THERMAL STABILITY OF THE U-Zr FUEL AND ITS INTERFACIAL REACTION WITH LEAD

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Abstract

The effect of heat treatment on fuel rods at 630°C and 700°C and the interfacial reaction between fuel and lead were investigated. The U-Zr metallic fuel was fabricated by mixing, pressing, sintering and extrusion. There were two kinds of phases – α-Zr precipitates and a δ-UZr_2 matrix in the U-Zr metallic fuel. After heat treatment of the extruded rod at 630°C and 700°C, the volume changes of the samples increased slightly and the density variation was negligible. Therefore, it is evident that U-Zr fuels have good thermal stability. The interface between U-55Zr fuel and Pb according to annealing time at 650°C consisted of two distinctive regions – a reaction zone in the vicinity of the surface and an initial zone in the inner area. It should be noted that the thickness of the reaction zone was 26 μm, 36 μm and 46 μm at 100 hrs, 200 hrs and 1 000 hrs, respectively. Also, the reaction zone consisted of an α-Zr layer and a Zr-depleted area.

Introduction

The blanket fuel assembly for HYPER (hybrid powder extraction reactor) contains a bundle of pins arrayed in a triangular pitch, which has a hexagonal bundle structure. The reference blanket fuel pin consists of the fuel slug of the TRU-xZr (x = 50-60 wt.%) alloy and is immersed in lead for thermal bonding with the cladding. The blanket fuel cladding material is ferritic-martensitic steel HT9.

Although there are lots of experimental data on the metallic alloys of U-Pu-Zr and U-Zr, they are for fuel types with a Zr fraction of less than 20 wt.%. Therefore, little data is available for the HYPER system fuel where the Zr fraction is higher than 30 wt.%. As a basic study on HYPER fuel, we fabricated U-55 wt.% Zr fuel instead of the actual TRU-Zr fuel. It appeared that no experimental data pertinent to the TRU-xZr (x = 50-60 wt.%) alloy existed. The U-55 wt.% Zr metallic fuel was fabricated by mixing, pressing, sintering and extrusion. This work was performed in order to investigate the microstructures and the thermal stability of the U55 wt.% Zr metallic fuel and the interfacial reaction between U-55 wt.% Zr fuel and Pb according to annealing time at 650°C.

Experimental procedure

The uranium powder was manufactured by a centrifugal atomiser and the zirconium powder (Sejong materials Co. Ltd, Korea) was prepared via the hydride-dehydride process.

The U-Zr metallic fuel was performed by mixing, pressing and sintering in optimum compaction and sintering conditions [1]. The sintered U-60 wt.% Zr was extruded by an indirect extrusion machine at 760°C and with a 13:1 extrusion ratio. Specimens of 25 mm in length were cut, vacuum-sealed in quartz tubes and annealed in a box furnace at 630°C and 700°C for up to 1 500 hrs. Sample swelling as a function of temperature and time was determined from dimensional changes. The densities of the annealed samples were calculated from the weights and dimensions. The microstructures and phase identification of the extruded rod and the annealed rod were examined by SEM and EDS. The area fraction of each phase obtained by SEM was analysed by a BMI plus ver. 4.0 (Winatech, Korea).

In order to clarify interfacial reaction between fuel and lead, a series of experiments were executed with fuel in Pb melt at 650°C for 100, 200 and 1 000 hrs. The composition of diffusion layers and the diffusion depth were analysed using SEM/EDS.

Results and discussion

Figure 1 shows uranium and zirconium particles. The mean particle sizes of the uranium and zirconium powders are 55 μm and 60 μm, respectively. Most of the uranium particles have a smooth surface and a generally near-perfect spherical shape with a few attached satellites. On the other hand, the zirconium particles fabricated by the hydride-dehydride process have an irregular morphology.

Figure 2 shows the back-scattered electron (BSE) image of the sintered sample. As can be seen in phase diagrams, the α-Zr phases are distributed in the δ phase, which is observed as a white matrix [2]. Also, small amounts of pores are found throughout the sample.

