

Dependency of Delayed Hydride Crack Velocity on the Direction of an Approach to Test Temperatures in Zirconium Alloys

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1. Introduction

Recently, Kim proposed a new DHC model where a driving force for the DHC is a supersaturated hydrogen concentration as a result of a hysteresis of the terminal solid solubility (TSS) of hydrogen in zirconium alloys upon a heating and a cooling^{1,2)}. This model was demonstrated to be valid through a model experiment where the prior plastic deformation facilitated nucleation of the reoriented hydrides, thus reducing the supersaturated hydrogen concentration at the plastic zone ahead of the crack tip and causing hydrogen to move to the crack tip from the bulk region³⁾. Thus, an approach to the test temperature by a cooling is required to create a supersaturation of hydrogen, which is a driving force for the DHC of zirconium alloys. However, despite the absence of the supersaturation of hydrogen due to an approach to the test temperature by a heating, DHC is observed to occur in zirconium alloys at the test temperatures below 180 °C³⁾. As to this DHC phenomenon, Kim proposed that stress-induced transformation from γ -hydrides to δ -hydrides is likely to be a cause of this, based on Root's observation that the γ -hydride is a stable phase at temperatures lower than 180 °C⁴⁾. In other words, the hydrides formed at the crack tip would be δ -hydrides due to the stress-induced transformation while the bulk region still maintains the initial hydride phase or γ -hydrides. It should be noted that Ambler has also assumed the crack tip hydrides to be δ -hydrides³⁾. When the δ -hydrides or $ZrH_{1.66}$ are precipitated at the crack tip due to the transformation of the γ -hydrides or ZrH , the crack tip will have a decreased concentration of dissolved hydrogen in zirconium, considering the atomic ratio of hydrogen and zirconium in the γ - and δ -hydrides³⁾. In contrast, due to no stress-induced transformation of hydrides, the bulk region maintains the initial concentration of dissolved hydrogen. Hence, there develops a difference in the hydrogen concentration or ΔC between the bulk and the crack tip, leading to the transfer of hydrogen from the bulk region to the crack tip. In short, this is the driving force for the DHC of zirconium alloys upon an approach to the test temperature by a heating. It should be noted that Kim's proposition¹⁾ that the terminal solid solubility for dissolution (TSSD) of the γ -hydride is higher than that of the δ -hydride is based on the experimental results reported by Cann⁵⁾ and Mishra⁶⁾. The objective of this study is to demonstrate if the water-quenched Zr-2.5Nb will have the DHC crack growth even at the temperatures above 180 °C where the furnace-cooled

Zr-2.5Nb will not. This experiment will provide supportive evidence that the TSSD of γ -hydrides is higher than that of δ -hydrides and furthermore, it will confirm the feasibility 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 determine the DHCV in the axial and radial directions, respectively. Hydrogen was charged electrolytically into the Zr-2.5Nb tube. The water-quenched specimens were subjected to a homogenization treatment at 370 °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 °C to contain 10 to 100 ppm H and then to furnace-cooling. The water-quenched Zr-2.5Nb had very fine hydrides precipitated while the furnace-cooled one had coarse and long hydrides..

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 the growth of a DHC crack, whose rate increased 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 °C. As expected, however, the water-quenched Zr-2.5Nb CT specimens even with no thermal cycle had the growth of a DHC crack even at 250 °C, whose rate was found to be almost similar to that of the furnace-cooled Zr-2.5Nb subjected to a thermal cycle. When the CB specimens were subjected to no thermal cycle, the same path dependency of the DHC crack growth was also observed: the water-quenched CB specimens had the growth of a DHC crack even at 250 °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.

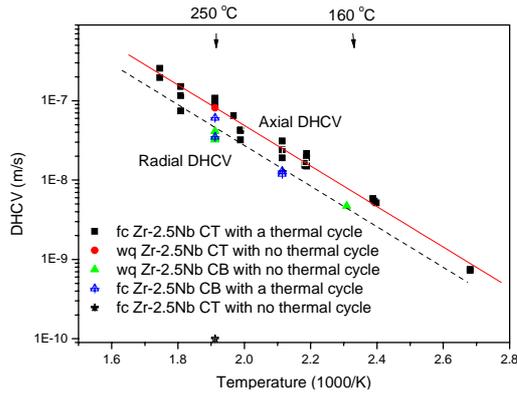


Fig. 1. Axial and radial DHCV of the water-quenched and furnace-cooled Zr-2.5Nb CB and CT with or without a thermal cycle.

