Effects of Diffusion Barriers on the Fuel/Cladding Interaction of SFR Metallic Fuel

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1. Introduction

U-Pu-Zr metallic fuels have been developed as a fuel for a sodium-cooled fast reactor (SFR). In order to transmutate the long-lived minor actinide (MA) fission products such as Np, Am, Cm in spent fuel, transuranic element (TRU) bearing metallic fuel is being developed for a advanced burner reactor (ABR) or a advanced recycle reactor (ARR). One of the performance limiting factors of the metallic fuel is a fuel-cladding chemical interaction (FCCI) during irradiation. FCCI limits the fuel temperature due to a eutectic melting of interaction products at a temperature much lower than a melting point of a fuel alloy. Fuel cladding interaction behaviors of U-Pu-Zr alloys against Fe, Fe-Cr, D9 and HT9 have been investigated by out-of-pile annealing tests of their diffusion couples[1,2]. Recently, usage of a thin coating on the inside wall of a cladding as a diffusion barrier has been proposed in order to suppress the interaction between metallic fuel and a cladding. Tokiwai et al. showed that Zr liner suppressed interdiffusion of U and HT9 [3]. Keiser and Cole showed that Zr and V did not react actively with uranium in out-of-pile annealing tests of simulated metallic fuels(U-Pu-Zr-Nd-Mo-Ru) and Zr or V liner metals [4].

In this study, interdiffusion behaviors of U-Zr and U-Zr-Ce alloys (U-Zr-X) against FMS disks were investigated by observing the microstructures of the diffusion couples after out-of-pile annealing tests. In order to evaluate the diffusion barrier performance of metallic foils, Ti, V, Cr, Zr, Nb and Mo foils were inserted between the U-Zr-X and FMS.

2. Experimental Procedures

U-Zr and U-Zr-Ce alloys were fabricated by an induction melting of elemental lumps in zirconia crucibles. Small amount of Ce (~2 wt%) were added in order to simulate rare earth elements and minor actinide elements in a TRU bearing metallic fuel. Nominal Zr content was 10 wt%.

Because ferritic martensitic steels (FMS) have been considered as candidate cladding materials for a SFR fuel due to their excellent irradiation performance, disks of HT9 and T91 ferritic steels were used as cladding materials in a diffusion couple. Diffusion couple annealing tests were carried out in a muffle furnace at 740^oC for 25 hours because the eutectic melting temperature of U-Zr vs. Fe-Cr was known to be ~725^oC. Mechanical clamps joining the diffusion couples of U-Zr-X and FMS slices were vacuum sealed in quartz tubes.

Commercially available metallic foils of Ti, V, Cr, Zr, Nb and Mo were used as diffusion barrier layers inserted between the U-Zr-X and FMS disks. The thickness of each foil ranged from 20~30 μm.

Microstructures of diffusion couple specimens were observed by using a scanning electron microscopy (SEM) and the elemental composition of the interaction layers were measured by using an energy dispersive X-ray spectroscopy (EDS).

3. Results and Discussion

When a eutectic melting occurs between U-Zr and Fe, complex microstructures were observed due to a formation of multi-phase structures in a multi-component system. The interaction layer on the FMS side consisted of gray $(U,Zr)Fe_2$ and a bright liquid phase (as-quenched structure). On the U-Zr side, dark $(Zr,U)Fe_2$ and gray $(U,Zr)Fe_2$ were mixed in the $(U,Zr)_6Fe$ matrix.

When thin foils of Ti, V, Cr, Zr, Nb and Mo were used as the diffusion barrier layer between the U-Zr-Ce and FMS, the diffusion couple tests at 740^oC resulted in a very limited interaction between the metallic foils and U-Zr-X as shown in Fig. 1(a) and Fig. 2(a). Based on the interaction thickness, V, Ti and Cr resulted in a minimal interaction after the annealing tests, whereas the other foils showed some interaction with U-Zr-Ce. When the contents of the uranium and zirconium which diffused into the diffusion barrier foil were measured, Ti, Zr, and Nb layers showed some uranium content in their layer whereas V, Cr and Mo showed no uranium content in their layers as shown in Fig. 1(b) and Fig. 2(b).

