

A MODEL FOR DELAYED HYDRIDE CRACKING OF ZIRCONIUM ALLOYS

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ABSTRACT

A new model for delayed hydride cracking (DHC) of zirconium alloys is proposed where its driving force is not the stress gradient but a supersaturated hydrogen concentration or ΔC resulting from a hysteresis of the terminal solid solubility of hydrogen in zirconium upon heating and cooling. For the feasibility of the newly proposed DHC model, the DHC tests were conducted on a cold-worked Zr-2.5Nb tube with a hydrogen concentration of 12 to 100 ppm where a tensile stress was applied at different times of a thermal cycle. The delayed hydride cracking velocity (DHCV) increased with an increase in the supersaturated hydrogen concentration, ΔC and then leveled off to a constant value at the ΔC in excess of the ΔC_{max} , corresponding to a difference of the terminal solid solubility of the hydrogen on cool-down and on heat-up. The applied tensile stress was found to promote precipitation of reoriented hydrides in the plastic zone ahead of the crack tip, developing the ΔC between the bulk region and the crack tip. With the reoriented hydrides precipitated all over the cross section of the specimen on cooling, however, no DHC occurred due to disappearance of the ΔC . Constant DHCV of zirconium alloys independent of an applied stress intensity factor, K is discussed with the new DHC model.

Keywords: Delayed hydride cracking velocity, zirconium, Zr-2.5Nb, terminal solid solubility, supersaturated hydrogen concentration.

1. INTRODUCTION

One unique feature of delayed hydride cracking (DHC) of zirconium alloys is a constant crack growth rate independent of applied stress intensity factors at the second stage where the applied stress intensity factor far exceeds the critical stress intensity factor, K_{IH} required to initiate DHC. (Coleman, 1977). Furthermore, this unique feature occurs not only in the DHC process of zirconium alloys but in the hydrogen embrittlement (HE) (Kerns, 1972, Kim, 2004 Hirth, 1984) or the stress corrosion cracking (SCC) of metals (Staehle, 1966, Magdowski, 1988). A driving force for all these cracking processes of metals was suggested to be a stress gradient at the crack tip (Dutton, 1977, Gangloff, 1977). This hypothesis was based on an assumption (Li, 1966) that an applied tensile stress acted as a hydrostatic tensile stress at the crack tip. However, the problem with this hypothesis is that it cannot explain the constant crack growth rate independent of the applied stress intensity factor in the cracking

processes such as DHC, HE and SCC. The aim of this work is to review the feasibility of the existing DHC models and propose a new DHC model which can contribute to a better understanding of the DHC behavior of zirconium alloys including a constant DHCV independent of the applied stress intensity factors at the 2nd stage.

2. DRIVING FORCE FOR DHC

2.1. Existing DHC models

Dutton (1977) and Puls (1982) suggested a DHC model where under a tensile stress gradient, hydrogen moved from the bulk region to the crack tip due to the effect of tensile stress on the terminal solid solubility of hydrogen arising from the difference in the molar volume of the hydrogen in the zirconium matrix and the hydride. They assumed that the hydrogen source moving to the crack tip was the hydrides in the bulk region, not in the dissolved hydrogen. Later, however, independent experiments revealed that the molar volume of hydrogen in zirconium was very similar to that in the hydride (MacEwen, 1985, Carpenter, 1973). Shi (1994, 1995) then proposed a revised DHC model where the source of hydrogen was the dissolved hydrogen, assuming the transfer of hydrogen towards the crack tip due to the stress gradient. As a result, the crack tip was assumed to have an increased hydrogen concentration by accompanying a decrease in the concentration of the dissolved hydrogen in the bulk region. When the hydrogen concentration at the crack tip increases to the terminal solid solubility for precipitation (TSSP) of hydrogen, the hydrides would instantaneously precipitate, leading to the initiation of DHC. Shi and Puls's hypothesis (1994, 1995) was proposed based on two assumptions: firstly, the applied tensile stress would act as a hydrostatic stress at the crack tip and suck in the hydrogen from the bulk region to the crack tip and, secondly, as soon as the hydrogen concentration in the crack tip would increase to the TSSP, hydrides with a larger volume would precipitate in the zirconium matrix even without cooling.

