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Thermal stability of co-extruded U–Zr/Zr–Nb alloys

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Abstract

A co-extruded U–80 at.%Zr/Zr–1 at.%Nb rod was heat-treated at 590 and 700 °C for up to 4000 h in order to investigate the thermal stability of a U–Zr fuel and the interface between the U–Zr fuel and the Zr–Nb clad. The U–Zr fuel was composed of the δ-UZr_2 matrix and α-Zr precipitates. There was little change in the microstructure during a heat-treatment at 590 °C. However, after a heat-treatment for 4000 h, uranium-rich precipitates were formed at the interface near the grain boundary of the α-Zr phase possibly due to a diffusion from the uranium supersaturated in the α-Zr phase. A heat-treatment at 700 °C induced a considerable dissolution of α-Zr into the matrix, which continuously formed a single (γ-U, β-Zr) phase. After a heat-treatment for 2000 h, the thickness of the diffusion layer in the interface between the U–Zr fuel and Zr–Nb clad was observed to be about 150 and 250 μm at 590 and 700 °**C**, respectively.

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1. Introduction

Metallic fuels such as U–80 at.%Zr could be utilized as a nuclear fuel for advanced light water reactors in the future due to their excellent thermal conductivity [1]. A Zr–Nb alloy can be recommended as a cladding material of a metallic fuel because of its excellent corrosion resistance along with reliable mechanical properties [2].

As one of the fabrication processes of metallic fuels, a billet composed of U–80 at.%Zr alloy as a metallic fuel and Zr–1 at.%Nb alloy as a cladding material can be extruded by using their excellent workability properties. An extruded metallic fuel rod usually shows a mechanical bond between a fuel and a clad [3,4]. This fact induces not only a reduced temperature gradient but also an interdiffusion at a fuel and clad interface.

A metallic fuel usually has a temperature gradient; its center part is very high while its outer region is relatively low. This temperature gradient may create two temperature regions; above and below 610 °C which is the temperature range for a phase transformation (γ-U, β-Zr ↔ α-Zr + δ-UZr_2). From this point of view, the thermal stability of a metallic fuel above and below 610 °C has become a subject of interest. The interdiffusion behavior between a fuel and clad should also be considered at these temperatures in terms of the performance of a fuel rod. However, there has been little research on the thermal stability and interdiffusion of a metallic fuel rod fabricated by an extrusion process.

In this study, an extruded metallic fuel rod consisting of U–80 at.%Zr fuel and Zr–1 at.%Nb clad was heat-treated at 590 and 700 °C in order to investigate the thermal stability of the fuel and the interdiffusion behavior of the fuel and clad interface.

2. Experimental

The uranium powders were prepared by a centrifugal atomizing process, and their shape showed spherical-type particles with an average size of about 48 μm. The zirconium powders were prepared by a hydriding–dehydriding

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process, and their average size was found to be about 88 μm. Within a glove box filled with a purified argon gas, 20 at.% uranium and 80 at.% zirconium powders were mixed together in a V-type off-axis rotating mixer at 75 rpm for 2 h. The mixed powders were uniaxially cold compacted at a compaction pressure of 428 MPa by using a double action press. The sintering of the U–Zr powder compacts was performed at 1500 °C under a vacuum of 1×10^{-4} Pa for 2 h. As a cladding material, an extruded and heat-treated Zr-1.0 at.%Nb alloy rod (30 mm*Φ*) was prepared. The U–80 at.%Zr and Zr–1 at.%Nb alloys were machined to make a billet. Both ends of the billet were sealed by using an electron beam welding machine. The billet was then covered with Cu-can which was used as both a protector from an oxidation during a heat-treatment and a lubricant during an extrusion. The billet was then heated in a box furnace under an argon atmosphere at 700 °C for 1 h and then co-extruded with an extrusion ratio of 30. The extruded rod ($5 \times 5 \times 1000$ mm) was pickle-treated to remove the Cu film formed on the outer surface of the rod, and then heat-treated in a box furnace at 570 and 700 °C for up to 4000 h.

