Effect of Marangoni migration on the formation of the liquid-state U-Zr-RE surface layer

Seung Uk Mun[a,b,c], Jungsu Ahn[c], Jun Hwan Kim [c], Sang-Gyu Park [c],
ByungMook Weon[a,b,*]
[a)]Soft Matter Physics Laboratory, School of Advanced Materials Science and Engineering,
Sungkyunkwan University, 066, Seobu-ro, Jangan-gu, Suwon-si, Suwon, 16419, gyeonggi-do, Republic of Korea
[b)]Research Center for Advanced Materials Technology, Sungkyunkwan University, 066,
Seobu-ro, Jangan-gu, Suwon-si, Suwon, 16419, gyeonggi-do, Republic of Korea
[c)] 3 Advanced Nuclear Fuel Technology Development Division, Korea Atomic Energy Research Institute (KAERI),
111 Daedeok-daero beon-gil, Yuseong-gu, Daejeon, Chungcheongnam-do 34057, Republic of Korea.
E-mail: bmweon@skku.edu

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1. Introduction

SFR reactors are being investigated in conjunction with pyroprocessing technology to increase the efficiency of high-level radioactive waste disposal using spent nuclear fuel as the main raw material. The inhomogeneous distribution of uranium and rare earth elements due to their immiscibility has a major impact on the production of microstructurally safe nuclear fuel. As an alternative to this, strain injection casting has been used for the production of metal fuel slugs. Strain injection casting prevents the evaporation of volatile elements in a pressurized argon atmosphere during the melting process [1]. In uranium-zirconium-rare earth (U-10wt.%Zr-5wt.% RE (RE: 53%Nd, 25%Ce, 16%Pr, 6%La)) alloys, the rare earth elements and uranium are not intermixed, resulting in microstructural inhomogeneities [3-6]. Rare earth element layers have been found to form on the surface of fuel cores. At high temperatures, rare earths are highly reactive and contribute to the generation of nuclear waste during the fuel casting process. In addition, the rare earth layer causes reactions with the cladding in the high temperature environment inside the reactor. In this study, we investigate the mechanism of rare earth layer formation on the surface and suggest research directions for surface layer suppression.

2. method and result

The quartz mold was preheated to 600°C, while the charged metal-fuel material was heated to 1470°C. Subsequently, the quartz mold was immersed in the molten metal fuel material at 1550°C, and Ar gas was infused to facilitate the injection of the molten metal into the quartz mold. Y_2O_3, known for its excellent performance in U-Zr-RE alloy, was applied to coat the quartz tube mold using a slurry coating method [2]. The resulting cast U-10Zr-5RE fuel slugs had a diameter of 5.56mm and a length of 300mm. The injection casting process was executed at a higher injection pressure of $2 kgf/cm^2$. Fig. 1. shows the separation of uranium and rare earth elements due to their immiscibility. Uranium and zirconium are alloyed and distributed in the same region. However, the rare earth elements are distributed separately from the uranium zone. Also, all four rare earth elements are distributed in the same region.

