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INTRODUCTION

Historical interest in metallic fuel, including U-Zr and U-Pu-Zr, is due to the high thermal conductivity, high burnup capability, high fissile atom (^{239}Pu) density, and good compatibility with the coolant in sodium-cooled fast reactor conditions. The notable fuel application issue is fuel-cladding chemical interaction (FCCI). The major cause of FCCI is lanthanide fission products that migrate to the fuel periphery and deposit on fuel cladding. Chemical interactions between the lanthanides and cladding will result in a cladding breach. Thus additives were sought that would immobilize the lanthanides by forming stable additive-lanthanide compounds. Palladium (Pd) is being explored as a potential additive. The microstructure of metallic fuels, including U-Zr and U-Pu-Zr, with Pd addition have been characterized [1-4]. The post-irradiation examination on the U-13Zr-4Pd wt% alloy has found a pronounced effect of Pd on the fuel-cladding interface [5]. The current study performed preliminary tests of diffusion couple and microstructure analysis, in order to investigate the chemical interactions between cladding and fuel with Pd addition.

EXPERIMENT

The alloy compositions are listed in Table 1, with the alloy fabrication method previously reported [2][3]. The contact surface was polished to 1 µm. The surface oxide of U-Zr-Pd and U-Zr-Pd-Ln was removed in an argon-filled glovebox. The surface oxide of U-Pu-Zr-Pd and U-Pu-Zr-Pd-Ln was not removed due to limited polishing capabilities for Pu-based fuels.

Table 1. Alloy compositions.

Alloy	at%	wt%
U-Zr-Pd	U-25.2Zr-7.2Pd	U-12Zr-3.86Pd
U-Zr-Pd-Ln*	U-24.5Zr-7.0Pd-6.6Ln	U-12Zr-3.86Pd-5Ln
U-Pu-Zr-Pd	U-16.2Pu-21.7Zr-7.2Pd	U-20Pu-10Zr-3.86Pd
U-Pu-Zr-Pd-Ln*	U-15.8Pu-21.2Zr-7.0Pd-5.8Ln	U-20Pu-10Zr-3.86Pd-4.3Ln

* Ln = 53Nd-25Ce-16Pr-6La wt% (52.3Nd-25.4Ce-16.2Pr-6.1La at%)

The U-Zr-Pd and U-Zr-Pd-Ln diffusion couples were clamped in a jig, and heat treated at 923 K for 500 hours under vacuum in an argon-sealed quartz tube inside a furnace. After heat-treatment, the samples were quenched, and the cross sections were prepared for microstructural analysis. The U-Pu-Zr-Pd and U-Pu-Zr-Pd-Ln diffusion couples were clamped in a jig and heat treated at 923 K for 500 hours in a furnace inside an argon glovebox. The diffusion couples were allowed to cool to room temperature, followed by preparation for microstructural analysis. A JSM-700f scanning electron microscopy (SEM) equipped with energy dispersive X-Ray spectroscopy (EDS) was used to analyze the diffusion region. The samples for transmission electron microscopy (TEM) were prepared using a focused ion beam (FIB). A FEI 200 kV Titan Themis scanning transmission electron microscopy (STEM) equipped with super-X EDS was used for the nano-scale microstructure and composition analysis.

RESULTS AND DISCUSSION

Matrix diffusion

In the U-Zr-Pd(-Ln)/Fe diffusion couples, SEM analysis found a diffusion layer comprised of UFe_2, shown in Fig. 1(a). In the Pu-containing diffusion couples, $(U,Pu)Fe_2$ is formed, shown in Fig. 1(b). U and Pu tend to move together, i.e. phases high in U are also high in Pu.

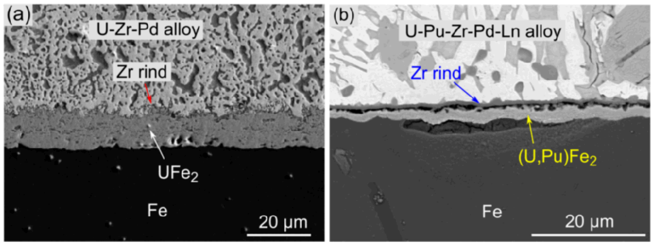

Fig. 1. SEM BSE images of diffusion between Fe and fuel matrix of (a) U-Zr-Pd, and (b) U-Pu-Zr-Pd-Ln alloys.