Although Keiser and Cole reported that V appeared to be the best liner material from their out-of-pile diffusion couple test using U-Pu-Zr-Nd-Mo-Ru[4], Cr or Mo might be another candidate barrier material based on their diffusion barrier performances shown in this study. Because the effects of lanthanide fission products on FCCI have not been clearly understood yet, diffusion couple tests with some lanthanide elements is necessary. Higher temperature tests at around 800^oC with rare earth element are under way in order to select the most performing barrier material for the fuel/cladding interaction. Because oxides or nitrides of V, Cr and Mo can also be good diffusion barrier materials, oxide or nitride coated FMS disks are being tested. At the same time, fabrication processes of diffusion barrier coatings on the inside wall of cladding is being investigated. The effects of Pu and MA on the eutectic melting of the interaction layer will be studied as cooperation research with INL.

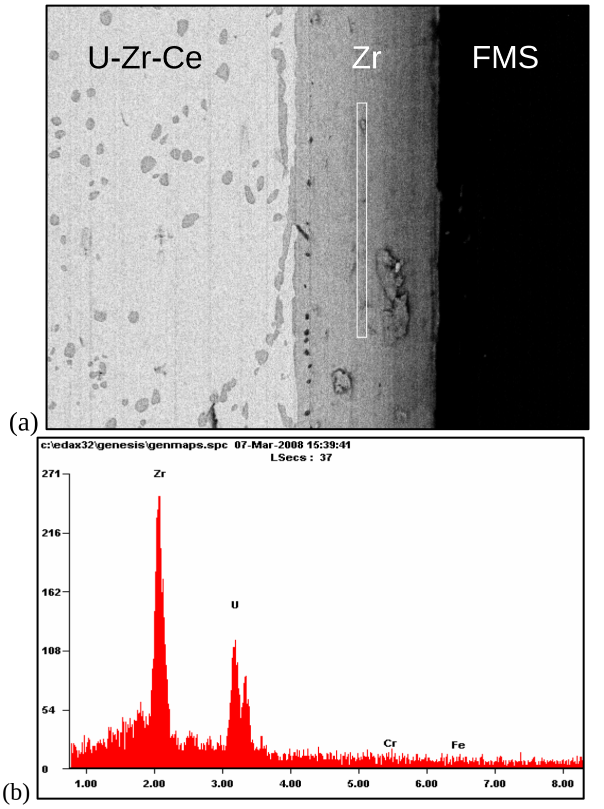

Fig. 1. (a) An SEM micrograph of a U-Zr-Ce vs. FMS diffusion couple with a Zr barrier foil. (b) An EDS spectrum of Zr barrier after annealing at 740^{o}C for 25 hours.

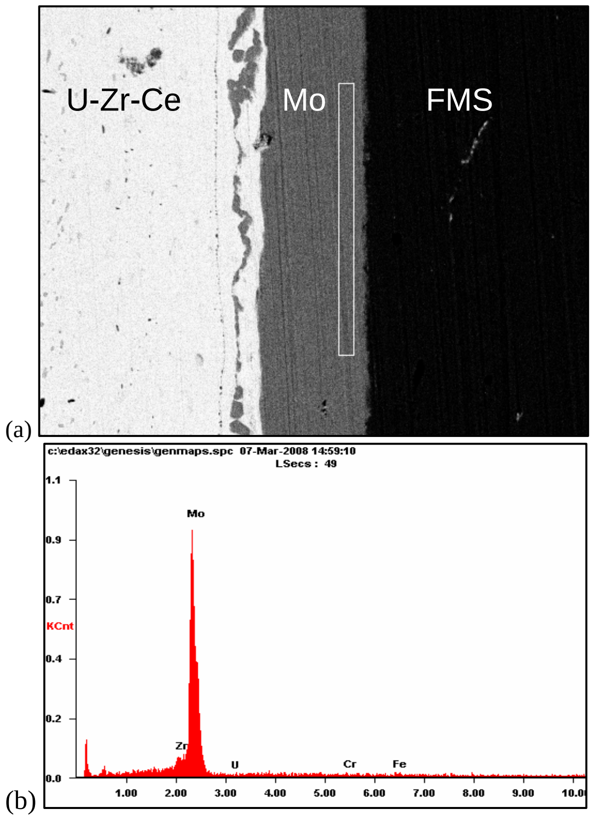

Fig. 2. (a) An SEM micrograph of a U-Zr-Ce vs. FMS diffusion couple with a Mo barrier foil. (b) An EDS spectrum of Mo barrier after annealing at 740^{o}C for 25 hours.

4. Conclusions

High temperature annealing tests of diffusion couples for U-Zr-X vs. FMS showed sound microstructures without a eutectic melting, when metallic diffusion barrier foils were inserted between the metallic fuel and the cladding materials. V, Cr, and Mo showed promising diffusion barrier performances whereas Zr, Ti and Nb allowed U to penetrate into their layers.

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