The sintered density of the sintered U–Zr alloy was measured and the resultant phase was identified by using a X-ray diffraction (XRD). The microstructure was examined by using a scanning electron microscope (SEM) for a compositional contrast by using a back scattered electron image, a transmission electron microscope (TEM), and an image analyzer (IA). Elemental analyses were made by using an energy dispersive spectroscope (EDS).

3. Results and discussion

3.1. Microstructure

Fig. 1 shows the microstructure of the sintered U–Zr fuel. The sintered fuel consisted of a hcp δ-UZr_2 matrix and hcp α-Zr precipitates. The chemical compositions of the δ-UZr_2 matrix and α-Zr precipitates were found to be (at.%) 80.12U, 19.88Zr and (at.%) 0.94U, 99.06Zr, respectively. Un-reacted zirconium and uranium particles were not detected while a small amount of pores was occasionally observed. The sintered density was measured to be 8.56 g/cm^3 (98.7%TD).

Fig. 2 shows the microstructures in the transverse and longitudinal sections of the extruded U–Zr/Zr–Nb rod. The microstructures in the transverse section of the rod revealed the δ-UZr_2 matrix with lath-type α-Zr precipitates (Fig. 2(a)) and a wavy-shaped interface due to a difference in the deformation strength during an extrusion at a high temperature (Fig. 2(b)). Meanwhile, the microstructures in the longitudinal section of the specimen exhibited an elongated α phase in the δ matrix (Fig. 2(c)) and a metallurgical bond due to the plastic flows of the two-kind materials (Fig. 2(d)).

3.2. Thermal stability of the U–Zr fuel

Fig. 3 shows the microstructures of the U–Zr fuel heat-treated at 590 and 700 °C for 500, 2000 and 4000 h. There was little change in the microstructure during a heat-treat-

