Delayed Hydride Cracking in Zr-2.5Nb Tubes with the Direction of An Approach to Temperature

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1. Introduction

One of the unique features of delayed hydride cracking (DHC) of zirconium alloys is that the DHC velocity (DHCV) of zirconium alloys strongly depends on the path to the test temperature. Ambler reported that the DHCV of Zr-2.5Nb tubes at temperatures above 180 ^{o}C depended upon the direction of an approach to the test temperatures, and reported on a presence of the DHC arrest temperature or T_{DAT} above which the DHCV decreased upon an approach to the test temperature by a heating[1]. Ambler proposed a hydrogen transfer from the bulk to the crack tip assuming that the hydrides formed at the crack tip and in the bulk region are fully constrained and partially constrained at the crack tip, respectively. In other words, the terminal solid solubility (TSS) of hydrogen would be governed by elastic strain energy induced by the precipitating hydrides, leading to a higher TSS in the bulk region than that at the crack tip. In a sense, his assumption that the hydrogen concentration is higher in the bulk region than that at the crack tip due to a higher TSS in the bulk region is, in a way, similar to Kim's DHC model [2,3]. Even though Ambler assumed a different strain energy of the matrix hydrides with the direction of an approach to the test temperature, the peak temperature, hydrogen concentration and the hydride phase, a feasible rationale for this assumption is yet to be given.

In this study, a path dependence of DHC velocity of Zr-2.5Nb tubes will be investigated using Kim's DHC model where a driving force for DHC is the supersaturated hydrogen concentration between the crack tip and the bulk region. To this ends, the furnace-cooled and water-quenched Zr-2.5Nb specimens were subjected to DHC tests at different test temperatures that were approached by a heating or by a cooling. Kim's DHC model predicts that the water-quenched Zr-2.5Nb will have DHC crack growth even at temperatures above 180 ^{o}C where the furnace-cooled Zr-2.5Nb will not. This experiment will provide supporting evidence that the TSSD of γ-hydrides is higher than that of δ-hydrides and furthermore it will confirm the plausibility of Kim's DHC model.

2. Experimental Procedures

17 mm compact tension (CT) specimens and 38 mm long cantilever beam (CB) specimens were taken from a cold worked and stress relieved Zr-2.5Nb pressure tube to determined the DHCV in the axial and radial directions, respectively. Hydrogen was charged electrolytically into the Zr-2.5Nb tube to form a thick hydride layer on the surface followed by annealing in a salt bath where the specimen temperature could be controlled to within ±0.5 ^{o}C. The water-quenched specimens were subjected to a homogenization treatment at 370 ^{o}C for 11 h to contain 89 ppm H and then to water quenching. The furnace-cooled specimens were subjected to homogenization treatments at various temperatures from 200 to 360 ^{o}C to contain 10 to 100 ppm H and then to furnace-cooling.

CT and CB specimens were subjected to a dead load in a creep machine and in a cantilever beam tester[1,2], respectively, while the initiation and growth of a crack were monitored by a dc potential drop method or an acoustic emission method [4]. Most of the furnace-cooled specimens experienced a thermal cycle with an approach to the test temperatures by a cooling from the peak temperatures where all the charged hydrogen was able to be completely dissolved in zirconium. All the water-quenched specimens and a few furnace-cooled ones had no thermal cycle with an approach to the test temperature by a heating. The load corresponding to 15 MPa√m was applied 30 minutes after the test temperature was reached. The crack length was determined on the fractured surfaces by dividing the area of the DHC crack calculated by an image analyzer by the width.

3. Results and Discussion

Fig. 1 shows the DHCV of the furnace-cooled and water-quenched specimens in the radial and axial directions of the Zr-2.5Nb tube. Here, a thermal cycle represents an approach to the test temperature from above or by a cooling while no thermal cycle means an approach to the test temperature from below by a heating. The furnace-cooled Zr-2.5Nb CT specimens subjected to a thermal cycle had DHC crack growth, whose rate increases with an increasing temperature. In contrast, the furnace-cooled Zr-2.5Nb CT specimens with no thermal cycle were not able to initiate a DHC crack at 250 ^{o}C. As expected, however, the water-quenched Zr-2.5Nb CT specimens even with no thermal cycle had DHC crack growth even at 250 ^{o}C, whose rate was found to be almost similar to that of the

