

## Effects of Extrusion and Heat-treatment on the Corrosion of Zr-Nb Alloy

T.K. Kim, P.S. Choi, S.K. Yang, U.J. Lee, G.S. Joo, Y.M. Ko, C.T. Lee, D.S. Sohn  
Advanced Nuclear Fuel Development Division, KAERI, 105, Deogjin, Yuseong, Daejeon, 305-353, tkkim2@kaeri.re.kr

### 1. Introduction

Zr-Nb alloys have drawn wide attention as cladding materials of nuclear fuels due to their superior corrosion resistances and excellent mechanical properties [1-2]. In the fabrication processes of metallic fuel rods, the billets combined with a Zr-U alloy for a fuel core and the Zr-Nb alloy for a clad are usually subjected to a hot-extrusion at high temperatures [3]. The deformed fuel rods should be annealed to relieve the stress from the hot-deformation. The heat-treatment conditions thus became a subject of main concern [4]. This study was focused on the effects of the extrusion and the heat-treatment on the corrosion of Zr-Nb alloy.

### 2. Methods and Results

#### 2.1 Experimental procedure

The Zr-Nb alloy for the clad materials of the fuel rods was prepared in extruded and annealed conditions. The alloy was machined, and then extruded at high temperatures. The extruded rods were heat-treated at 580°C for 3 and 16 hours. The microstructures of the alloys were observed by TEM/EDS. The microhardness was observed, and the corrosion tests were performed at 400°C in steam for 60 days using a static autoclave.

#### 2.2 Microstructure

Figure 1 shows the microstructures of Zr-Nb alloys. The as-received alloy revealed the equiaxed structure with an average grain size of about 4.5  $\mu\text{m}$  (Fig.1a). Most of precipitates in the alloy were identified to be enriched- $\beta$  phases with a Nb-concentration range from 25 to 70 wt.%, and they also showed a spherical shape with a size range from 10 to 50 nm. On the other hand, the as-extruded alloy revealed the average grain size of about 0.5  $\mu\text{m}$  along with a high density of dislocations possibly formed during the plastic deformation of extrusion (Fig 1b). Two kinds of Nb-containing precipitates,  $\beta$ -Zr and enriched  $\beta$  were observed. Most of precipitates were  $\beta$ -Zr with Nb-concentration of less than 20 wt. %. The heat-treatment of the extruded alloys at 580°C for 3 hours produced the grain growth along with the reaction of phase transformation ( $\beta$ -Zr  $\rightarrow$   $\alpha$ -Zr + enriched  $\beta$ ), resulting in the increase in the Nb-concentration of precipitates (Fig. 1c). The additional heat-treatment of the extruded alloys

at 580°C up to 16 hours provided the grain growth up to about 45  $\mu\text{m}$  as well as the enrichment of Nb-concentration in the precipitates (Fig. 1d).

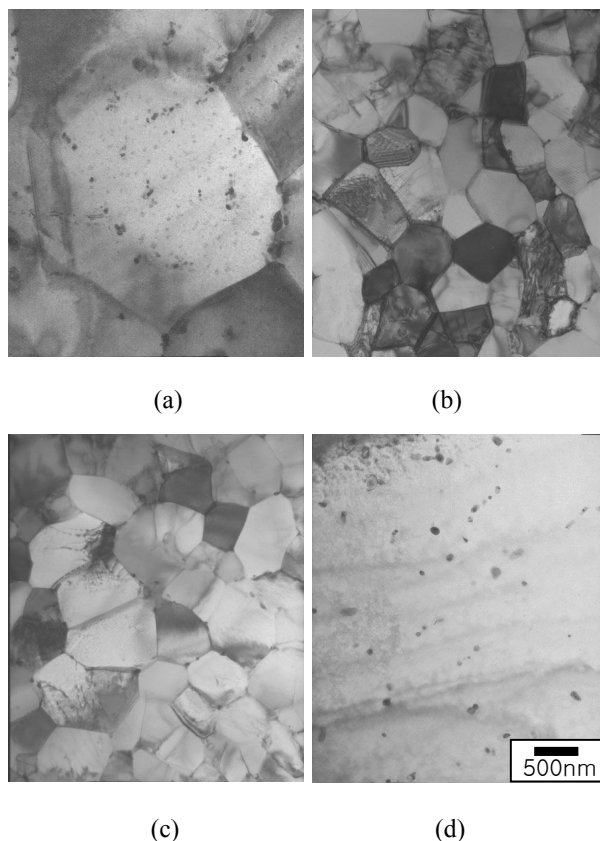


Figure 1. Bright field TEM images of Zr-Nb alloys; (a) as-received, (b) as-extruded, and heat-treated at 580 °C for (c) 3 and (d) 16 hours.

#### 2.3 Effects of extrusion and heat-treatment on the microhardness

Fig. 2 shows the effects of an extrusion and heat-treatment on the microhardness of the as-received Zr-Nb alloy. It is apparent that the extrusion led to an increase in the hardness mainly due to the reduction in the grain size. The heat-treatment of the as-extruded alloy resulted in the reduction in the hardness mainly due to the increase in the grain size. In the aspect of mechanical properties, this observation would provide fundamental data to select the

optimum heat-treatment condition after an extrusion of Zr-Nb alloy which is the cladding materials of metallic fuel.