However, the first assumption seems erroneous because the experiments on the dog-bone-shaped test specimens with a linear stress gradient along their length demonstrated no transfer of hydrogen into or out of the stressed region in the absence of thermal cycling (Kammenzind, 2000). Under thermal cycling only, hydrogen was observed to migrate into high tensile regions and out of high compressive regions (Kammenzind, 2000). Though a hypothesis was suggested that the measurable transfer of hydrogen during heat-up and high temperature-thermal anneal must have occurred (Kammenzind, 2000), the precipitation of hydrides upon cooling may be its cause, which will be more clearly discussed later. This is based on the fact that the terminal solid solubility for dissolution (TSSD) of zirconium alloys is well known to be insensitive to an applied stress (Puls, 1981) and little transfer of hydrogen would occur during thermal annealing even under a stress gradient according to Kammenzind's observation (2000). Furthermore, even if it is favorably conceded that the transfer of hydrogen may occur under a higher tensile stress gradient as with the CT specimens than that of the dog-bone specimens, Shi's calculation demonstrates that the crack tip cannot reach the TSSP as long as the bulk region maintains the hydrogen concentration equivalent to the TSSD as shown in Table 1. Here, the calculated ratio of the hydrogen concentrations at the crack tip and in the bulk region (Shi, 1995) is compared to the ratio of the measured TSSP (Pan, 1996, Slattery, 1967) and TSSD (Kearns, 1967). Accounting for this weak point of the proposed DHC models, Eadie (1989, 1993) suggested that an approach to the test temperature by cooling was required for the bulk region to have an increased hydrogen concentration over the TSSD or the supersaturated hydrogen concentration for the crack tip to reach the TSSP.

Table 1. The predicted ratio of the maximum hydrogen concentration at the crack tip over the hydrogen concentration, C_o in the bulk region under an applied tensile stress gradient for a Zr-2.5Nb tube. The measured ratios of the TSSP and the TSSD, where the TSSP in the 3rd and the 4th column were taken from Pan (1996) and from Slattery (1967), respectively and the TSSD was taken from Kearns (1967) are also presented.

| Temperature (°C) | C_{max}/C_o (Shi) | TSSP*(Pan)/TSSD(Kearns) | TSSP**(Slattery)/TSSD(Kearns) |
|------------------|---------------------|-------------------------|-------------------------------|
| 50 | 2.54 | 2.77 | 3.52 |
| 200 | 1.83 | 2.14 | 2.73 |
| 250 | 1.5 | 1.74 | 2.22 |
| 300 | 1.35 | 1.46 | 1.87 |

The second assumption that hydrides would precipitate as soon as the crack tip region has the increased hydrogen concentration to the TSSP is also questionable. To induce precipitation of solid particles from a metal matrix, a supersaturation of solutes is, generally required to 5 to 1000 times the equilibrium solute concentration enough to compensate for the increased strain energy accompanied by the precipitated particles (Cahn, 1965). The best way to induce the supersaturation of hydrogen in zirconium is cooling which would contribute to compressing of the zirconium lattices hydrostatically, resulting in the supersaturation of hydrogen. Thus, without cooling, it seems impossible to precipitate any hydrides in the zirconium matrix unless the concentration of hydrogen increases to several times the equilibrium hydrogen concentration or the transformation of the precipitated phase is accompanied (Cahn, 1965). Experimental evidence to the role of cooling can be found from the results of Fig. 1 (Shi, 1996) where the TSSP temperatures of the Zr-2.5Nb tube generally coincide with the DHC initiation temperatures. In other words, the results of Fig. 1 demonstrate that cooling corresponding to the TSSP-TSSD temperatures is required to initiate precipitation of the hydrides and finally DHC in Zr-2.5Nb tubes. It should be noted that at as low a hydrogen as 7 ppm H, the DHC initiation occurs at the TSSD temperature, requiring no cooling, which is also in conflict with the previously proposed DHC models where the hydrogen concentration must increase to TSSP. Consequently, the proposed DHC model hypothesizing that the stress gradient is a driving force for DHC looks unfeasible. Therefore, a new DHC model is needed to be established based on a driving force other than the stress gradient.