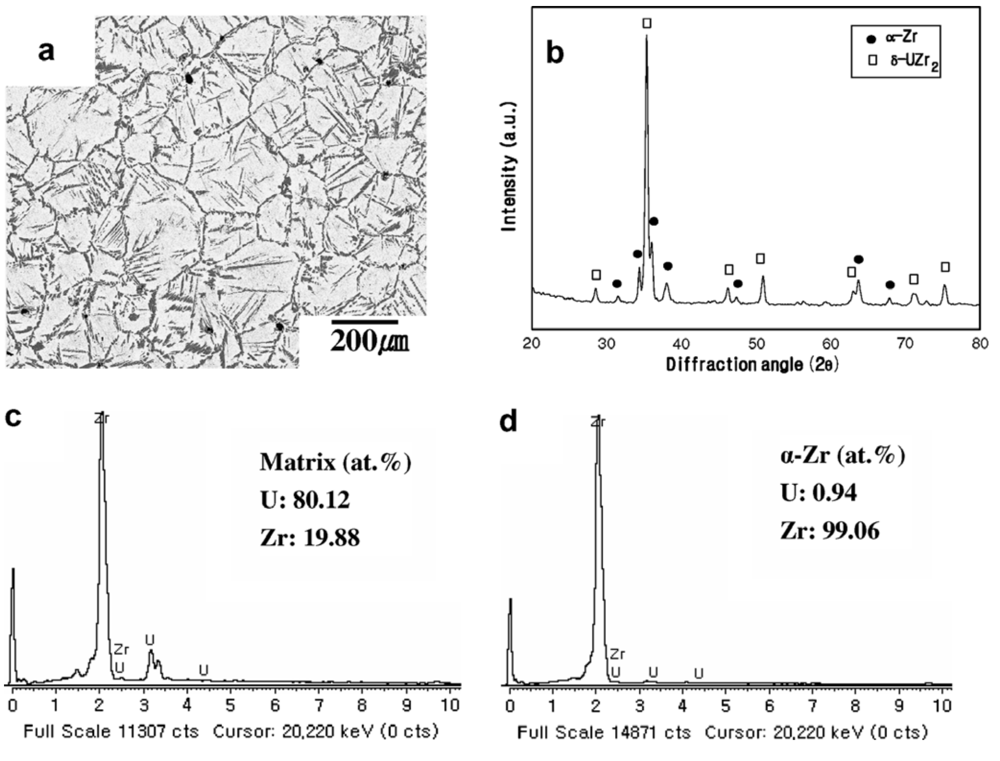


Fig. 1. (a) Microstructure, (b) X-ray diffraction pattern of the sintered U–Zr fuel, EDS analysis results of (c) white δ-UZr_2 matrix and (d) gray α-Zr phase.

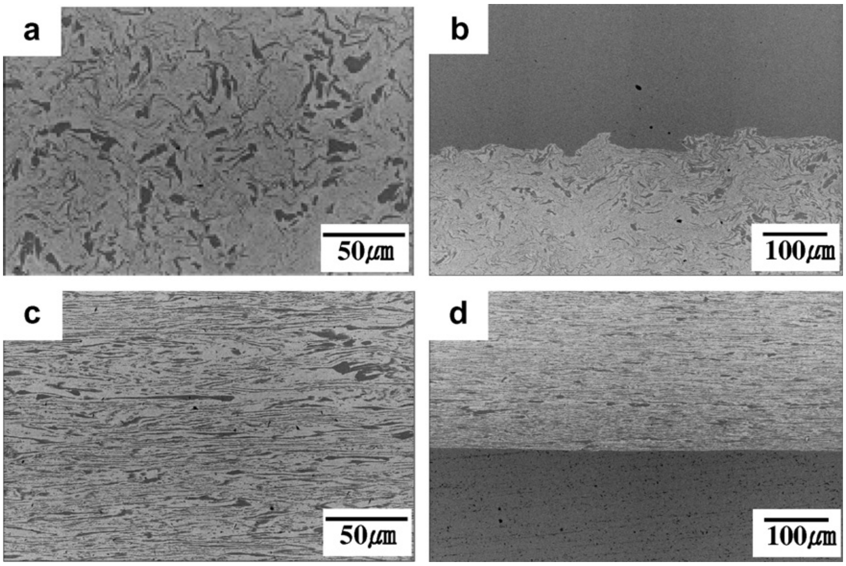


Fig. 2. Microstructures of the extruded U–Zr/Zr–Nb rod; (a) fuel, (b) fuel and clad interface in the transverse section, (c) fuel and (d) fuel and clad interface in the longitudinal section.