The Zr-rich layer (Zr rind) on the fuel surface was formed due to dissolved oxygen, shown in Fig. 1. Zr rind can act as a barrier, and suppress the diffusion of fuel and Fe. Since the oxide layer was removed, Zr rind on the fuel alloys with no Pu is extremely thin (< 1 µm), thus the effect is negligible. For the Pu-containing fuel alloys, the surface oxide not removed, and the resulting Zr rind is thick (~ 4 µm) and decreases diffusion of fuel and Fe. Due to this, comparison of the diffusion depth of the samples is not practicable; however, the current result indicates what reactions might occur in a reactor during FCCI.

Minor constituent phase diffusion

The diffusion area in U-Zr-Pd-Ln/Fe and U-Pu-Zr-Pd-Ln/Fe diffusion couples is larger than U-Zr-Pd/Fe and U-Pu-Zr-Pd/Fe, primarily caused by lanthanides. Ln-Pd binary phases and (Ln,Pu)Pd are the minor constituent phases present in the U-Zr-Pd-Ln and U-Pu-Zr-Pd-Ln alloys, respectively; however, lanthanides were found to release from the phases and diffuse toward the Fe side.

In the U-Zr-Pd-Ln/Fe diffusion couple, the diffusion area is comprised of U-Zr-Fe and Ln-Fe phases, shown in Fig. 2. The mottled area (i.e., U-Zr-Fe) on the fuel side is formed by Fe diffusion into the fuel. The Ln-Fe diffusion layer is comprised of $LnFe_4U$ and Ln_2Fe_{17}, which were confirmed by both STEM EDS and TEM diffraction analysis. STEM EDS analysis found Ce diffused the fastest among four lanthanides. The diffusion front of lanthanides is free of U, thus lanthanides diffused faster than U.

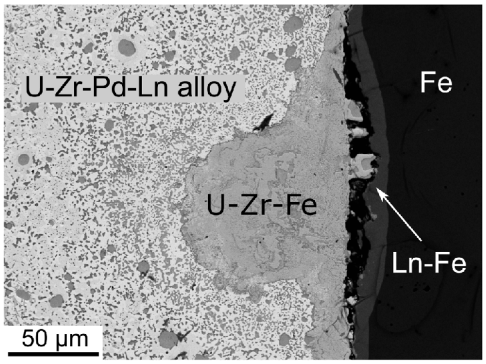

Fig. 2. SEM BSE image of diffusion in U-Zr-Pd-Ln/Fe diffusion couple.

In the U-Pu-Zr-Pd-Ln/Fe diffusion couple, it was found (Ln,Pu)Pd diffused with Fe. Pu was released from (Ln,Pu)Pd and readily reacted with Fe, forming $PuFe_2$ layer at the interface, shown in Fig. 3. Pd did not immobilize Pu, which is not the point of adding Pd in the fuel. Next to $PuFe_2$ is a Ln-rich layer. It is speculated that the lanthanides were migrating from the Ln inclusion trapped in the (Ln,Pu)Pd precipitate, or releasing from (Ln,Pu)Pd, which needs further investigation. Pd did not prevent all lanthanides-caused FCCI, but greatly decreased the FCCI. In comparison with the past studies on Nd/Fe and Ce/Fe diffusion couples [6,7], diffusion areas comprised of Ln and Fe, shown in Figs. 2 and 3, are much smaller.

CONCLUSION

The diffusion couples between U-Zr-Pd, U-Zr-Pd-Ln, U-Pu-Zr-Pd, and U-Pu-Zr-Pd-Ln alloys and Fe have been investigated, with SEM data presented here. Diffusion layers comprised of $(U,Pu)Fe_2$ and UFe_2 are formed in the diffusion couple samples with and without Pu, respectively. Pd did not prevent all lanthanides-caused FCCI, some lanthanides were released from Ln-Pd phase to react with Fe. However, Pd has greatly mitigated the diffusion between lanthanides and Fe, and immobilized most of lanthanides in the fuel.

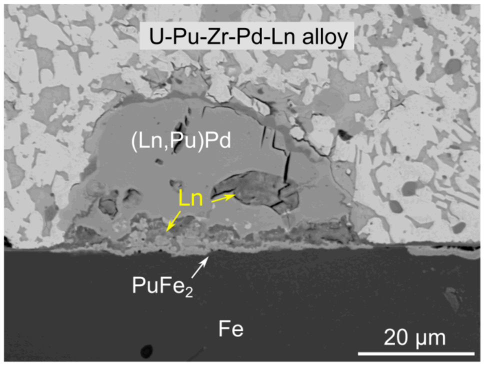

Fig. 3. SEM BSE image of diffusion between (Ln,Pu)Pd and Fe in U-Pu-Zr-Pd-Ln/Fe diffusion couple.

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