Crack Growth Rate of Zirconium Alloys with Temperature

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1. Introduction

One of the unresolved issues related to delayed hydride cracking (DHC) of zirconium alloys is arrest of a DHC crack above 300°C despite the test temperatures being approached by cooling [1]. Two hypotheses were suggested according to the old DHC models [2], which argued that crack arrest occurs either when a difference in hydrogen concentration, ΔC between the bulk and a crack tip is reduced to zero [2] or when the bulk hydrogen concentration is equal to the terminal solid solubility for hydride dissolution (TSSD) [3]. Given that ΔC or the magnitude of the bulk hydrogen concentration in solution is a factor to affect nucleation of hydrides in the matrix, those hypotheses argued that crack arrest at higher temperatures is associated with nucleation of hydrides. The fact is that DHC is arrested despite sufficiently large hydrides being precipitated at a crack tip [4]. Therefore, it is clear that arrest of a DHC crack has nothing to do with nucleation of the hydrides, demonstrating that the old DHC models are defective.

In contrast with the old DHC models assuming that DHC is a hydrogen diffusion-driven process [2-3], DHC consists of three consecutive processes: nucleation, growth and cracking of hydrides. Hence, according to Kim's DHC model [5-6], the crack growth rate by DHC (CGR) is governed by the slowest process among the three consecutive processes. When the hydrogen concentration charged to zirconium alloys are large enough to exceed the critical supersaturation of hydrogen, ΔC_{max} above which the CGR becomes constant, the hydride nucleation rate becomes too fast to govern the CGR [7]. Thus, it is either hydride growth or hydride cracking that determines the CGR of zirconium alloys with sufficient hydrogen. The aim of this work is to understand which of the two factors affect the CGR of zirconium alloys, leading to DHC arrest. To this end, the CGR and striation spacing corresponding to a critical hydride length were determined for a cold-worked Zr-2.5Nb tube with sufficient hydrogen after DHC tests at different temperatures ranging from 144°C to above 300°C.

2. Experimental Methods

The DHC tests were conducted on compact tension (CT) specimens taken from a cold-worked Zr-2.5Nb tube at temperatures ranging from 144°C to above 300°C which were approached by cooling from a peak temperature [6]. The peak temperatures were set at
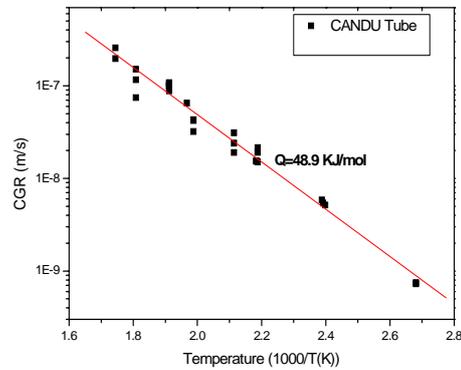

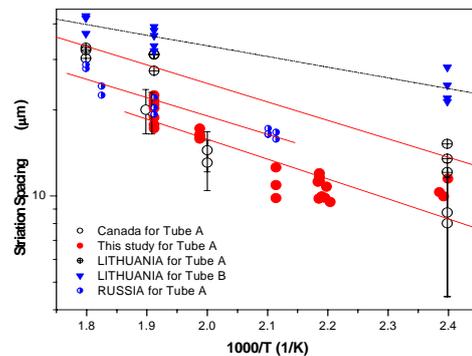Fig. 1. Temperature dependence of the crack growth rate of a cold-worked Zr-2.5Nb tube below 300°C.

Fig. 2. Temperature dependence of the striation spacing for the Zr-2.5Nb tube

10°C above the TSSD temperature to completely dissolve all the hydrides in the bulk. Two hours after reaching the test temperature, the specimens were subjected to constant loads corresponding to 15 MPa√m at the start of the DHC tests. All the detailed DHC procedures are given elsewhere [8].

3. Results and Discussion

Fig. 1 shows the CGR of the Zr-2.5Nb tube over a temperature range of 144 to 283°C. The CGR increased with increasing temperature. This kind of positive temperature dependence of the CGR clearly has also been observed not only in Zr-2.5Nb but also in Zircaloys [9] unless the test temperatures exceed a critical temperature. Given that diffusion of hydrogen increases with temperature, a positive temperature dependence of the CGR, as shown in Fig. 1, may suggest that the CGR is primarily governed by hydrogen diffusion or the hydride growth rate. In other words, it seems that the hydride growth rate becomes slower than the hydride cracking rate. To understand

the effect of hydride cracking rate on the CGR, we measured striation spacing with temperatures using the striations observed on the fracture surfaces. The striation spacing was narrow at 144°C but became wider at 283°C. Plotting the striation spacing as a function of temperatur, clearly shows that the striation spacing increased with increasing temperature. Considering that the striation spacing represents a critical hydride length over which hydrides precipitated at the crack tip can crack, it is clear that the wider striation spacing manifests higher resistance of hydride cracking. In short, the results shown in Figs. 2 demonstrate that the hydride cracking rate became slower with temperature. Despite this negative temperature dependence of the hydride cracking rate, however, the positive temperature dependence of the CGR as shown in Fig. 1 indicates that the hydride growth rate dictated by hydrogen diffusion governs the CGR, demonstrating that hydride growth is a slower process than hydride cracking.

At higher temperatures above 300°C, the CGR of the Zr-2.5Nb tubes decreased with increasing temperature [1-3] despite the hydride growth rate being much faster above 300°C. This negative temperature dependence of the CGR or crack arrest has also been seen to occur in Zircaloys [9]. Considering the fact that the hydride cracking rate becomes slower with increasing temperature, this rapid drop of the CGR above 300°C may be caused by a sharp decrease in the hydride cracking rate at higher temperatures. Thus, it is expected that the striation spacing and K_{IH} or the threshold stress intensity factor for DHC increases rapidly above 300°C. Experimental evidence is presented by Levi and Puls [4] who have demonstrated a rapid increase of the K_{IH} above 300°C. Consequently, it is clear that the hydride cracking rate became slower above 300°C as compared to the hydride growth rate, which governs the CGR. This is the cause of the negative temperature dependence of the CGR in zirconium alloys.

4. Conclusions

The CGR of the Zr-2.5Nb tube increased with temperature below 300°C but decreased rapidly above 300°C. Striation spacing delineating the critical hydride length increased gradually with temperature below 300°C but increased rapidly above 300°C. Thus, according to Kim's DHC model where the CGR is governed by a slower process between the two processes of hydride growth and hydride cracking, a positive temperature dependence of the CGR below 300°C suggests that the CGR below 300°C is governed by the hydride growth rate or hydrogen diffusion. However, the negative temperature dependence of the CGR above 300°C, despite an increase of the hydride growth rate, suggests that the governing process above 300°C is not hydrogen diffusion but hydride cracking. Concrete evidence for this suggestion was demonstrated

by the experimental facts where K_{IH} of the Zr-2.5Nb tube increased rapidly above 300°C and the activation energy for the CGR was a sum of the activation energies for hydrogen diffusion and hydride cracking. Consequently, this study demonstrates the validity of Kim's DHC model.

ACKNOWLEDGMENTS

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