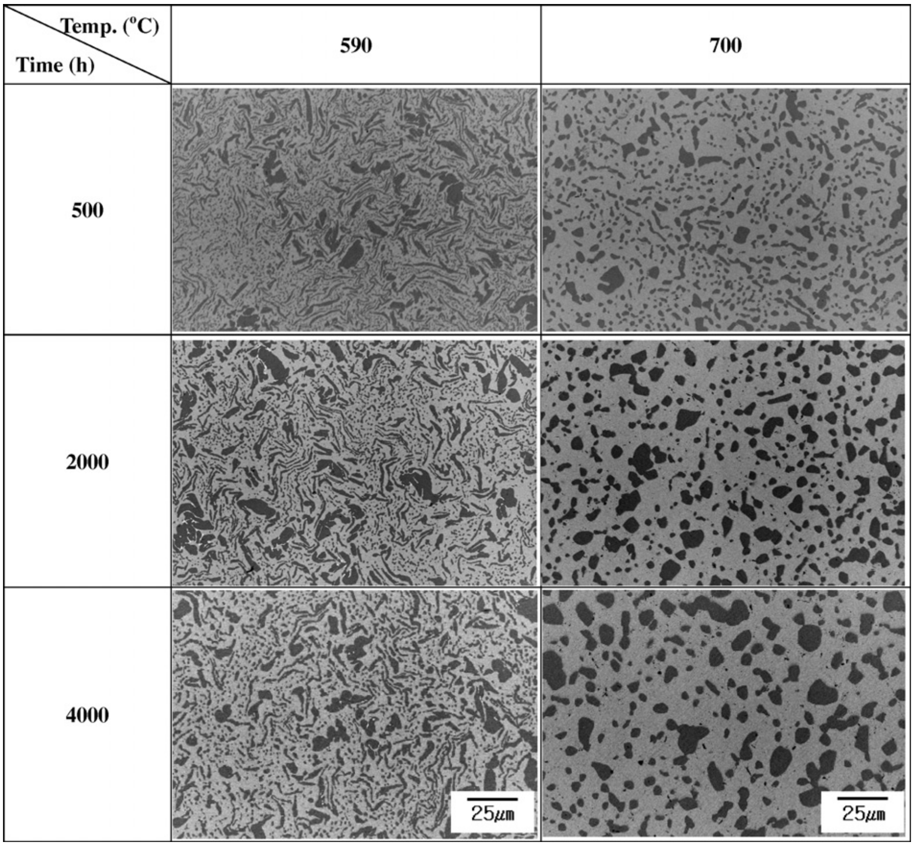
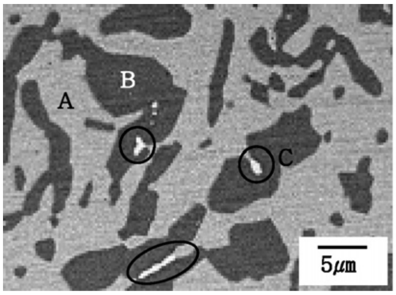


Fig. 3. Microstructures of the U–Zr fuel after a heat-treatment at 590 and 700 °C for 500, 2000 and 4000 h.

ment at 590 °C. However, the heat-treatment at 700 °C induced a morphological change of the α-Zr precipitates from a lath-type to a round-type. This observation means that the α-Zr phase which had been dissolved into the δ-UZr_2 matrix continuously formed a single phase of (γ-U, β-Zr) above 610 °C.

The uranium-rich precipitates formed in the U–Zr fuel after a heat-treatment at 590 °C for 4000 h are shown in



Position	Phase	U(at.%)	Zr(at.%)
A	δ-UZr_2	29.7	70.3
B	α-Zr	0.9	99.1
C	U-rich	65.3	34.7

Fig. 4. Uranium-rich precipitates formed in the U–Zr fuel after a heat-treatment at 590 °C for 4000 h.

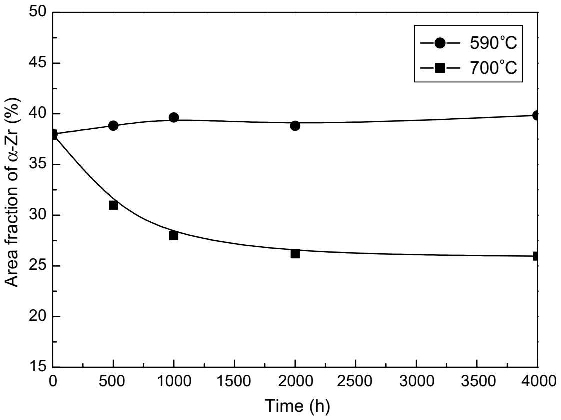


Fig. 5. Variation of the area fraction of the α-Zr phase in the U–Zr core with a heat-treatment at 590 and 700 °C.

Fig. 4. It was interesting to note that particles with a brighter white color were observed near the boundary of the α-Zr phase, and its composition was determined to be 65.3 at.%U and 34.7 at.%Zr. The solubility of uranium in a α-Zr phase is below 1.0 at.% [5,6]. It is considered that the uranium element which was supersaturated in the α-Zr phase was diffused from the grain to the grain boundary, thus forming uranium-rich precipitates during a heat-treatment at this temperature.