Figure 1. Photographs of the atomised (a) U powder and (b) Zr powder

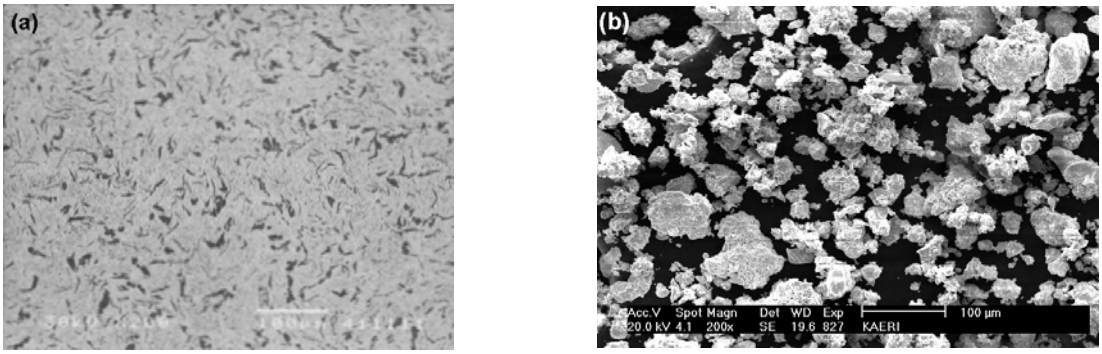
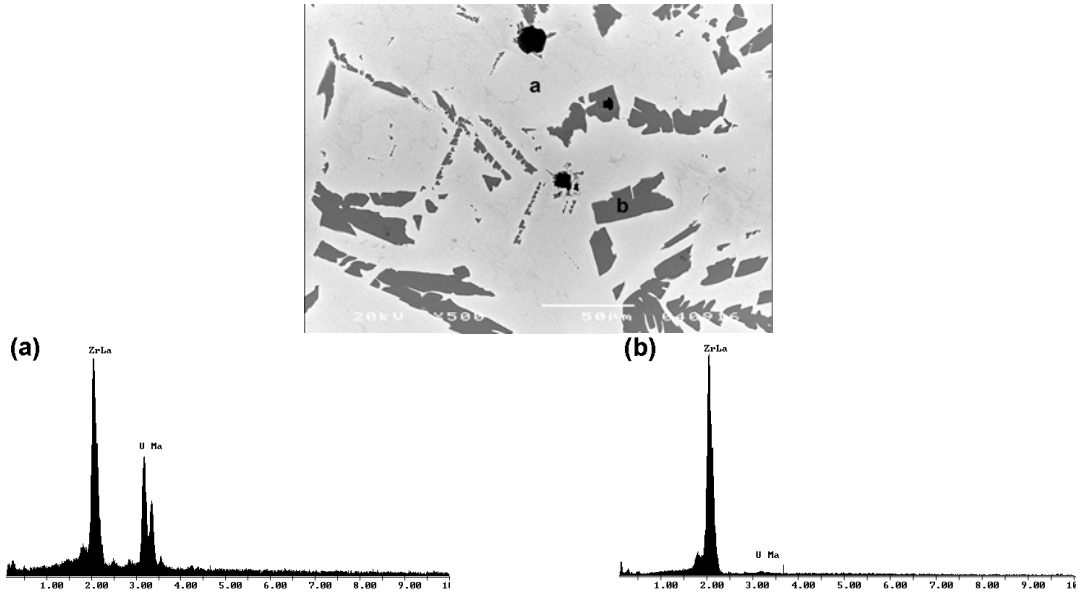


Figure 2. SEM micrograph and EDS analysis results for U-60 wt.% Zr sintered sample

(a) δ-UZr_2 matrix, (b) α-Zr phase



BSE images of the hot extruded rod are shown in Figures 3 and 4. In the sintered samples mentioned above, the α-Zr phases were also distributed (homogenously) in the white δ phase matrix and the porosity was drastically decreased because of the densification by the extrusion process. It was confirmed that the U-55 wt.% Zr alloy with a high melting point could be fabricated via a sintering process at a relatively low temperature in lieu of a conventional casting process.