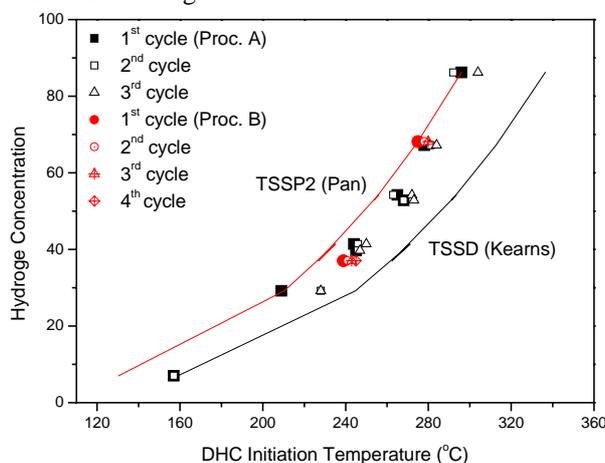


Fig. 1. DHC initiation temperatures of a Zr-2.5Nb tube containing a hydrogen concentration of 7 to 86.2 ppm with the number of a thermal cycle along with the TSSD and TSSP temperatures.

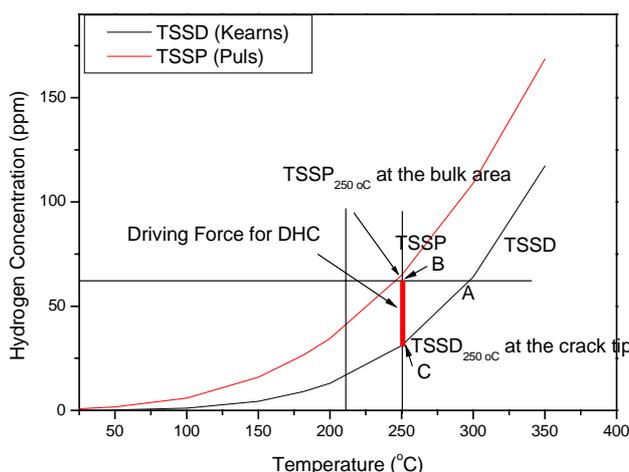


Fig. 2. Terminal solid solubility hysteresis of hydrogen during a thermal cycle to which the Zr-2.5Nb tubes are subjected during DHC tests. The driving force for DHC is the supersaturated hydrogen concentration ΔC corresponding to the distance BC in Zr-2.5Nb with 60 ppm H.

2.2. New DHC model

It should be noted that under a stress gradient, the transfer of hydrogen occurs only under thermal cycling, not under thermal annealing at a fixed temperature without any prior cooling (Kammenzind, 2000). This experimental fact hints some effect of cooling on the hydrogen transfer, which is in conflict with Kammenzind's argument (2000).

Zirconium alloys are well known to have hysteresis of the TSS of hydrogen on heat-up and cool-down due to a volume difference of around 17% between the δ -zirconium hydrides and the zirconium matrix as shown in Fig. 2 (Puls, 1984). Though the zirconium lattice is contracted on cooling with a driving force to precipitate dissolved hydrogen into hydrides, the increased lattice strain energy resulting from the precipitating hydrides with a larger volume by around 17%, thus, suppresses precipitation of the hydrides. This leads the zirconium matrix to have a higher dissolved hydrogen concentration upon cooling as opposed to the equilibrium hydrogen concentration upon heating. That is why the TSSP becomes larger than the TSSD as shown in Fig. 2.