The variation of the area fraction of the α-Zr phase in the U–Zr fuel during a heat-treatment at 590 and 700 °C is plotted in Fig. 5. There was little change in the area fraction of the α-Zr phase during a heat-treatment at 590 °C since the δ-UZr_2 and α-Zr phases were stable below 610 °C [5]. However, the heat-treatment time at 700 °C

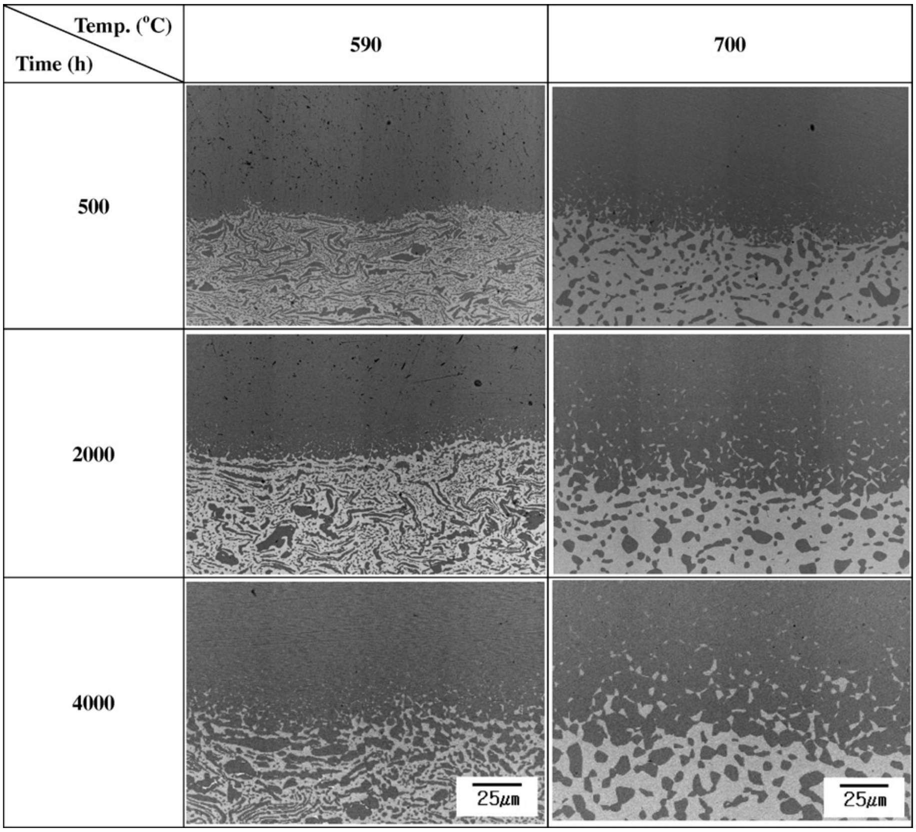


Fig. 6. Microstructural change of the interface between the U–Zr fuel and Zr–Nb clad during a heat-treatment at 590 and 700 °C.