Figure 5 shows the dependency of swelling behaviour and the density changes of temperature and time for the samples. The volume changes of the samples increased slightly and the density variation was negligible. Therefore, it is evident that U-55 wt.% Zr fuel has good thermal stability.

Figure 3. SEM micrographs of the extruded rod

(a) Transverse direction, (b) longitudinal direction

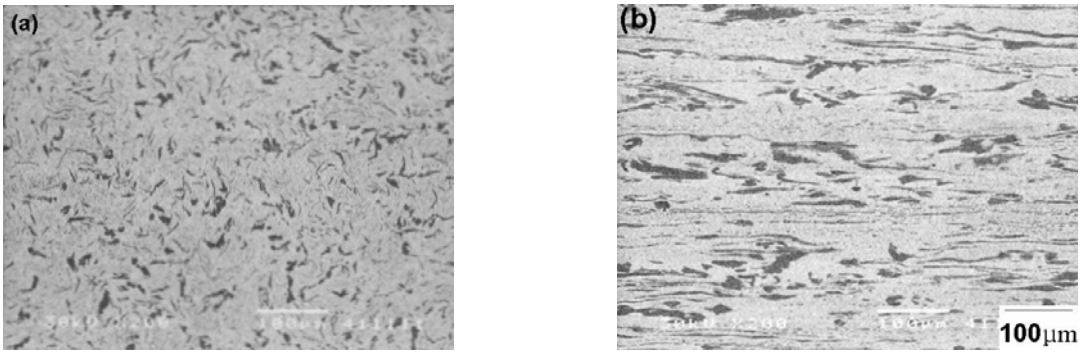


Figure 4. SEM micrograph and EDS analysis results for the extruded rod

(a) δ-UZr_2 matrix, (b) α-Zr phase

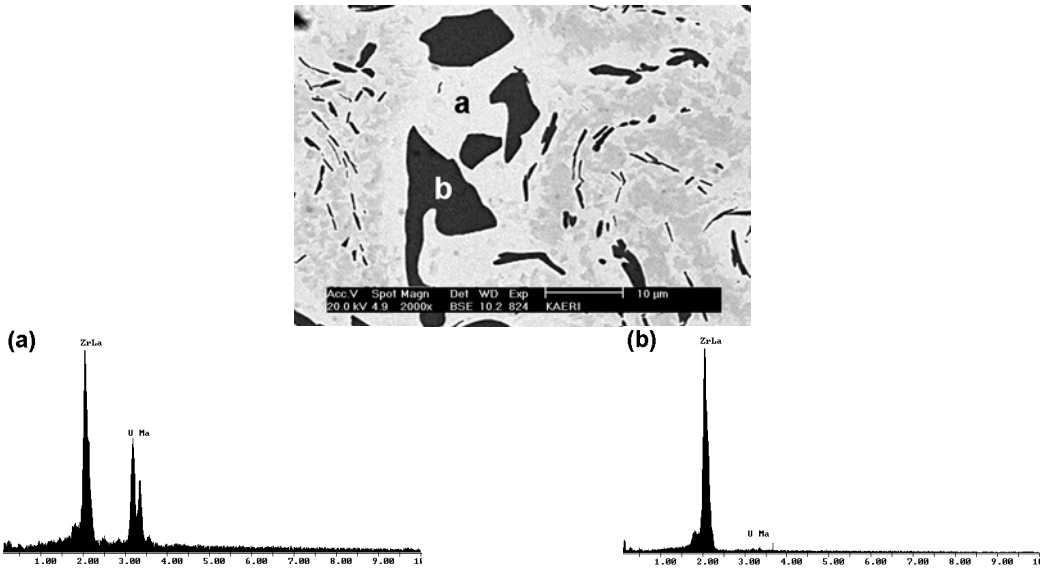


Figure 5. Volume and density changes of the extruded rod on temperature and time

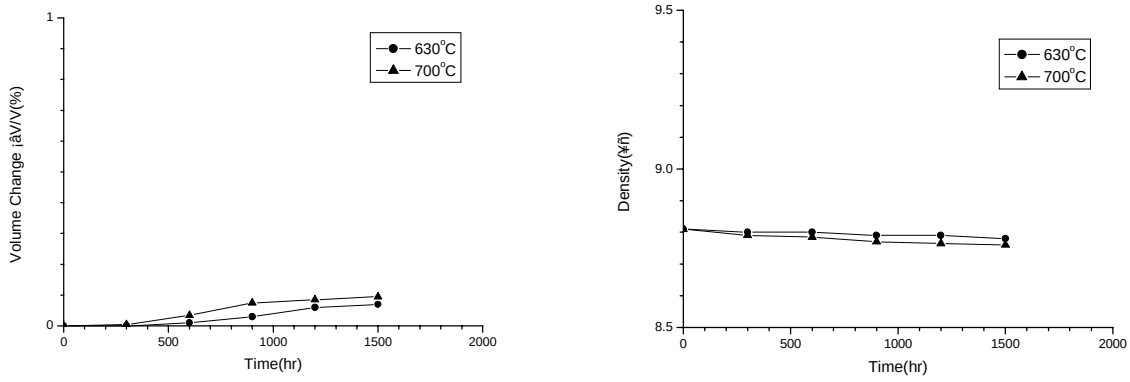


Figure 6 shows BSE images of the rod annealed at 630°C and 700°C for 600 hrs and 1 500 hrs. These figures show that as the annealing time increased, the lathed α-Zr phases began to break into pieces and grow into spherical particles. As the annealing temperature increased, spheroidising of the α-Zr phases was accelerated, with transformation to a spherical shape. We can interpret that the geometrically unstable lathed α-Zr phase transformed to the spherical phase with a lower surface area by coalescence. In addition, we think that the α-Zr and δ-UZr_2 phases dissolved and formed a solid solution at an annealing temperature above 617°C, and that the spheroidised α-Zr and δ phases appeared again during cooling.

As the annealing time increased, the hardness decreased and the decreasing rate of the hardness was much higher at 700°C than at 630°C (Figure 7). This is attributed to the increase of the area fraction of the δ phase and the decrease of the area fraction of the relatively hard α-Zr phase as the annealing temperature and time increased.

Figure 8 shows the area fraction of the α-Zr phase according to the annealing time at 630°C and 700°C. In the case of 630°C of annealing time, the area fraction of the α-Zr phase gradually decreased as the annealing time increased, whereas the area fraction of the α-Zr phase sharply decreased at the early stage of annealing time and then the decreasing rate declined at 700°C. This phenomenon agreed with the results on hardness values vis-à-vis annealing time as shown in Figure 7. When the U-55 wt.% Zr alloy is annealed at above 617°C, which is the unstable region of α-Zr phase, the alloy is diffused into the uranium matrix and finally forms δ phase at a lower temperature. Hence, it can be interpreted that the drastic decrease of area fraction of the α-Zr phase at the early stage of annealing time was due to the higher concentration gradient of Zr, which is the driving force of Zr diffusion.

