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A Study on the Radial Hydride Assisted Delayed Hydride Cracking of Zircaloy

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1. Introduction

Delayed hydride cracking (DHC) is a time-dependent crack growth mechanism occurring in Zircaloy that requires the repetition of precipitation, growth, and fracture of brittle hydride at crack tip. The basic process is illustrated in Fig. 1. Extensive studies have been done on understanding of DHC phenomenon since several zirconium alloy pressure tubes failed in nuclear reactor in the 1970s.

Recently, long-term dry storage strategy has been considered seriously in order to manage spent nuclear fuel in Korea and other countries around the world. Consequentially, many researches[1] have been investigated the degradation mechanisms which will threaten the spent fuel integrity during dry storage and showed that hydrogen related phenomenon such as hydride reorientation and DHC are the critical factors. Especially, DHC is the direct cracking mechanism which can cause not only a through-wall defect but also a radiation leak to the environment. In addition, DHC can be enhanced by radial hydride as reported by Kim[2] who demonstrate that radial hydrides clearly act as crack linkage path. This phenomenon is known as the radial hydride assisted DHC (RHA-DHC). Therefore, study on DHC is essential to ensure the safety of spent fuel.

Recently experimental works on DHC of LWR cladding were conducted and could be divided into two areas. The one was to measure the DHC velocity in axial direction using pin-loading test (PLT) specimen, which was suggested by Grigoriev[3]. Later IAEA led the coordinated research project that measured axial crack propagation rate in Zircaloy-4 cladding using the PLT technique[4]. The other was reported by Kubo[5] who measured the radial DHC velocity using chemically notched ring specimen by in-situ method.

It should be noted the fact that hydride reorientation phenomenon should be involved definitely in DHC process regardless of its propagation direction. Since crystallographic orientation of zirconium is controlled by cold pilgering during the cladding manufacturing process so that hydride platelets precipitate circumferentially around the cladding. In this preliminary study, several tests were conducted to investigate the conditions for precipitation of radial hydride at stressed region around notch tip.

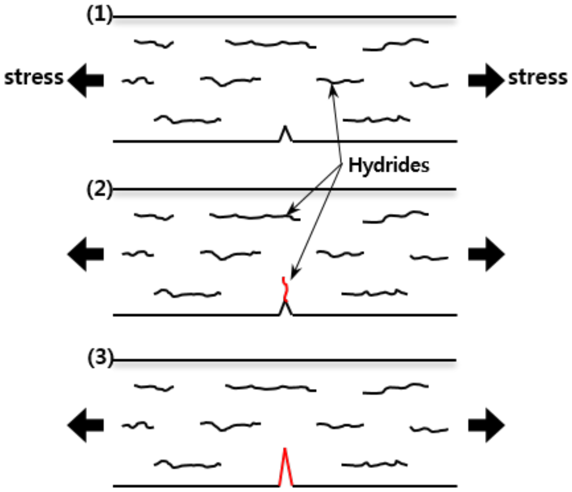

Fig. 1. A schematic illustration of DHC process.

2. Experimental

2.1 Specimen preparation

Firstly, specimens were cut into 9 mm long segment from the commercial Zircaloy-4 cladding tube with an outside diameter of 9.5 mm and wall thickness of 0.57 mm. Secondly, the specimens were charged with hydrogen in Sieverts-type apparatus. We followed the detailed hydrogenation method presented in Ref. [2]. In our preliminary test, hydrogen concentration in all specimens was controlled to have a value of 100 wppm approximately. Finally, the radially notched specimens were made by electric discharge machining whose detailed geometry is shown in Fig. 2.

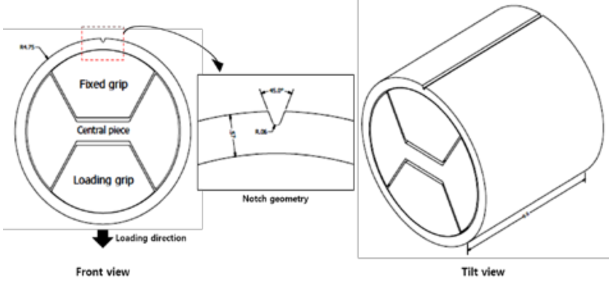

Fig. 2. Detailed geometry of notched specimen.

2.2 Test procedure

Constant load apparatus (Fig. 3) was used to produce a radially reoriented hydride at notch tip. Three jig pieces were used and set up on the apparatus as shown in Fig. 2. We followed similar experimental procedure for radial hydride treatment as presented in Ref. [6] which have explained the detailed method to make radial hydrides. According to Kim[6], it is imperative to heat the specimens up to peak temperature in order to dissolve hydrogen into the Zirconium (Zr) matrix. Test conditions for each specimen in this preliminary test are presented in Table 1. Table 1 showed that the pre-heating temperatures are 60 °C higher than the loaded temperature which corresponds to the temperature gap between terminal solid solubility for dissolution (TSSD) and terminal solid solubility for precipitation (TSSP) of hydrogen in Zircaloy-4. Fig. 4(a) and Fig. 4(b) indicate a thermal history and expected hydrogen solid solution trajectory, respectively.

