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Delayed Hydride Cracking Velocity of CANDU Zr-2.5Nb Tubes with Orientation

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1. Introduction

Though CSA N285.8-05 stipulates that anisotropic delayed hydride cracking velocity (DHCV) of CANDU Zr-2.5Nb tubes should be used for their mechanical integrity analysis [1], the cause of the anisotropic DHCV is yet to be understood. Another thing is that even though the Zr-2.5Nb tubes have microstructural evolution during their operation in reactors [2], its effect on anisotropic DHCV of CANDU Zr-2.5Nb tubes is disregarded.

Recently, Kim demonstrated that hydrogen diffusion mainly governs DHCV at below 300 ^{o}C, using Kim's DHC model [3]. Thus, since the decomposition of the β-Zr phase is accompanied during an operation of CADNU Zr-2.5Nb tubes in reactors, it will affect their DHCV and anisotropy. The aim of this study is to elucidate the cause of an anisotropic DHCV in CANDU pressure tubes by correlating it with hydrogen diffusion with orientation and thereby to demonstrate the plausibility of Kim's DHC model [4]. To this end, we determined axial and radial DHCVs using compact tension and cantilever beam specimens containing hydrogen which were taken from a CANDU pressure tube with an anisotropic distribution of the β-Zr. As decisive evidence for the role of an anisotropic hydrogen diffusion in the anisotropic DHCV, we cited Levi and Sagat's experiment [5] where DHC tests were conducted on annealed Zr-2.5Nb tubes and plates with a fully discontinuous β-Zr.

2. Experimental Procedures

Compact tension (CT) specimens of 17 mm long and cantilever beam (CB) specimens of 38 mm long were taken from a CANDU Zr-2.5Nb tube with elongated α-Zr grains in the axial direction. It should be noted that the CANDU Zr-2.5Nb tube has the β-Zr phase semi-continuously distributed between the elongated α-Zr grains in the longitudinal direction of the tube [2]. The compact tension and cantilever beam specimens were used for determining the DHC velocities and the threshold stress intensity factors, K_{IH}'s for the onset of delayed hydride cracking, respectively, in the axial and radial directions of the CANDU Zr-2.5Nb tube. All the specimens were subjected to electrolytic charging to form a thick hydride layer on their surface followed by homogenization treatment at different temperatures to dissolve hydrogen concentration ranging from 27 to 100 ppm H. At the end of homogenization treatment, they were water-quenched or furnace-cooled to change the size and phase of the hydride precipitates. The details of the hydrogen charging procedures have been reported elsewhere [11].

DHC tests were conducted using a creep machine to apply a constant load to the 17 mm CT specimens or using a cantilever beam tester to apply constant stress intensity factors to the CB specimens with a 0.5 mm deep notch and the notch root radius of 0.05 mm. The initiation and growth of a crack was monitored by a direct current potential drop method for the CT specimens and by an acoustic emission method for the CB specimens. The stress intensity factor ranging from 6.13 to 18.4 MPa√m was applied during the DHC tests on the CB specimens through an auto-control program. Most of the specimens were subjected to a thermal cycle during the DHC tests where the test temperatures were approached from an upward direction by a cooling as shown in Fig. 3. The water-quenched specimens after a homogenization treatment of hydrogen were not subjected to a thermal cycle but they approached the test temperature directly by a heating. The threshold stress intensity factor, K_{IH} was determined by the load decreasing mode [6] where the applied load decreased from 20 MPa√m step-wise by 0.5 MPa √m until the crack growth stopped. DHCV was determined from post-fracture measurements of the DHC crack length by an image analyzer method divided by the cracking time.

3. Results and Discussion

Fig. 1 shows the measured DHCV in the axial and radial directions of the CANDU Zr-2.5Nb tube at temperatures ranging from 100 to 300 ^{o}C. As expected, the axial DHCV was around 1.8 to 2 times higher than the radial DHCV and the difference between the radial DHCV and the axial one remained constant at a temperature range of 160 to 250 ^{o}C. This constant difference in the DHCV with the orientation agrees with Sagat's anisotropic DHCV data for an unirradiated CANDU Zr-2.5Nb tube but it differs from that for irradiated CANDU Zr-2.5Nb tubes [2]. Fig. 2 shows the threshold stress intensity factors, K_{IH}'s for the onset of delayed hydride cracking that were determined for the axial and radial directions of the CANDU Zr-2.5Nb tube at temperatures ranging from 160 to 280 ^{o}C. The radial K_{IH} was higher than the axial K_{IH} and the ratio of the radial and axial K_{IH} was kept constant in this temperature interval.

