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Diffusion behavior in an interface between U–10Zr alloy and HT-9 steel

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A B S T R A C T

The diffusion behavior in an interface between a U–10 wt.%Zr alloy and HT-9 steel at 700 °C and 730 °C was investigated. The Zr-rich layer near the interface between two alloys was formed mainly due to the decomposition of the δ-UZr_2 phase in U–Zr alloy, and acted to interrupt the interdiffusion of the alloying elements. Considerable interdiffusion was observed only when the Zr-rich layer was destroyed locally. As a result, the diffusion–reaction layer was mainly composed of a U-rich, (U,Zr)$(Fe,Cr)_2$ compound, Zr-rich and Zr-depleted layers. Fe elements which had diffused from HT-9 were mainly observed in the boundaries of the Zr-rich particles in the U–Zr alloy.

1. Introduction

Metallic fuels such as U–Zr and U–PU–Zr alloys are being considered as nuclear fuels for sodium-cooled fast reactors (SFR) due to their excellent thermal conductivities, high densities and good breeding performance [1–5]. Ferritic/martensitic steels such as HT-9 and 9Cr steels are being recognized as candidate materials for the metallic fuel cladding tubes in SFRs due to their high thermal conductivities, low expansion coefficients and superior irradiation resistances to void swelling [6–10]. When the metallic fuels are applied to a nuclear fuel, their swelling results in an interdiffusion between the metallic fuels and cladding tubes. This interdiffusion induces the formation of intermetallic compounds that cause a degradation of the mechanical properties of the cladding tubes [11,12]. Hence, such interdiffusion behavior has become a subject of concern.

Some studies have reported that the interdiffusion behavior appeared to be dependent of the composition of the cladding materials, and HT-9 showed a thinner diffusion–reaction layer than Fe–18Cr–8Ni steel [13–15]. However, only limited results regarding U–Zr/HT-9 interdiffusion were reported. In addition, these papers report a diffusion behavior at 700 °C for less than 100 h, which is a relatively short term. Hence, this study focuses on the interdiffusion between U–10 wt.%Zr and HT-9 at 700 °C for 1000 h, as well as at 730 °C for 300 h.

2. Experimental procedure

A U–10 wt.%Zr alloy was prepared by a vacuum induction melting (VIM) process (Table 1). The alloy was homogenized at 900 °C for 100 h. HT-9 steel was also prepared by the VIM process (Table 2). The steel ingot was hot-forged at 1200 °C, followed by a normalizing at 1050 °C for 1 h and a tempering at 750 °C for 1 h. Air cooling was applied for all the heat treatments. The disk-type specimens with an 8 mm diameter and a 2 mm thickness were prepared by a machining of the U–10 wt.%Zr alloy and HT-9 steel, and their planes were polished by up to 0.05 μm-size alumina powders. The disk-type U–Zr specimen was arrayed between the disk-type HT-9 ones, and a set of these specimens was wrapped with a Ta foil to prevent a reaction with the apparatus made of type 316 stainless steel (Fig. 1). The specimens were compressively pressed with a torque of 70 Nm. The assembled diffusion couple was then sealed in a quartz tube, and an isothermal heat treatment was performed at 700 °C for 300, 500, 800 and 1000 h. An additional diffusion couple was heat-treated at 730 °C for 300 h.

After a heat treatment, the cross-sectional microstructure was observed by using a scanning electron microscope (SEM) with a back scattered electron mode for a compositional contrast, and elemental analyses were made by using an energy dispersive spectroscope (EDS) attached to a SEM. The crystalline structures were identified by using an X-ray diffraction (XRD).

3. Results and discussion

3.1. Diffusion behavior between U–10 wt.%Zr alloy and HT-9 steel

Fig. 2 shows the SEM images of an interface between the U–10 wt.%Zr alloy and HT-9 steel after a heat treatment at 700 °C for 300, 500, 800 and 1000 h. It was apparent that the diffusion–reaction layer formed in the interface between the two specimens increased with time, finally forming about a 10 μm thickness after 1000 h. After a heat treatment for 300 h, protruded U-rich phases and a Zr-rich layer were observed as an interface between the two specimens (Fig. 2a). As the diffusion proceeded, the U-rich

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Table 1
Chemical composition of U–Zr alloy (wt.%).

U	Zr	C	O	N
Bal.	10.2	0.0010	0.0151	0.0008

Table 2
Chemical composition of HT-9 steel (wt.%).

Fe	Cr	Ni	Mo	Mn	Si	C	W	V	Ti
Bal.	12.09	0.65	1.04	0.61	0.42	0.21	0.46	0.29	0.015

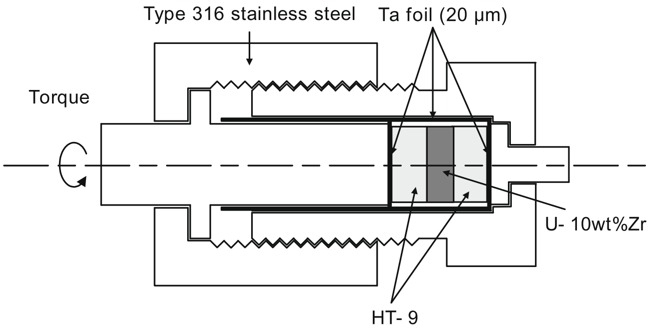

Fig. 1. Schematic drawing of the diffusion couple apparatus.

