Effect of Hydrides on Mechanical Properties of Zirconium Alloy with Oxidation

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1. Introduction

In mechanical aspects, degradations during wet storage are relatively minor. On the other hand, when spent fuel moves to dry storage, potential degradations will be revealed such as hydride embrittlement, creep, stress corrosion. Particularly during vacuum drying, there is high probability to occur massive hydride reorientation which makes cladding vulnerable to mechanical interaction [1]. When cladding moves to dry storage, cladding will be stored for at least several decades at the conditions of internal pressure up to 10MPa, hoop stress up to 65MPa at 400℃. By the time cladding discharged, integrity margin already extremely drops. Therefore, it is required a caution to build a qualification to secure the integrity of dry storage for several decades.

The paper has an object to obtain applicable mechanical properties for establishment of initial conditions to secure the integrity thought investigation cladding mechanical properties. To secure the integrity, it needs to clearly understand degradation mechanisms which are mainly hydride, radiation damage, and oxidation. In this paper, uniaxial tensile test were conducted for precise measurement of mechanical properties. Assessment factors for mechanical test are yielding, reorientation which is formed during vacuum drying. Moreover, influences of crack induced by oxidation to reorientation were additionally considered.

2. Experimental

2.1. Sample preparation

The Zircaloy-4 sheet in accordance with ASTM specification B352 was fabricated at KNFC(Korea Nuclear Fuel Company). Tensile specimens with gauge section of $25.4 \times 3.7 \times 0.5$ mm were made (Fig.1). The specimen was standardized for HANARO loading which has a limit charging space. Therefore, the specimen was considered the balance of test reliability and compact size with the reference of ASTM E8/E21.

Figure 1. Tensile specimen geometry

In the paper, to produce hydride zirconium alloy, the Advanced Sieverts Law Apparatus using gaseous diffusion method was manufactured. To prevent phase change of hydride for predictable hydrogen content of specimens, hydriding was conducted on 550℃.

To investigate the influences of pre-transition and post-transition oxide layer to hydride, oxidation was conducted to have weight gains as $2{\sim}10\mu$ m using an autoclave at 700℃ [2].

2.2. Tensile test

 Uniaxial tensile test were chosen to perform mechanical test. Because uniaxial tensile test is simple and reproducible, it is easy to perform in hot-cell. In addition, uniaxial tensile test is defined well theoretically, so yield point in the complex stress condition such as cladding which is in the between plans-strain stress and equal-biaxial stress can be predicted by using Tresca Criterion and Von Mise's Criterion [3]. The reason of conservative assessment, yield stress, which is most important factor of Tresca Criterion, was evaluated intensively.

Tensile test were conducted with INSTRON 5582 Universal Testing Machine at room temperature and 400℃. For uniaxial tensile test, in the case of high temperature, ASTM E21 recommends $0.5-1 \times 10^{-3} \text{ s}^{-1}$ strain rate. In the test, 1×10^{-3} s⁻¹ strain rate was taken to both room temperature test and high temperature test. Young's modulus was measured by strain gauge, because the measurement of modulus is extremely difficult on conversational mechanical test.

3. Results and Discussion

3.1. Hydride effect

The result of tensile test, 0.2% offset yield stress hardly changes until 300ppm, but it decreases at 600 ppm. Total elongation decreases with hydrogen contents, to figure out ductile or brittle of 600 ppm specimen, SEM Fractograph were observed. Fig. 2 shows that the intact specimen has ductile dimples, on the other hand, both 300 and 600 ppm specimen have brittle cleavages and a lot of small cracks. As look at the middle of Fig 2, primary void induced hydride is observed. The reason of 0.2% offset yield stress and young's modulus declining with hydrogen contents is hydride formation and micro-cracking existing. In these hydride specimens, precipitation hardening is hardly expected because micro-cracks are formed all over the specimen. In

addition, as look at stress-strain curve, the elastic region of 600 ppm specimen is shorter than intact specimen and strain hardening starts early during tensile test. Thereby, yielding occurs at low stress.

3.1. Oxide effect

The result of tensile test, UTS increases quietly, elongation decreases at 10um specimen, but 0.2% offset yield stress is lower than intact specimens because it has shorter elastic region likewise hydride specimens. As look at 2μm oxide layer specimens, it could regard that 2μm specimens does not have especial mechanical changes comparing with intact specimens. However, at Fig.3 0.2% offset yield stress of 2μm specimens with hydrided have lower value than non-oxidation hydride specimens and 10μm oxide layer specimens.

4. Conclusions

In present study, the effect of hydride on mechanical properties of Zircaloy-4 sheet specimen was evaluated.

As the hydrogen increases, most mechanical properties such as UTS, yield stress, total elongation, young's modulus decrease overall. Primary cause is micro-cracking induced by hydride. At the specimens with oxidation, 2μm oxide layer can be regarded as minor, but 10μm oxide layer specimens have lower total elongation. Temperature effect is distinguish, strength decrease abruptly, and elongation increase slightly.

It infers that strength of cladding can be degraded during dry storage. The results show that hydride can make micro-crack that is regarded as dominant factor more than hardening mechanism. It affects the declining both strength and elongation.

Figure 2. OM(left), SEM(right) image (from top –intact, 300 ppm)

Figure 3. 0.2% offset yield stress versus hydrogen contents

Figure 4. Total elongation versus hydrogen contents

Figure 5 Young's Modulus versus hydrogen contents

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