wt.%Zr-5wt.%RE metallic fuel slug surface reaction layer

Fig. 2. shows the scanning electron micrographs of U-10wt.%Zr fuel slug surface layer. A dark gray phase and a light gray phase were distinguished from the matrix in the top, middle, and bottom parts of metallic fuel slugs. the surface reaction layer thickness increased as the RE content of the metal fuel slugs increased, and the surface reaction layer of about 30 μ m was formed in the U-10wt.% Zr-5wt.%RE fuel slugs. Recent studies reported that Si-Zr forms a compound and it is thought that a uniform Si reaction layer is formed due to the

high-temperature reaction in the boundary layer between the quartz (SiO₂) template and the U-10wt.%Zr alloy.

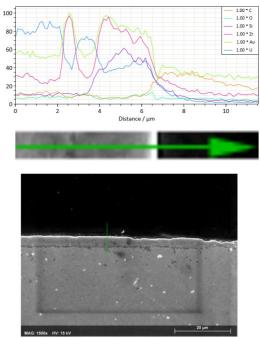


Fig. 3. EDS line scanning image on surface reaction layer of the U-10wt.%Zr metallic fuel slug.

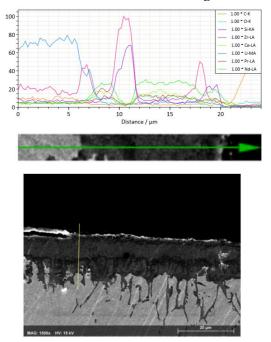


Fig4. EDS line scanning image surface reaction layer of the U-10wt.%Zr metallic fuel slug.

Fig. 3 shows differences in composition according to the surface reaction layer from top to bottom. The dark gray part of the surface layer confirmed that there was more Zr than in the light gray layer. In U, it is presumed that a U-Si compound was formed in the Si penetration region due to a small amount in the outer layer. A zone where Si was detected simultaneously with U and Zr was observed and other studies have reported that Zr-Si and U-Si form compounds with each other. [3, 4] Haefling et al. demonstrated that Nd, Ce, Pr and La are immiscible and do not react with molten U. Furthermore, the melting temperatures of Nd, Ce, Pr and La are 1,024°C, 795°C, 935°C, and 920°C, respectively [5, 6]. Zr-5.10wt.% The U-10wt.% RE allov was approximately 1,300 $^\circ C$, which indicated that the immiscible RE elements would still be liquid when the U-10wt.% Zr-5wt.% RE alloy solidified during cooling after casting. In this situation, it seems that Si diffuses from the quartz mold and reacts with the RE elements. RE elements have no independent distribution by component, and the tests confirmed the RE-rich zone. This appears to be due to the immiscibility of between U and RE elements. Si from the quartz was detected in the surface reaction layer of the metallic fuel slug, but there was no Y₂O₃ slurry coating layer in the surface reaction layer of the metallic fuel slug. It seems that the Y₂O₃ coating layer appears to be deteriorated by the molten metal during injection casting.

3. Conclusions

The surface reaction layer thickness increased as the RE content of the metal fuel slugs increased, and the surface reaction layer of about 30 μ m was formed in the U-10wt.%Zr-10wt.%RE fuel slugs. Due to the reaction between the metal fuel and the quartz (SiO₂) mold, the fuel slug surface reaction layer was predominantly consisted of precipitates formed by the Si element penetrating the metal fuel slugs and reacting with the RE element. The immiscibility between a RE element and a U element affects the formation of a surface reaction layer. For this reason, it causes the depletion of the U element at the surface reaction layer of the ternary U-Zr-RE metallic fuel slug.

REFERENCES

[1] J.H. Kim, H. song, K.H. kim, C.B Lee, J. Radioanal. Nucl. Chem, vol. 301, pp794-803 ,2014.

[2] Philip S. chen, Ward C. stevens and C. L. Trybus, Reusable molds for casting U-Zr alloys, Sixth international Conference on Surface Modification Technology The Metallugical Society, Chicago IL, 1992

[3] T. Yamauchi, S. Zaima, K. Mizuno, H. Kitamura, Y. Koide, and Y. Yasuda, Solid-phase reactions and crystallographic structures in Zr/Si systems, Journal of Applied Physics 69, 7050, pp1-3, 1991.

[4] A. Berche a, C. Rado b, O. Rapaud b, C. Guéneau a, J. Rogez c, Thermodynamic study of the U-Si system, Journal of Nuclear Materials 389, 1, pp101-107, 2009.

[5] J.H haefling, A.H Daane, The immiscibility limits of uranium with the rare-earth metals, Trans. Met. Soc. AIME 215, pp 336-338, 1959.

[6] H. Okamoto, La-U (Lanthanum-Uranium), J. Phase equilib, vol. 20, p639, 1999.