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Interaction Barrier Effect of Zr-ZrO_2 Thin Film Deposited by Hydrothermal Deposition for Sodium-cooled Fast Reactor

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1. Introduction

The great attention has been shown to the unique properties of a metal fuel, such as high thermal conductivity, high proliferation resistance, ease of fabrication, and a good compatibility for the sodium in Sodium-cooled Fast Reactor (SFR) for burning the long-lived fission products. [1, 2] The U-Pu-Zr alloy with the Zr content of 10~20 % for high solidus temperature has received much attention for the most desirable metal fuel. However, It has several problems to use the U-Pu-Zr metal fuel directly since a fuel-clad chemical reaction (FCCI) caused by the eutectic reaction between actinide elements (U, Pu) and a cladding material based on stainless steel at above 650℃. It is researched that lanthanide elements (La, Ce) generated by nuclear fission of actinides (U, Pu) exacerbate FCCI phenomena.[3]

In recent years, numerous studies have attempted to prevent FCCI during the fuel burn-up since the FCCI decreases the life of a fuel element by decreasing of mechanical strength and reliability of clad gradually. Therefore, it is necessary that the novel research for the interaction barrier to prevent the FCCI between the cladding and the metal fuel with high strength and adhesion without the cladding wastage. It was reported that a Zr foil on the U-Pu-Zr fuel rod is very effective to mitigate the clad-fuel interaction because the Zr foil acts as a diffusion barrier. [4] However, the study of the interaction barrier to prevent the FCCI has not been reported to best knowledge. To maximize the performance of Zr as a diffusion barrier, Zr thin film was deposited by a RF magnetron sputtering. Then, the Zr thin film was treated by a hydrothermal method to change the crystalline structure for the interaction barrier.

In this study, Zr-ZrO_2 thin films were deposited on metal clad by by RF magnetron sputtering and hydrothermal method with different pH to propose the possibility of Zr thin film as the barrier materials can be deposited onto the inner surface of the cladding.

2. Methods and Results

2.1 Experimental procedure

The Zr thin film to prevent the FCCI and the eutectic reaction was deposited on a Ferritic-Martensitic Steel (9Cr-2W) clad by RF magnetron sputtering with a Zr target(99.99%). The chamber was kept base pressure of 5 x 10^{-5} torr and working pressure of 8 X 10^{-3} torr. Thickness and deposition rate of the Zr thin film was 3um and 15nm/min, respectively. Zr thin films were treated from Zr thin film in zirconium hydroxide mixed DI water by hydrothermal method with various pH values at the 150℃ for 24 hours. Nitric acid (HNO_3) and ammonia (NH_4OH) was used for various pH. The pH was controlled from 2~9. X-ray Diffraction (XRD) and Field Emission Scanning Electron Microscopy (FESEM) with EDS were contacted to analyze the crystalline structure and surface and cross-section image. Besides, a diffusion couple experiment between lanthanide alloy (Misch metal, 70Ce-30La) and the clad was performed at 600℃ for 24 hours.

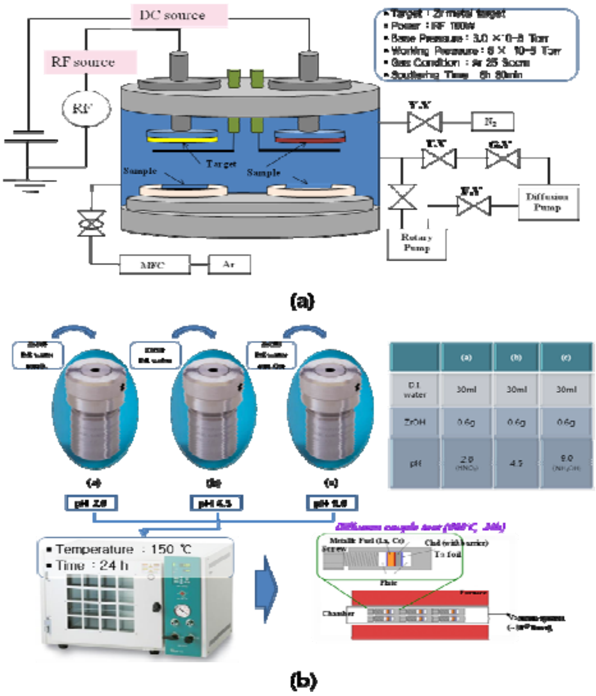

Fig. 1. Experimental procedures in this research.

2.2 Crystalline structure of the Zr thin film

Fig. 2 shows the crystalline structure of Zr thin films. The crystalline structure of the Zr thin film by RF sputtering was a hexagonal phase. The crystalline structure was changed to a monoclinic phase by hydrothermal method. Grain size was increased from 8.56nm to 21.12nm. The ZrO_2 layer was deposited on surface of Zr thin film by hydrothermal method. The surface on Zr thin film was oxidized by hydrothermal method. The crystalline structure of ZrO_2 layer on Zr thin film was tetragonal structure (220) which was analyzed by using XRD. We expected that the crystalline structure of ZrO_2 layer will be changed by various pH. However, there was no effect of pH.