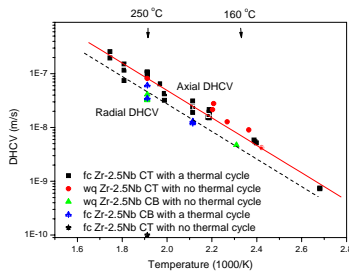furnace-cooled Zr-2.5Nb subjected to a thermal cycle. When

Fig. 1 Axial and radial delayed hydride crack velocity (DHCV) of the water-quenched and furnace-cooled Zr-2.5Nb specimens with and without a thermal cycle that were determined by using compact tension (CT) and cantilever beam (CB) specimens, respectively.

the Zr-2.5Nb CB specimens with the crack growing in the radial direction were subjected to no thermal cycle, the same path dependency of DHC crack growth was also observed: the water-quenched CB specimens had DHC crack growth even at 250 ^{o}C where the furnace-cooled CB specimens had no DHC crack growth. Furthermore, the DHCV of the water-quenched CB specimens with no thermal cycle was similar to that of the furnace-cooled CB specimens with a thermal cycle.

Considering Shek's result[5] that no DHC occurs at temperatures in excess of 200 ^{o}C for Zr-2.5Nb specimens without a cooling of ΔT, it is very surprising that the water-quenched Zr-2.5Nb specimens had DHC crack growth even without a thermal cycle, as shown in **Fig. 1**. Our results that the water-quenched Zr-2.5Nb specimens with no thermal cycle had the DHC crack growth in the radial and axial directions even at 250 ^{o}C provide supporting evidence to the role of the stress-induced transformation from the γ-hydrides to the δ-hydrides, as suggested by Kim [2]. His hypothesis is that it is related to the formation of the γ-hydride in addition to the δ-hydride that DHC has occurred in both Zr-2.5Nb specimens at temperatures below 180 ^{o}C upon an approach to the test temperature by a heating. At the test temperature, the stress-induced γ-δ transformation will occur only at the crack tip subjected to a tensile stress, causing a difference in the hydrogen concentration or ΔC between the crack tip only with the δ-hydrides and the bulk region with a mixture of the δ- and γ-hydrides. This occurs if the γ-hydrides have a higher terminal solid solubility (TSS) than the δ-hydrides.

For experimental evidence on the stress-induced γ-δ transformation at the crack tip, X-ray diffraction analyses were carried out on the fractured surface and the bulk region at a distance of 20 mm away from the fractured surface in a water-quenched Zr-2.5Nb specimen. As expected, only the δ-hydrides appeared on the fractured surface as shown in Fig. 2 while most hydrides present in the bulk region were found to be γ-hydrides. The results shown in Fig. 2 definitively demonstrate that the stress-induced hydride transformation from γ to δ occurs at the crack tip as suggested by Kim's DHC model, which is a driving force for DHC.
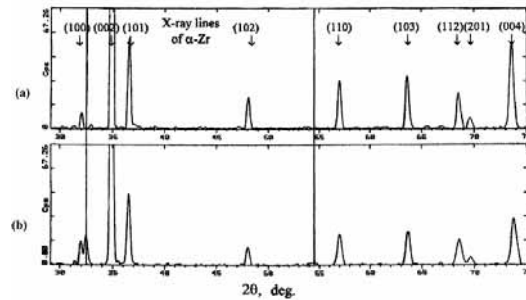

Fig. 2 X-ray diffraction analyses of (a) the bulk region at a distance of 20 mm from the crack tip and of (b) the crack tip in the water-quenched Zr-2.5Nb specimens subjected to a DHC test at 250 ^{o}C.

4. Conclusion

The DHCV of the water-quenched Zr-2.5Nb tubes was independent of the direction of the approach even at temperatures as high as 250 ^{o}C where the DHC of the furnace-cooled ones occurred only upon an approach by a cooling. This different DHC behavior of the Zr-2.5Nb tubes with the cooling rate after a homogenization treatment is due to precipitation of the γ-hydrides only in the water-quenched Zr-2.5Nb tubes. Likewise, the increased DHC arrest temperatures or T_{DAT} for the water-quenched Zr-2.5Nb and the hydrogen concentration dependency of the T_{DAT} was suggested to be related to the volume fraction of the γ-hydrides. Based on these results, we conclude that the stress-induced γ-δ hydride transformation at the crack tip, as Kim's DHC model has already suggested, is the cause for DHC in the water-quenched Zr-2.5Nb tubes and no DHC in the furnace-cooled ones upon an approach by a heating at a test temperature over 200 ^{o}C.

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