Effects of hydride precipitation on the mechanical property degradation of Zircaloy-4 and Zirlo

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1. Introduction

Mechanical integrity of zirconium alloy cladding is important to achieve the performance goal not only in high burn-up and extended fuel cycle operation and but also in long- term dry storage of spent fuel. Especially, it is well-known that hydride precipitation critically promotes the degradation of the mechanical integrity. Thus, many studies on the hydride-induced degradation and embrittlement of zirconium alloys have been reported [1, 2]. However, few studies on the quantitative and mechanistic analysis for the degradation have been carried out.

In this study, mechanical properties degradation of two popular zirconium alloys, Zircaloy-4 and Zirlo, with increasing hydrogen concentration was experimentally investigated and the degradation mechanism was discussed.

2. Experimental

2.1 Sample Preparation

Uni-axial tensile test was selected for current study not only because the test is simple and quite reproducible but also because these out-of-pile data will be compared with in-pile data generated later in the hotcell examination.

First, cold-rolled zirconium alloy sheet in fully recrystallized condition in accordance with ASTM specification B352 was fabricated in KNFC (Korea Nuclear Fuel Company). Then, according to ASTM E8/E21 tensile specimen (gauge section of 25.4 x 3.7 x 0.5 mm) was made with tensile axis in the transverse direction (Figure 1), which is one of the standard specimens for Hanaro test reactor. Hydrogen charging was performed at 500° C with advanced Sieverts' law apparatus.



Figure 1. Tensile specimen geometry

2.2 Uniaxial Tensile Test

Tensile test were conducted with Instron universal testing machine (model 5582) at room temperature and 400°C. Following ASTM E8/E21, strain rate was set to be 1 x 10-3 s-1 at both temperatures. Elongation was measured with strain gauge on the basis of assumption that strain of resistant and its resistance are proportional. Young's Modulus was derived from the measured strain from the strain gauge and stress from Instron machine.

2. Results

As expected, Figure 3 shows that Young's modulus decreases with increasing hydrogen concentrations.

It has been generally observed that Young's modulus drops in most materials when crack and porosity develops inside [3]. In this case of hydrided zirconium hydrogen precipitates immediately when alloys, concentration exceeds the solubility because of very low solubility limit, especially at room temperature. Once zirconium hydride forms, hydrided zirconium metal loses its ductility and becomes very brittle. In this case, micro-cracks develop easily in the boundary between the base metal and precipitated hydrides when load is applied. These micro-cracks can propagate rapidly to zirconium matrix, ending up with the fracture even at low stress [4]. Also, relatively small size hydride makes void at hydride - zirconium interface due to the loss of adherence between the hydride and zirconium metal, increasing porosity in zirconium matrix. This is the way that mechanical integrity of the hydrided zirconium metals degrades with increasing hydrogen concentrations [5].

In addition, as shown in Figure 4, the transition between elastic region and plastic region become dissipated with hydrogen concentration increase. This means that elastic region gets narrower with hydride increment while plastic region wider with micro-crack development and void growth [5], causing the loss of ductility, i.e. elongatio.

Fractographic analysis using SEM (Scanning Electron Micrography) demonstrates that lots of small cracks were developed and micro-voids were grown due to the hydride precipitation.



Figure 2. Stress-strain curve



Figure 3. Young's modulus vs hydrogen contents



Figure 4. Log-Log rescale in plastic region of stress-strain curve

3. Conclusions

In present study, mechanical property degradation of the hydrided zirconium alloys was experimentally investigated.

As hydrogen concentration increases, as expected, Young's modulus and total elongation decrease. Primary cause of the degradation is due to micro-crack development and void growth induced by hydrogen. It is because that in the hydrided zirconium matrix, microcracks develop easily in the boundary between the base metal and precipitated hydrides when load is applied. Also, relatively small size hydride makes void at hydride - zirconium interface due to the loss of adherence between the hydride and zirconium metal.

For reliable correlation modeling of the mechanical property degradation of zirconium alloys with hydrogen concentrations, more experimental examinations will be following with various conditions in the next step.

REFERENCES

[1] S. Oh, Effect of Nb on hydride embrittlement of Zr–xNb alloys, Materials Science and Engineering A 527 (2010) 1306–1313.

[2] F.PRAT, Behavior and Rupture of Hydrided ZIRCALOY-4 Tubes and Sheets, Metall. Trans. A, 29A, JUNE (1998) 1643.

[3] X. Feng, A simple method for calculating interaction of numerous microcracks and its applications, International Journal of Solids and Structures, 40 (2003) 447–464.

[4] M. GRANGE, Anisotropic Behavior and Rupture of Hydrided Zircaloy-4 Sheets, Metall. Trans. A, 31A, MARCH (2000) 679.

[5] M. Song, Multi-scale model for the ductility of multiple phase materials, Mechanics of Materials 41 (2009) 622–633.



Figure 3. a Void(at center) and micro-cracks SEM fractography (600 ppm, Zircaloy-4)