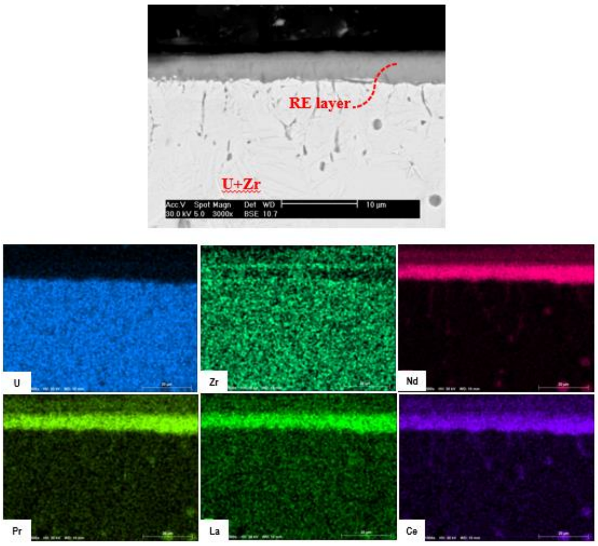

Fig. 1. SEM-EDS cross-section image of the U-10wt.%Zr metallic fuel slug

Fig. 2. shows XRD data of the fuel core surface layer. The peaks were identified as rare earth oxides and silicon compounds. The XRD analysis confirmed that the distribution of rare earth elements in the SEM-EDS analysis was consistent. Rare earth elements are immiscible in liquid form with uranium. The surface tension of uranium in liquid is higher than that of rare earth elements. The difference in surface tension causes Marangoni migration in the liquid state. Since the surface tension of uranium elements is greater than that of rare earth elements, the rare earth elements near the surface layer of the fuel slug will migrate to the surface layer. The surface layer thickness of the injection-cast metal fuel slug varied by region. The components of the surface layer were all rare earths.

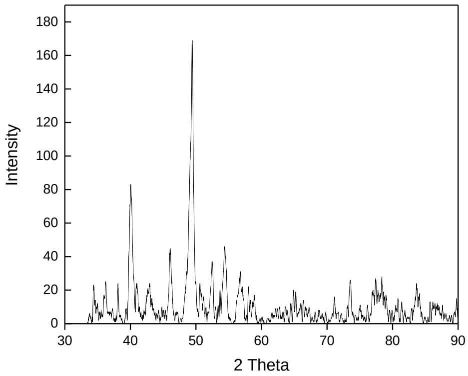

Fig. 2. XRD results of the U-Zr-RE metallic fuel slug surface layer

In the injection casting process, the area where the mold is immersed remains hotter than other areas. This will affect the cooling rate of the molten metal fuel slug components. The extended solidification time provides rare earth elements with a prolonged period to migrate, potentially influencing their distribution and concentration within the solidified matrix. SEM analysis showed that the thickest surface layer was observed at the bottom of the fuel slug. This means that the uneven distribution of temperatures in the process can affect the thickness of the fuel core surface layer.

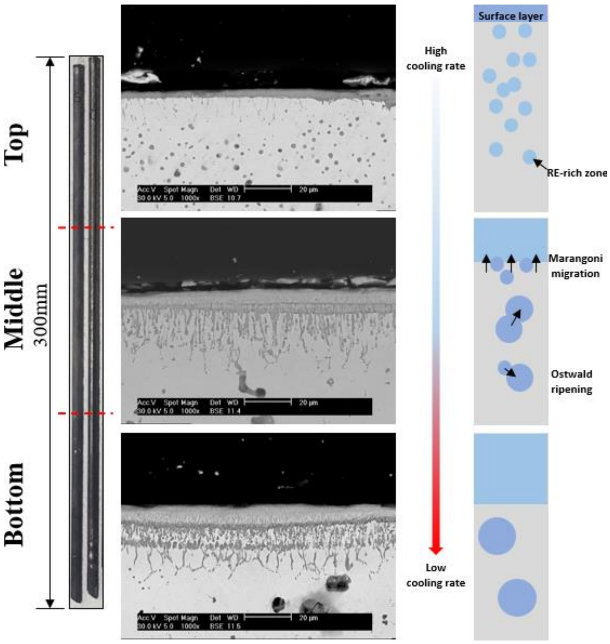

Fig. 3. SEM (BSE mode) images and Rare Earth Element Migration Schematic

3. Conclusion

In this study, the injection casting process for U-10Zr-5RE fuel slugs was meticulously analyzed, with particular attention to the distribution and behavior of rare earth elements (RE) within the alloy matrix.

The XRD analysis of the fuel core surface layer corroborated the presence of rare earth oxides and silicon compounds, further confirming the distribution observed in the SEM-EDS analysis. The surface tension disparity between uranium and rare earth elements was identified as the driving force behind the Marangoni migration observed during solidification. This migration resulted in the accumulation of rare earth elements at the surface, with the thickest surface layer noted at the bottom of the fuel slug, highlighting the impact of temperature gradients during the process.

The findings underscore the significant influence of solidification time on the migration and distribution of rare earth elements. The extended solidification time allowed for increased mobility of these elements, leading to their uneven distribution within the fuel slug. Consequently, the thermal management and control of cooling rates during the casting process are critical to achieving a uniform microstructure and optimizing the performance of U-Zr-RE metallic fuels. Further studies are warranted to refine these parameters and enhance the understanding of rare earth behavior in nuclear fuel alloys.

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