When the Zr-2.5Nb compact tension (CT) specimen with a hydrogen concentration in excess of 60 ppm H is heated up to a peak temperature of 310 °C in a thermal cycle, it dissolves hydrogen whose concentration corresponds to the A point (as shown in Fig. 2) following the TSSD line. On cool-down to the test temperatures or 250 °C from the peak temperature, the Zr-2.5Nb specimen keeps all the hydrogen dissolved without precipitating any hydrides out of the zirconium matrix hydrostatically compressed by cooling. This corresponds to a case where the TSSD at the peak temperature is less than the TSSP at the test temperature. It leads the specimen to have the supersaturation of the dissolved hydrogen because the equilibrium hydrogen concentration that the Zr-2.5Nb specimen can dissolve is equal to the C point of Fig. 2, following the TSSD line. The amount of supersaturation of hydrogen, thus, corresponds to the distance BC as shown in Fig. 2. In general, the maximum supersaturated hydrogen concentration that the zirconium matrix can have in the thermal cycle will be TSSP-TSSD at any temperatures when the specimen is charged to a hydrogen concentration that is sufficient to exceed the TSSP at test temperatures. When a tensile stress is applied to Zr-2.5Nb CT specimens with a crack maintaining the supersaturated concentration of the dissolved hydrogen, the work energy given by the applied tensile stress will compensate for part of the increased strain energy by the precipitating hydrides with a larger volume. Hence, this will facilitate precipitation of the hydrides only at the crack tip. This causes the supersaturated hydrogen concentration at the crack tip to be reduced ultimately to the equilibrium hydrogen concentration or TSSD. In other words, the tensile stress applied at the crack tip will act to trigger nucleation of the hydrides only at the crack tip, thus decreasing the supersaturated hydrogen concentration to the TSSD at the test temperature or the C point as shown in Fig. 2. In contrast, the bulk region, which is subjected to small tensile stress, still maintains the supersaturation of hydrogen. This consequently leads to the development of a difference in the hydrogen concentration or ΔC between the crack tip and the bulk region corresponding to the distance BC, as shown in Fig. 3. Therefore, there arises a driving force for hydrogen to move from the bulk region to the crack tip. This difference in the hydrogen concentration or the amount of supersaturation of hydrogen (ΔC) is a driving force for the DHC. It should be noted that the maximum hydrogen concentration difference, ΔC_{\max} is the TSSP-TSSD at any test temperatures as shown in Fig. 2. On the contrary, if the charged hydrogen concentration is less than ΔC_{\max} (=TSSP-TSSD) at the test temperature, then the DHCV will be greatly affected with the hydrogen concentration. In essence, the driving force for the DHC is a hysteresis of the TSS on heating and cooling..

A model experiment was conducted to demonstrate the feasibility of the new DHC model proposed where the effect of plastic deformation on precipitation of reoriented hydrides and their effect on DHC susceptibility were investigated during a thermal cycle treatment of a Zr-2.5Nb tube. Furthermore, the effect of the supersaturated hydrogen concentration on the DHCV was also investigated using the Zr-2.5Nb compact tension specimens with the hydrogen concentration changing from 12 to 100 ppm.

3. EXPERIMENTAL PROCEDURES

17 mm compact tension specimens were used to determine the delayed hydride cracking velocity (DHCV) in the axial direction of a cold worked and stressed relieved Zr-2.5Nb pressure tube while cantilever beam specimens, 38 mm long x 3.2 mm wide with a 0.5 mm deep notch were used for a model experiment where the effect of plastic deformation on the reoriented hydrides and their DHC susceptibility were investigated. 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 ± 5 °C. The details of the hydrogen charging procedures have been reported elsewhere (Kim, 1999). The hydrogen content of the specimen was obtained by averaging 5 different values measured with a LECO RH 404 analyzer. A pre-fatigue crack of 1.7 mm was introduced using an Instron 8501 to obtain the ratio of the fatigue length and the CT length (=a₀/W) equal to 0.5.

The applied stress intensity factor was $12 \text{ MPa}\sqrt{\text{m}}$ at the beginning stage of the pre-fatigue crack and decreased to $10 \text{ MPa}\sqrt{\text{m}}$ after the fatigue crack grew to 1.7 mm.

For DHC tests, CT and CB specimens were subjected to dead-weight loading in a creep machine and in a cantilever beam tester, respectively, while the initiation and growth of the crack were monitored by a direct current potential drop method. Fig. 3 shows a typical thermal cycle to which all the specimens were subjected during DHC tests. The specimens were heated to a peak temperature by $0.5\text{--}1 \text{ }^\circ\text{C}/\text{min.}$, held at the peak temperature for 1 h and cooled down to the test temperature followed by dead-weight loading 30 minutes after reaching the test temperature. The peak temperature was set at $10 \text{ }^\circ\text{C}$ higher than the TSSD temperature corresponding to the charged hydrogen concentration so as to completely dissolve the charged hydrogen into solution at the peak temperature. The crack length was determined on the fractured surface by dividing the area of the DHC crack calculated by an image analyzer by the width.