According to Kim's DHC model [4], DHCV in zirconium alloys is governed by hydrogen diffusion and the hydrogen concentration gradient at a flaw tip defined by the supersaturated hydrogen concentration, the yield strength and the threshold stress intensity factor:

$$V = (k_1 D_H)\Delta C\left[k_2 \frac{\sigma_{YS}}{K_{IH}}\right]^2 \qquad (1)$$

where V is the DHC velocity, D_H is a diffusivity of hydrogen, ΔC a supersaturated hydrogen concentration, σ_{YS} the yield strength of the CANDU tube used, k_1 and k_2 constants and K_{IH} the threshold stress intensity factor for the onset of DHC in the CANDU Zr-2.5Nb tube. Thus, since the supersaturated hydrogen concentration must be the same irrespective of the orientation, the ratio of the axial and radial DHCV can be determined as shown below:

$$\frac{V_{ax}}{V_r} = \frac{D_{H,ax}}{D_{H,r}}\left(\frac{\sigma_{YS,ax}}{\sigma_{YS,r}}\right)^2\left(\frac{K_{IH,r}}{K_{IH,ax}}\right)^2 \qquad (2)$$

where V_{ax} and V_r are the DHC velocity in the axial and radial directions of the CANDU Zr-2.5Nb tube, $\sigma_{YS,ax}$ and $\sigma_{YS,r}$ are its axial and radial yield strengths, and $K_{IH,ax}$ and $K_{IH,r}$ are the axial and radial threshold stress intensity factors. Since it was demonstrated that the yield strengths of the CANDU Zr-2.5Nb tube are almost the same in both directions [7], Eq. (2) shows that the anisotropic DHCV depends only upon

hydrogen diffusion and K_{IH} with the orientation. A problem arises in which of the D_H and K_{IH} shown in Eq. (2) controls DHCV. From the results shown in Fig. 3, the ratio of the axial and radial K_{IH} turned out to be 1.43. By placing this value into Eq. (2), we could suggest that the around two times higher V_{ax} than V_r is due to the higher radial K_{IH} than the axial K_{IH} as shown in Fig. 2 assuming the same hydrogen diffusivity with the orientation of the CANDU pressure tube. However, this hypothesis disagrees with Skinner's experiment [8] and Kim's demonstration [3]. Hence, a more reasonable rationale is that the DHC crack growth rate is governed primarily by a hydrogen diffusivity as experimentally demonstrated by Kim [3] and the effect of the K_{IH} on DHCV that governs the hydrogen concentration gradient at the flaw tip, is relatively minor at below 300 °C compared to diffusion of hydrogen.

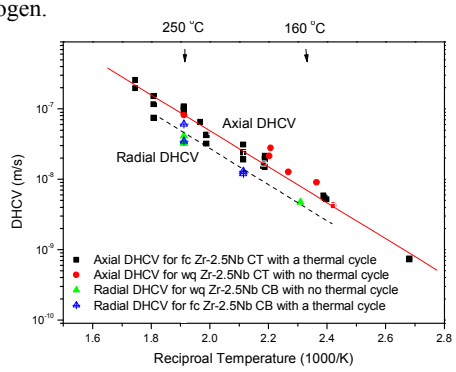

Fig. 1. DHCV of a CANDU Zr-2.5Nb tube with orientation.

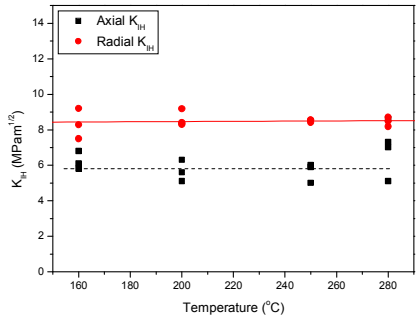

Fig. 2. Threshold stress intensity factor, K_{IH} for the onset of DHC for a CANDU Zr-2.5Nb tube with the orientation that were determined by the load-decreasing mode.

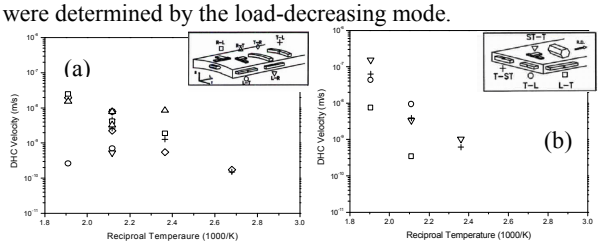

Fig. 3. DHCV with the orientation in (a) a Zr-2.5Nb tube annealed at 400 °C for 1000 h and (b) a Zr-2.5Nb plate annealed at 650 °C for 9h [10].

Another decisive evidence for this rationale can be provided by demonstrating that the anisotropic DHCV of the CANDU Zr-2.5Nb tube should disappear by causing the β-Zr phase to become uniformly distributed with its orientation. It is because a hydrogen diffusivity or D_H is influenced mainly by the distribution of the β-Zr where hydrogen diffuses much faster when compared to the α-Zr [8]. Fortunately, Levi and Sagat [5] conducted this kind of experiment where the axial and radial DHCV were measured from both a CANDU Zr-2.5Nb tube annealed at 400 °C for 1000 h and a Zr-2.5Nb plate annealed at 650 °C for 9 h. Fig. 3 shows their results where the T-R or T-ST specimens with the crack growing in the radial direction of the tube or the plate, respectively, had the similar growth rate to that of the T-L specimens with the crack growing in the longitudinal direction. As expected, no anisotropic DHCV with the orientation was observed for the annealed Zr-2.5Nb tube and plate where a non-uniform distribution of the β-Zr should be disrupted by annealing to become uniform. In other words, a difference in DHCV with the orientation disappeared at a temperature range of 100 to 250 °C.

4. Conclusion

DHC experiments demonstrated that anisotropic DHCV of CANDU Zr-2.5Nb tubes is caused by anisotropic diffusion of hydrogen through inhomogeneous distribution of the β-Zr, leading the axial DHCV of the CANDU Zr-2.5Nb tube to be higher than that in the axial direction, and not affected by the K_{IH} with the tube orientation.

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