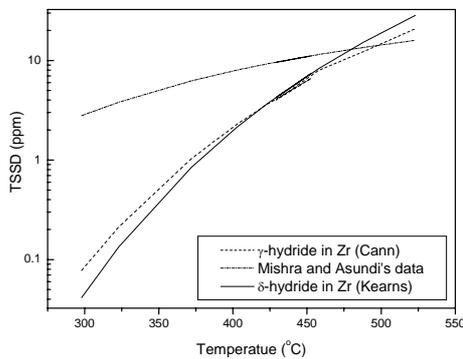


Fig. 2. Comparison of the hydrogen solubility of γ -hydride⁵⁾ and the Kearns data⁸⁾ extrapolated to lower temperatures that corresponds to the solubility of the δ -hydride along with Mishra and Asundi data⁶⁾.

The results shown in Fig. 1 show 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 °C, which provides supportive evidence to the role of stress-induced transformation from γ -hydrides to δ -hydrides, as suggested by Kim²⁾. His hypothesis is that it is related to the formation of the γ -hydride in addition to the δ -hydride that DHC occurred at temperatures lower than 180 °C upon approaching the test temperature from below. When the stable δ -hydrides are formed by the stress-induced γ - δ transformation only at the crack tip under a tensile stress, a difference in the hydrogen concentration will arise between the bulk region with mostly γ -hydrides and the crack tip with the δ -hydrides. This occurs because of a higher TSS of the γ -hydrides than that of the δ -hydrides as shown in Fig. 2.

Fine hydrides precipitated in the water-quenched specimens are mostly γ -hydrides³⁾, while the furnace-cooled ones have δ -hydrides and a few γ -hydrides^{3,7)}. Thus, due to the γ - δ transformation at temperatures above 180 °C⁷⁾, the furnace-cooled specimens will have mostly δ -hydrides with a little γ -hydrides at 250 °C. With little stress-induced γ to δ hydride transformation

at the crack tip, no driving force will develop or there will be no difference in the hydrogen concentration between the bulk region and the crack tip in the furnace-cooled specimen, leading to no DHC, as shown in Fig. 1. In contrast, the water-quenched specimens with many small γ -hydrides will still have some γ -hydrides left on a heating to 250 °C. When the tensile stress is applied, however, the stress-induced γ to δ hydride transformation occurs only at the crack tip. On the other hand, the bulk region will maintain γ -hydrides due to no stress-induced transformation. This will create a difference in the hydrogen concentration between the crack tip and the bulk region due to the assumption that the TSSD of the γ -hydride is larger than that of the δ -hydride, as shown in Fig. 2. Therefore, a finding that the water-quenched Zr-2.5Nb specimens had DHC even with an approach to the test temperature by a heating provides evidence to the stress-induced transformation of the γ - δ hydrides as well as a difference of the terminal solid solubility(TSS) for the γ - and δ -hydrides, respectively.

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 °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 the precipitation of the γ -hydrides only in the water-quenched Zr-2.5Nb tubes. 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 heating.

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REFERENCES

- [1] Y.S. Kim, S.S. Kim, S.B. Ahn et al., To have been presented at the 14th symposium on zirconium in the nuclear industry, ASTM, Stockholm, June 6-10, 2004.
- [2] Y.S. Kim, Metals and Materials Int., vol. 11, p.29, 2005.
- [3] J.F.R. Ambler, Zirconium in the Nuclear Industry: 6th International Symposium, ASTM STP 824, p653, 1984.
- [4] J.H. Root, W.M. Small, D. Khatamian et al., Acta Materialia vol. 51, p.2042, 2003.
- [5] C.D. Cann, A. Atrens, J. Nucl. Mater., vol. 88, p. 42, 1980.
- [6] S. Mishra, M.M. Asundi, Zirconium in Nuclear Applications, ASTM STP 551, American Society for Testing and Materials, p.63, 1974.
- [7] J.H. Root, R.W.L. Fong, J. Nucl. Mater. vol. 232, p.75, 1996.
- [8] J. J. Kearns, J. Nucl. Mater., vol. 22, p. 292, 1967.