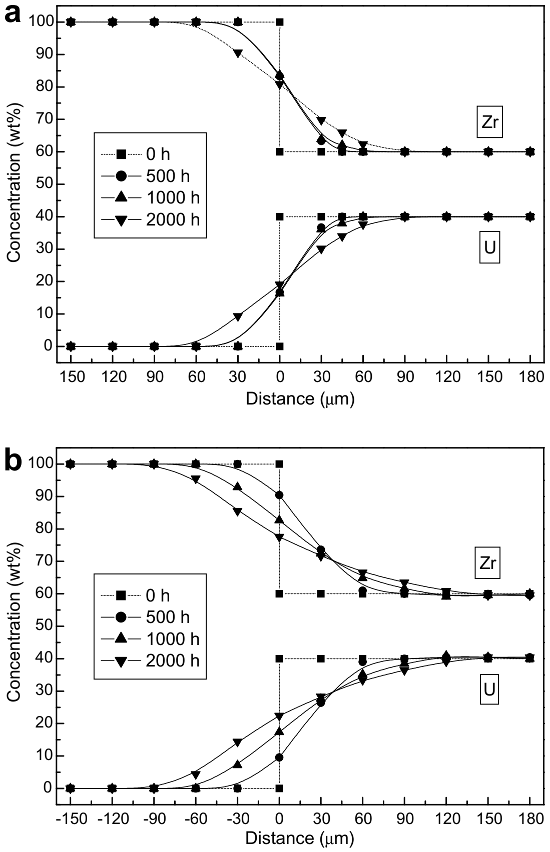


Fig. 7. Distribution of the uranium and zirconium concentrations in the U–Zr/Zr–Nb interface during a heat-treatment at (a) 590 °C and (b) 700 °C.

caused a reduced area fraction of the α-Zr phase in the U–Zr core with an increase in the time due to a considerable dissolution of α-Zr into the matrix to form a single (γ-U, β-Zr) phase.

3.3. Interdiffusion between the U–Zr core and Zr–Nb clad

Fig. 6 shows the microstructural change of the interface between the U–Zr fuel and Zr–Nb clad during a heat-treatment at 590 and 700 °C. Reaction layer in the interface increased with an increase in the heat-treatment time at these temperatures. The uranium element in the U–Zr fuel appeared to diffuse along the grain boundaries of the Zr–Nb clad, finally forming facet-type grains in the Zr–Nb clad especially after diffusion at 700 °C for 4000 h. Except for the niobium in the Zr–1%Nb clad, the interdiffusion system in the present study is simply composed of two phases, uranium and zirconium. This means that there is little possibility to make any other intermediate phases as a result of interdiffusion between the U–Zr fuel and the Zr–Nb clad. In the U–Zr–Nb ternary system considered the niobium, several intermediate phases would be formed. However, the niobium concentration in the Zr–Nb clad was as low as 1%, and the results of XRD study did not show any other intermediate phases due to its limited accuracy.

The distribution of the uranium and zirconium concentrations near the U–Zr/Zr–Nb interface during a heat-treatment at 590 and 700 °C is plotted in Fig. 7. As the heat-treatment time was increased, the uranium in the U–Zr fuel diffused into the Zr–Nb clad while the zirconium in the Zr–Nb clad diffused into the U–Zr fuel. After a heat-treatment for 2000 h, the width of the reaction layer was measured to be about 150 and 250 μm at 590 and 700 °C, respectively.

4. Conclusion

A billet composed of U–Zr fuel and Zr–Nb clad was extruded, and the extruded rod was heat-treated at 590 and 700 °C for up to 4000 h. The extruded rod revealed a metallurgical bond at the fuel and clad interface, with an excellent thermal conductivity. The thermal stability of the U–Zr fuel at 590 °C was found to be stable, and a constant area fraction of the α-Zr phase was observed. However, uranium-rich precipitates were detected near the grain boundary of the α-Zr phase, possibly due to a diffusion of a supersaturated uranium element from the α-Zr phase. Meanwhile, the heat-treatment at 700 °C induced a dissolution of the α-Zr phase into the matrix, resulting in a decrease in the area fraction of the α-Zr phase. The uranium element in the U–Zr fuel appeared to diffuse along the grain boundaries of the Zr–Nb clad, finally forming a facet type grain in the Zr–Nb clad. Reaction layer at the interface between the U–Zr fuel and Zr–Nb clad increased with an increase in the heat-treatment time. After a heat-treatment for 2000 h, the width of the reaction layer was found to be about 150 and 250 μm at 590 and 700 °C, respectively.

Acknowledgements

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