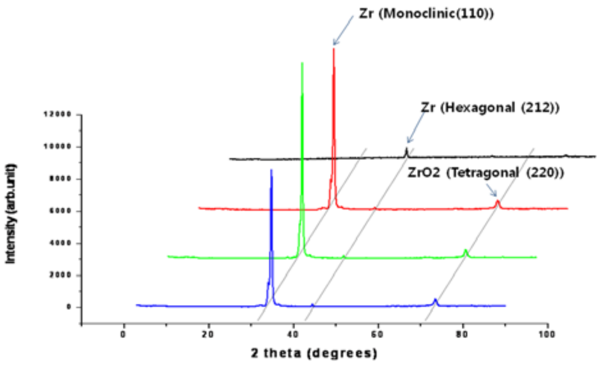

Fig. 2. XRD spectra of Zr thin films.

2.3 Diffusion couple test and discussion

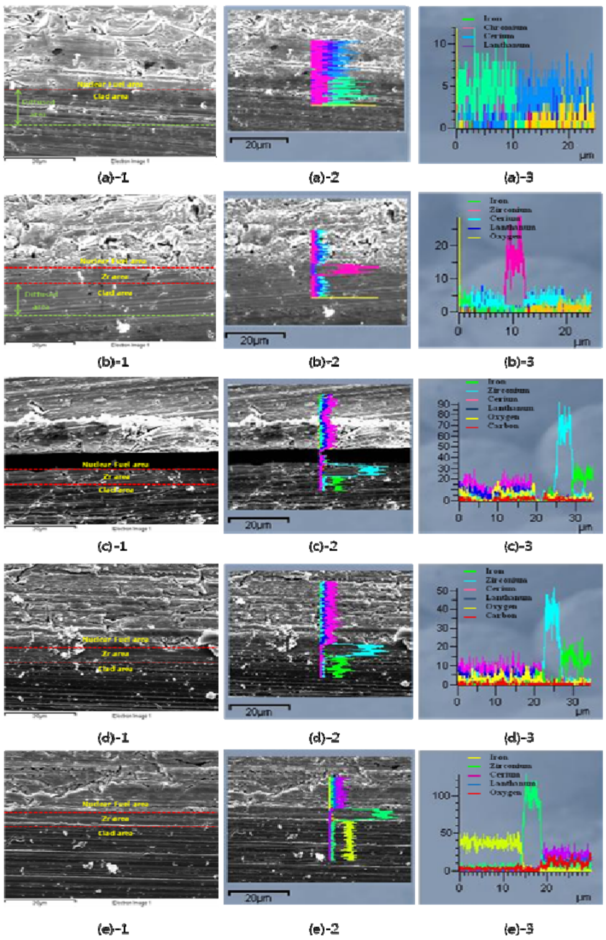

Fig. 3. SEM Images and EDS data in the sample (a) without diffusion barrier (b) Zr by sputtering (c), (d), (e) hydrothermal treated Zr film in various pH(2, 5, 9).

The cladding elements (Fe, Cr) and misch metal were totally inter-diffused in the sample (a) without diffusion barrier. The cladding elements (Fe, Cr) and Fuel elements (Ce, La) were also inter-diffused through the grain boundary in Zr thin film in the sample (b) with Zr thin film by sputtering. However, the cladding elements (Fe, Cr) and Fuel elements (Ce, La) were not inter-diffused in the sample (b) with Zr-ZrO_2 thin film by hydrothermal method. As a results, We demonstrated that the inter-diffusion was dependent on grain size. Besides, the ZrO_2 thin film on surface gives the improvement for inter-diffusion effect. However, There was no effect of pH in the diffusion test as the XRD results.

3. Conclusions

The Zr thin film was deposited by RF magnetron sputtering with Zr target. The hydrothermal deposition of Zr-ZrO_2 thin films onto a FMS clad was carried out to prevent a FCCI and a eutectic reaction of a Ce, La in metal fuel with clad material of nuclear power plant. Zr-ZrO_2 thin films were deposited with various pH values at the 150℃ for 24 hours. X-ray diffraction (XRD) patterns indicated that an monoclinic structure of the hydrothermal deposition grown Zr-ZrO_2 thin films regardless of growth temperature and ph variation even though tetragonal of ZrO_2 structure and monoclinic of Zr structure were found in plane and cross sectional images of the as deposited Zr-ZrO_2 thin films with thickness range of 3um.A depth profile of auger electron microscopy (EDS) revealed that Zr and O atoms were found in the as deposited Zr-ZrO_2 thin films. Diffusion couple tests at 600℃ for 24 hours showed Zr-ZrO_2 thin films give the improved diffusion barrier behavior for FCCI between the metal fuel and the clad. This result suggested that Zr-ZrO_2 thin films on inner cladding tube, even with the thickness below few micrometer levels, has very high possibility as an effective barrier against FCCI.

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