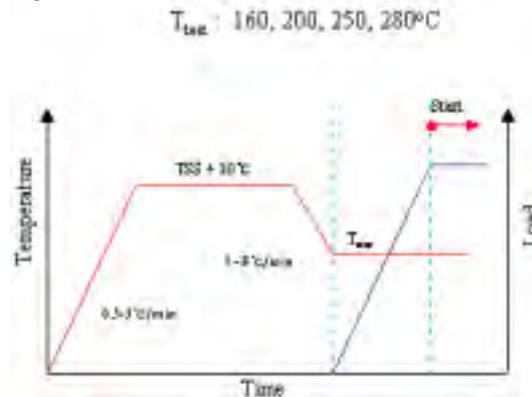


Fig. 3. A typical thermal cycle to which all the specimens were subjected for the DHC tests along with the loading schedule.

4. RESULTS

4.1 Applied Stress Effect on DHC Initiation in a Thermal Cycle

The feasibility of the new DHC model proposed here can be proven if the applied tensile stress facilitates precipitation of the hydrides from zirconium with supersaturated hydrogen, thereby causing a hydrogen concentration gradient. To this end, a model experiment was conducted where the same tensile stress corresponding to $18.4 \text{ MPa}\sqrt{\text{m}}$ was applied to the CB specimens at different times in a thermal cycle as shown in Fig. 4: first, at the beginning of the cycle or at room temperature (A point); second, at the end of a 50 hour-hold at the peak temperature (B point) and third, at the beginning of the test temperature (C point). It should be noted that the stress intensity factor of $18.4 \text{ MPa}\sqrt{\text{m}}$ (termed K) is calculated based on the assumption that a notch with 0.05 mm radius in the CB acts as a sharp crack (Yun, 2003). With the K applied at the points A or B, the reoriented hydrides precipitated over almost the whole cross-section and at a small area near the crack tip, respectively, as shown in Figs. 5a and 5b. With the same K applied at the test temperature of $250 \text{ }^\circ\text{C}$, however, slight precipitation of reoriented hydrides was observed, as shown in Fig. 5c.

The model experiment definitively demonstrates that the applied tensile stress facilitates precipitation of the reoriented hydrides in the deformed regions whose size depends upon when to apply the tensile stress during the thermal cycle. In contrast, with the same tensile stress applied at the test temperature, the relative small plastic deformation of the Zr-2.5Nb matrix suppressed precipitation of the reoriented hydrides somewhere else but the crack tip, as shown in Fig. 5. Surprisingly, the DHC crack did not grow in the specimens with the reoriented hydrides having precipitated all over the whole cross-section on cooling (Fig. 5a) but grew only in the specimen with a few reoriented hydrides precipitated at the crack tip (Fig. 5c). These results cannot be understood with the old DHC model (Dutton, 1977, Puls, 1982, Shi, 1994, 1995) where the tensile stress gradient is the driving force for the DHC. They are also in conflict with the current understanding that reoriented hydrides at the crack tip would facilitate growth of the DHC crack (Nuttall, 1979).

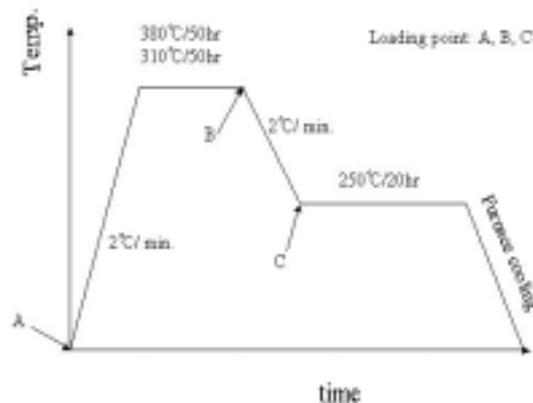


Fig. 4. Thermal cycle treatment to which the Zr-2.5Nb cantilever beam specimens were subjected during DHC tests where the loading time with the stress intensity factor of $18.4 \text{ MPa}\sqrt{\text{m}}$ changed: at RT (A point), at the end of the hold at the peak temperature (B point) and at the test temperature (C point).