Figure 6. SEM micrographs of the U-55 wt.% Zr fuel annealed at 630°C and 700°C for 600 hrs and 1 500 hrs

(a)(b) 630℃, (c)(d) 700℃, (a)(c) 600 hrs, (b)(d) 1 500 hrs

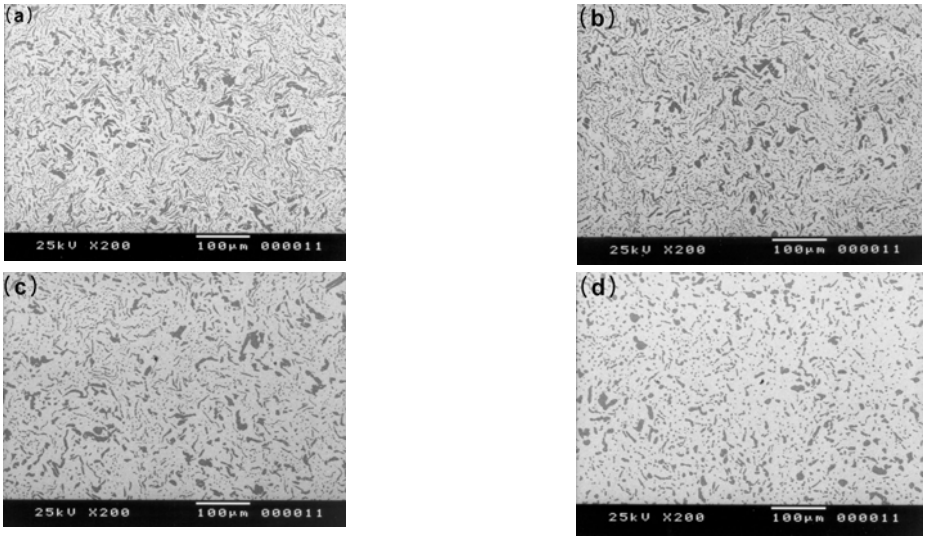


Figure 7. Variation of room temperature hardness of rods after annealing at 630°C and 700°C

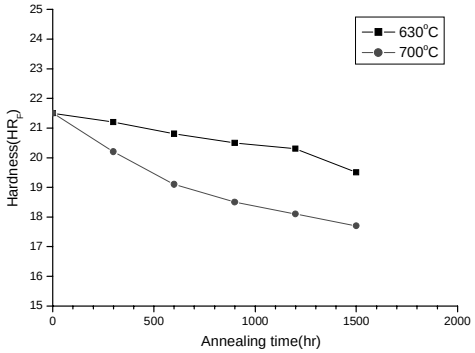


Figure 8. Variation of area fraction of α-Zr phase with annealing times at 630°C and 700°C

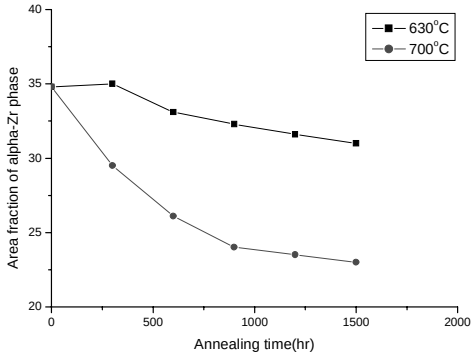


Figure 9 shows EDS line profile results on the interface between U-55Zr and Pb according to annealing time at 650°C. The microstructures of each sample consisted of two distinctive regions – a reaction zone in the vicinity of the surface and an initial zone in the inner area. It should be noted that the thickness of the reaction zone was 26 μm, 36 μm and 46 μm at 100 hrs, 200 hrs and 1 000 hrs, respectively, as the annealing time increased. The reaction zone also consisted of two regions – an α-Zr layer and a Zr-depleted area. The α-Zr layer may be formed by a diffusivity difference between U and Zr atoms (i.e. Zr atom diffuses into the Pb melt during annealing while U is relatively intact due to lower diffusivity). The TRU in the metallic fuel is reported to react with stainless, as a cladding material then forms eutectic at a low temperature. Thus, it is anticipated that the α-Zr layer should effectively act as a reaction barrier with the cladding material.