Table 1. Experimental conditions for each specimen.

No.	Peak temp.^{A} (°C)	Loaded temp. range$^{B\text{-}C}$ (°C)	Load (kg)
CR1	460	400-340	10
CR2	400	340-280	10
CR3	400	340-280	20
CR4	360	300-240	20

Superscript A or B-C indicates the point A, B and C in Fig. 4.

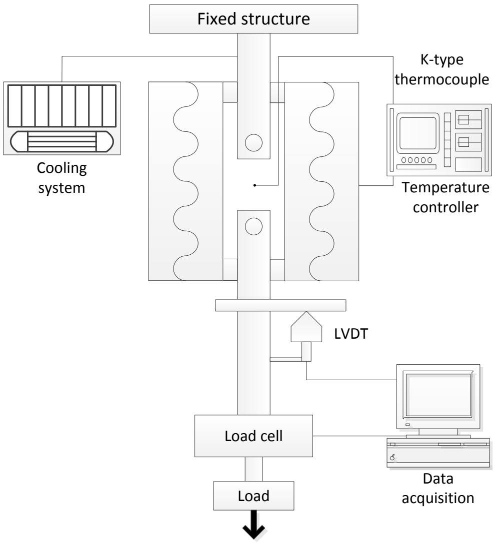

Fig. 3. A schematic diagram of constant load apparatus.

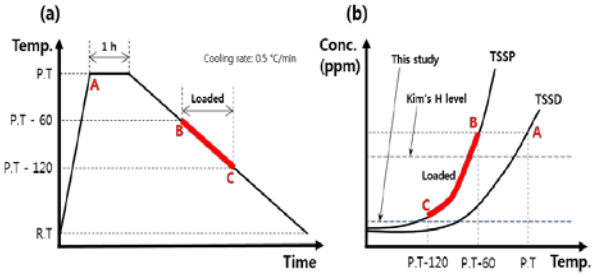

Fig. 4. Experimental conditions: (a) Thermal history, (b) expected hydrogen solid solution trajectory.

3. Results & Discussion

Fig. 5 shows the hydride morphologies around notch tip of each specimen before and after the preliminary test. Circumferential hydrides were precipitated uniformly before test in all cases. After tests according to different experimental conditions given in Table 1, there were no changes at all in the hydride morphology even though excessive hoop stress was applied. In the case of specimen CR3 after the test, although hydride was observed right in front of notch tip, it was not radial hydride as expected.

Kim[6] using un-notched ring specimen observed that the dissolved hydrogen precipitates radially, only when temperature decreases along TSSP curve under considerable stress. In Kim's case, there was enough dissolved hydrogen exceeding the TSSP to precipitate as shown in Fig. 4(b). On the other hands, in this preliminary test, although enough stress was applied during cool-down, hydrogen could not precipitate because hydrogen contained in specimen was too low so that all hydrogen has been dissolved in supersaturated state. Later when the load was removed, dissolved hydrogen might precipitate naturally in circumferential direction during cool-down to room temperature. Then it can be explained why no radial hydride was observed at all. The relation between hydrogen concentration and hydride reorientation phenomenon will be studied further.

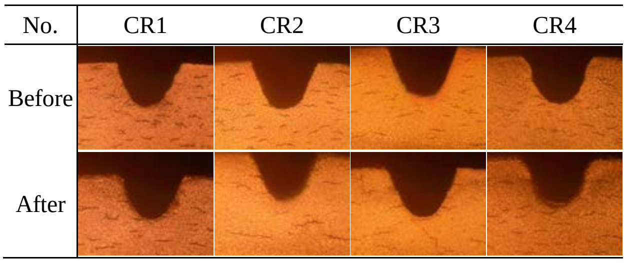

Fig. 5. Hydride morphologies around notch tip of each specimen before and after the preliminary test.

4. Future plans

It is insufficient to suggest any conclusions from these preliminary test results. Following are future plans for study on radial HRA-DHC.

- We will increase hydrogen concentration contained in specimen to simulate high burn-up fuel cladding.
- Finite element analysis will be carried out for the stress gradient evaluation around notch tip.
- A variation in thermal cycle which leads to change in hydrogen solid solution trajectory may be required.
- If the radial hydride precipitates at notch tip, we will investigate what conditions should be met. Ultimately, we will suggest the regulation criteria for long-term dry storage of spent nuclear fuel.

- We will measure the radial HRA-DHC velocity caused by fracture of reoriented hydride at notch tip and compare three of the zirconium alloys, Zircaloy-4, ZIRLO, and HANA which are being used in domestic nuclear industry.

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