layers were extended along to the Zr-rich layer (Fig. 2b). As the diffusion proceeded further, they were further extended (Fig. 2c) and finally connected to each other after a heat treatment for about 1000 h (Fig. 2d). The SEM/EDS results of the reaction layer formed in an interface between the U–10 wt.%Zr and HT-9 after a heat treatment at 700 °C for 1000 h are shown in Fig. 3. The interdiffusion between the two specimens produced four distinctive diffusion–reaction layers from HT-9 to U–Zr alloys: U-rich, U–Fe compounds, Zr-rich, and Zr-depleted layers. The representative chemical composition of the U-rich phase was determined to be (at.%) 60U, 29Fe, 10Cr and 1Zr. In the U-rich layer marked as A in Fig. 3a, unreacted HT-9 particles were also observed. This observation means that the U atoms diffuse along to the grain boundaries of HT-9, and they spread parallel to the boundary between two alloys. Finally, they connect each other, forming the isolated HT-9 particles in the U-rich layer. Next was the U–Fe compound layer with about 6 μm in width marked as B, and its chemical composition was analyzed to be (at.%) 32U, 7Zr, 57Fe and 4Cr. Then, a Zr-rich layer with about 3 μm in width was observed marked as C, and its chemical composition was analyzed to be (at.%) 96Zr, 2U and 2Fe. The Zr-depleted layer marked as D was also observed in the U–Zr alloy, and its width appeared to be in the 5 μm range. The presence of Zr-rich and Zr-depleted layers indicates that the UZr_2 phases are decomposed into U and Zr elements, and the decomposed Zr elements are formed to the Zr-rich layer near an interface. It was reported that the Cr in the HT-9 might lead to an activation of the Zr in the UZr_2 to form a Zr-rich layer [15].

Fig. 4 shows the X-ray diffraction pattern obtained form the reaction layer between the U–10 wt.%Zr and HT-9 after a heat treatment at 700 °C for 1000 h. It was found that the reaction layer was mainly composed of several phases: U, Zr, U_6Fe, UFe_2, and Fe. It is believed that the U_6Fe, UFe_2 and Zr peaks are representative of the U-rich, U–Fe compounds and Zr-rich layers, respectively. Hence, the chemical formulations of the U-rich and U–Fe compound phases are believed to be U_6Fe and $(U,Zr)(Fe,Cr)_2$, respectively, which are in agreement with the previous studies [12–14].

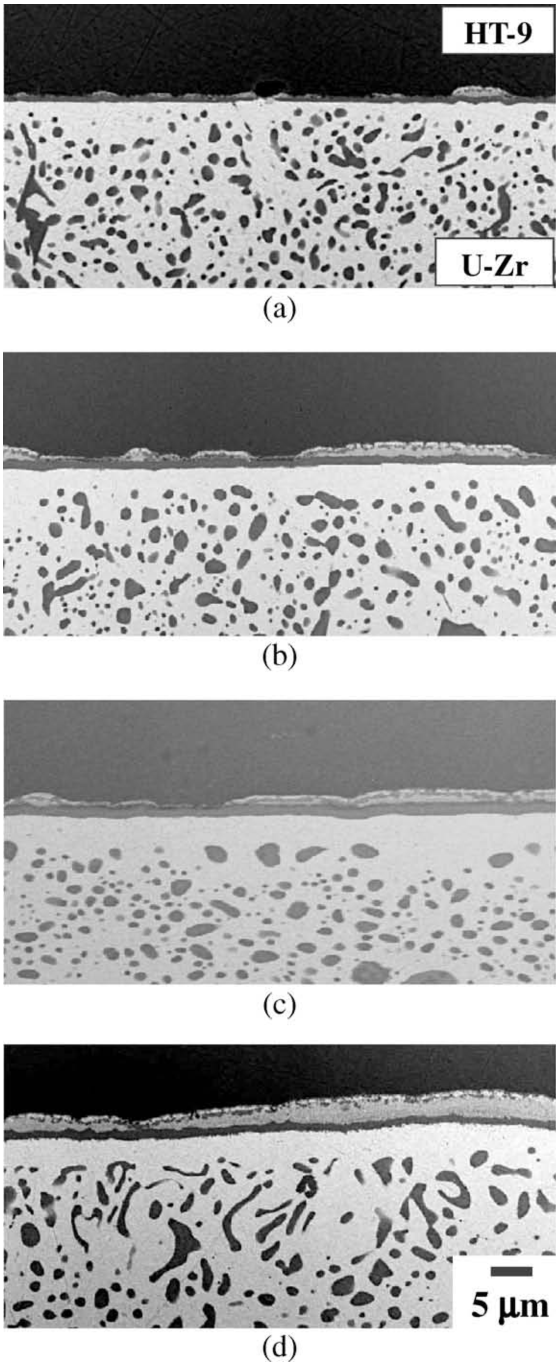

Fig. 2. SEM images on the interface between U–10 wt.%Zr alloy and HT-9 steel after a heat treatment at 700 °C for (a) 300, (b) 500, (c) 800 and (d) 1000 h.

Fig. 5 shows the SEM/EDS results from the reaction layer formed in an interface between the U–10 wt.%Zr and HT-9 after a heat treatment at 700 °C for 500 h. An examination of the distribution of the alloying elements showed that an Fe element was especially observed around the Zr-rich particles in the U–Zr alloy. This means that the Fe atoms which are diffused from the HT-9 to U–Zr alloy preferentially react with the Zr-rich particles more so than the U and δ-UZr_2 phases. It is thus considered that Fe has a higher affinity with Zr than U.

3.2. Correlation between diffusion–reaction and the Zr-rich layer

Fig. 6 shows the SEM images of the reaction layer formed in an interface between the U–Zr and HT-9 after a heat treatment at