In order to closely investigate the reaction layer, EDS analysis was performed as shown in Figure 10. The analysis shows that a negligible amount of Pb is present in the very outer layer of the reaction zone with Zr-rich phase (region A). And the α-Zr phase forms a thick layer of ~10 μm underneath the surface (region B). Region C consists of U-rich phase and Zr formed by the decomposition of δ phase. As the annealing time increased, the thickness of the reaction layer from A to C increased to the direction of as-extruded area, region D.

Figure 9. Line profiles of interface of U-55Zr fuel with Pb bonding according to annealing time at 650°C

(a) 100 hrs, (b) 200 hrs, (c) 1 000 hrs

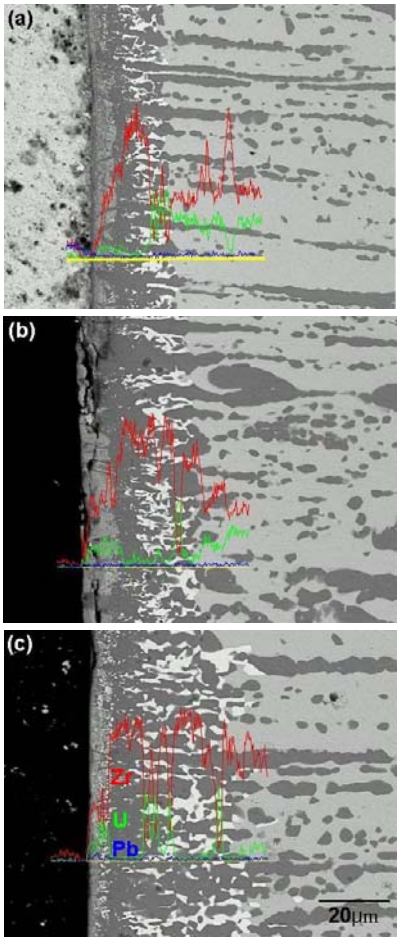
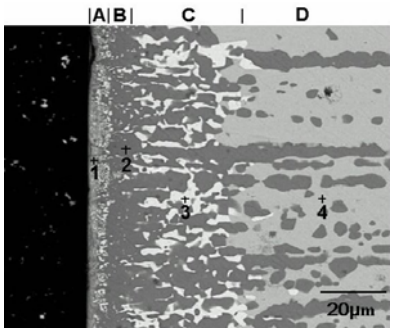


Figure 10. SEM/EDS results of U-55Zr with Pb bonding for 1 000 hrs at 650°C



Mark	Pb	U	Zr
1	0.76	19.11	80.13
2		0.37	99.63
3		95.11	4.89
4		31.82	68.18

Figures in at.%.

Conclusions

- The α-Zr phases were distributed in the white δ-UZr_2 phase matrix and the porosity was drastically decreased due to densification from the extrusion process.

- The volume changes of the samples increased slightly and the density variation was negligible. Therefore, it is evident that U-55 wt.% Zr fuel has good thermal stability.

- As annealing time increased, hardness decreased and the decreasing rate of hardness was much higher at 700°C than 630°C. This is attributed to the increase of area fraction of δ phase and the decrease of area fraction of relatively hard α-Zr phase as annealing temperature and time increased.

- The interface between U-55Zr fuel and Pb at annealing time and 650°C consisted of a reaction zone and an initial zone in the inner area. The reaction zone consisted of a α-Zr layer and a Zr-depleted area with thicknesses of 26 μm (100 hrs), 36 μm (200 hrs), 46 μm (1 000 hrs).

REFERENCES

[1] Cho, H.S., *et al*., *Proceedings of the Korean Nuclear Society Spring Meeting*, Jeju, Korea, May 2001.

[2] *Phase Transformation in Materials*, 2nd Ed., D.A. Porter, K.E. Easterling (1992).

SUMMARIES OF TECHNICAL SESSIONS

Chairs: R. Sheffield, B-H. Choi

The summaries contained in this section reflect the views of the respective Chairperson(s) and do not necessarily reflect the views of the technical session group members.

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