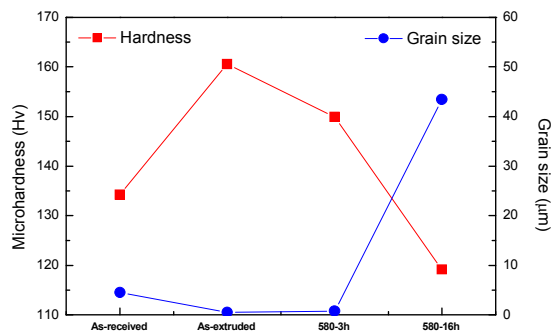


Figure 2. Effects of extrusion and heat-treatment on the grain size and microhardness of the Zr-Nb alloys.

#### 2.4 Effects of extrusion and heat-treatment on the corrosion

Figure 3 shows the effects of extrusion and heat-treatment on the corrosion of the Zr-Nb alloy at 400°C in steam. The corrosion behavior exhibited that the corrosion rate was rapid in the initial corrosion period, but decreased greatly after approximately 30 days. This is closely correlated with the increase in the thickness of a protective oxide layer formed in the surface of Zr-Nb alloy. After corrosion for 60 days, the final weight gain of the as-received alloy showed to be 37 mg/dm<sup>2</sup> whereas that of the as-extruded alloy revealed to be 45 mg/dm<sup>2</sup>. This means that the extrusion would act to decrease the corrosion resistance of the alloy. It would be mainly ascribed to the phase transformation of the precipitates from enriched-β to β-Zr during a pre-heating at high temperatures before the extrusion, including the change in the size, morphology and Nb-concentration of the precipitates. However, it was shown that the heat-treatment of the extruded alloy at 580°C for 3 hours provided to increase the corrosion resistance. Prolonged heat-treatment up to 16 hours also promoted to increase the corrosion resistance. It would be mainly attributed to the change in the properties of precipitates; the heat-treatment led to the phase transformation of precipitates from β-Zr to enriched-β (β-Zr → α-Zr + enriched β) which was identified from TEM/EDS studies as shown in Fig. 1. In the aspect of corrosion behavior, these results would provide another fundamental data to select the optimum heat-treatment condition after an extrusion of Zr-Nb alloy.

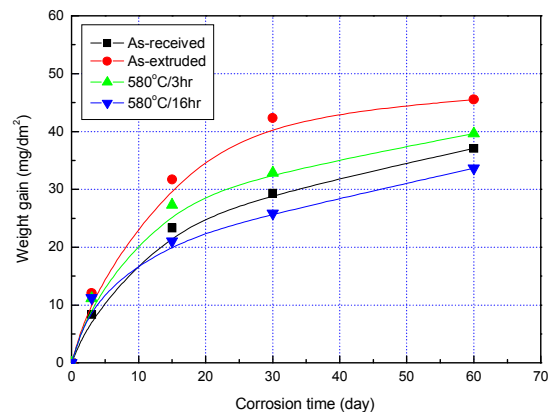


Figure 3. Effects of extrusion and heat-treatment on the corrosion of Zr-Nb alloy at 400°C in steam.

### 3. Conclusion

The extrusion of the as-received Zr-Nb alloy resulted in the increase in the hardness and decrease in the corrosion resistance. However, the heat-treatment at 580°C of the extruded alloy induced to decrease in the hardness and increase in the corrosion resistance. It would be ascribed to the grain size, the phase transformation of precipitates, and so on. This indicates that the mechanical properties and corrosion behavior of the extruded alloy intimately depends on the heat-treatment conditions. Therefore, these results would be useful to select the optimum heat-treatment conditions after an extrusion of the as-received Zr-Nb alloy.

#### Acknowledgements

The authors would like to express their appreciation to the Ministry of Science and Technology (MOST) of the KOREA for the support of this work.

#### REFERENCES

- [1] G.P. Sabol, G.R. Kiop, M.G. Balfour and E. Roberts, Zirconium in the Nuclear Industry, ASTM Spec. Tech. Publ. **1023**, p. 227, 1989.
- [2] J.P. Mardon, G. Garner, P. Beslu, D. Charquer, J. Senevat, in: Proceedings of the 1997 International Topical Meeting on LWR Fuel Performance, Portland, OR, 2-6 March, p. 405, 1997.
- [3] T.K. Kim, J.H. Park, S.K. Yang, G.S. Joo, J.S. Song, Y.M. Ko, C.T. Lee, D.S. Sohn, "Proceedings of the Korean Nuclear Society Spring Meeting", May 27-28, 2004, Gyeongju, Korea.
- [4] T.K. Kim, B.S. Choi, Y.H. Jeong, D.J. Lee, M.H. Chang, J. Nucl. Mater. Vol. **301**, p. 81, 2002.