Effect of Niobium on hydride embrittlement of zirconium alloys

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1. Introduction

Zirconium based alloys have been used in cladding materials. Nowadays, advanced fuel cladding materials is developed to enhance the corrosion resistance to keep pace with the high burnup operation in a nuclear power plant. Recent trends of alloying design are increasing of Nb contents and decreasing of Sn contents in order to enhance the corrosion resistance. However, there has existed little idea that the change of niobium (Nb) has an impact on the hydrogen behavior of zirconium alloys.

The objectives in this study are to investigate the effect of Nb on hydrided zirconium alloy. Hydrogen was charged into various zirconium alloys with different Nb contents. Tensile test was conducted on the hydrogen charged these alloys and the mechanical properties such as the strength and elongation were evaluated.

2. Materials

Materials used in this study are the zirconium alloys with the different Nb contents. Unalloyed zirconium was used as a reference material. P2 and P3 which contain 1, 2wt% Nb were manufactured in order to investigate the effect of Nb on hydrogen behavior of zirconium alloy. All the materials contained about 1000 ppm of oxygen. It was thought that the oxygen was absorbed during manufacturing process.

All the materials are fabricated by hot rolling and cold rolling as shown in Figure 1. Final heat treatment of all the manufactured materials was performed at 510 °C for 2.5 hours.



Figure 1 The manufacturing process of zirconium alloys

3. Experimental procedure

The samples were charged with hydrogen using high temperature cathodic hydrogen charging method [1]. Hydrogen was charged with the current density of 0.2 A/cm² at 80 °C in 1N H₂SO₄ solution and hydrogen concentration (C_H) was controlled by charging time. After hydrogen charging, samples were homogenized at 400 °C for 24 hr.

Sheet type tensile test specimen was cut from samples such that the tensile axis lied parallel to the transverse direction of the plate. It was the gage length of 12.5 mm and the thickness of 0.88 mm as shown in figure 2. tensile test was performed at room temperature and a strain rate of 10^{-4} s⁻¹. Absorbed hydrogen contents (C_H) in each specimen were measured after the tensile test. Fracture surface of tested specimens was observed by SEM.



Figure 2 The schematic of tensile test specimen

4. Result and Discussion

4.1 hydride morphology

Unalloyed Zr had fully recrystallized structure. Nb added Zr alloy showed partially recrystallized structure. With increasing of Nb contents, the fraction of recrystallized structure was reduced. In the behavior of hydride morphology, the effect of microstructure was dominant. Intergrain hydrides and intragrain hydrides were observed in recrystallized grain so macroscopic hydride morphology was short and thick by tangled hydrides. On the other hand, cold-woked grain had only intergrain hydrides. they are precipitated with aligning to rolling direction. So the hydrides in macroscopic scale show thin and long morphology.



Figure 3 the hydride morphology of (a) Unalloyed Zr, (b) Zr-1.0%Nb and (c) Zr-2.0%Nb with 500 ppm hydrogen charged

4.2 Effect of Nb on tensile properties of hydrided Zr alloys

In Zr-2.0%Nb, yield stress was decreased slightly when hydrogen was charged. B-Zr precipitates by addition of Nb enhanced TSS of hydrogen. This cause the dissolved hydrogen concentration near the b-Zr precipitates is very high at room temperature. Therefore, the softening of mechanical properties due to high hydrogen concentration reduces the yield stress. Also elongation was decreased significantly with increasing of hydrogen contents because of high volume fraction of hydride precipitates. The addition of Nb enhanced the resistance to hydride embrittlement. The reason is that the softening of Zr alloys is accrued by hydrogen solid solution because b-Zr precipitates by addition of Nb enhanced TSS of hydrogen. Consequently, it is thought that b-Zr precipitates inhibit the hydride embrittlement.



Figure 4 YS of zirconium alloys with $C_{\rm H}$



Figure 5 Elongation of zirconium alloys with C_H 4.3 fractography

After tensile test, fracture surface morphology was observed. The fracture morphology is affected by microstructure. In recrystalized grain, facet fracture surface was observed. On the other hand, cold-worked grain caused dimple fracture surface. The secondary cracks were observed in hydride Zr alloy. The behavior of secondary cracks was similar to hydride morphology so it was thought that the secondary cracks were accrued by hydride and the secondary crack reduced the ductility.



Figure 6 fracture surface of (a) unalloyed Zr, (b) Zr-1.0%Nb and (c) Zr-2.0%Nb which are charged

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