In essence, the role of an applied tensile stress is to facilitate precipitation of the hydrides in the zirconium matrix with the supersaturated hydrogen by cooling, not to move dissolved hydrogen up against a tensile stress gradient as suggested in the existing DHC models. The hydrides can precipitate either during cooling (Fig. 5a) or at the test temperature lowered by 50°C from the peak temperature (Fig. 5c). However, precipitation of the hydrides during cooling decreases the concentration of supersaturated hydrogen in the zirconium matrix finally to the equilibrium hydrogen concentration. Thus, with the reoriented hydrides precipitated all over the cross section during cooling (Fig. 5a), then the whole area of the specimen does not maintain the supersaturated hydrogen but has the hydrogen concentration reduced to the equilibrium hydrogen concentration in accordance with the TSSD. Even after the specimen is cooled to the test temperature and is held there under an applied tensile stress, then, no hydrides can precipitate even at the crack tip due to no supersaturation of hydrogen, leading to no DHC as shown in Fig. 5a. In contrast, when the hydrides precipitate only at the test temperature, not on cooling as shown in Fig. 5c, the specimen becomes saturated with dissolved hydrogen in the zirconium matrix that is compressed hydrostatically by cooling. Since the applied tensile stress facilitates precipitation of the reoriented hydrides preferentially at the crack tip, the crack tip has the reduced hydrogen concentration but the bulk region that still maintains the supersaturated hydrogen in solution. Therefore, a gradient of the hydrogen concentration or a driving force for the transfer of hydrogen is developed between the crack tip and the bulk region, leading to the initiation of DHC crack as shown in Fig. 12c.

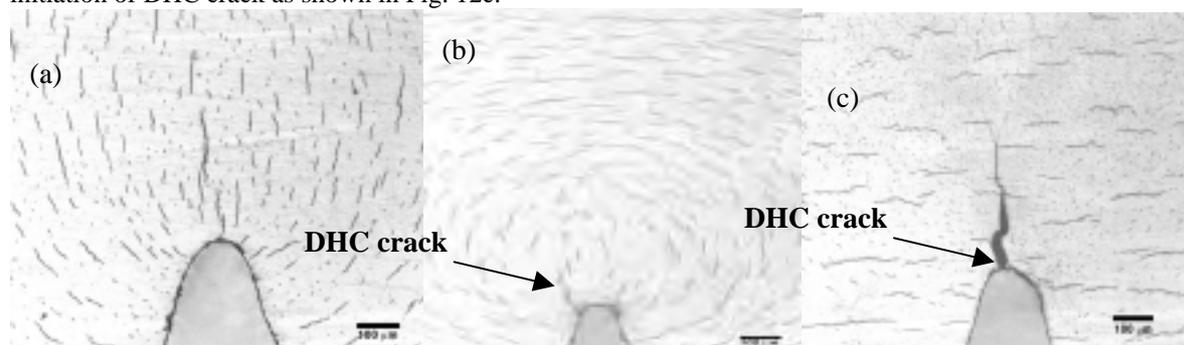


Fig. 5. Reorientation of hydrides in the Zr-2.5Nb tube with time of application of the stress intensity factor of $18.4 \sqrt{\text{m}} \text{ MPa}$ on the CB specimens during the thermal cycle treatment shown in Fig. 4: (a) at the beginning of the thermal cycle, (b) at the end of the hold at the peak temperature of 380°C and (c) at the test temperature of 250°C .

Kammenzind's experiment (2000) demonstrating the transfer of hydrogen only during thermal cycling provides other evidence to the enhanced precipitation of the hydrides by tensile stress. Under a gradual stress gradient, thus, the number of the precipitated hydrides in the region under the highest tensile stress becomes larger

than that in the region under the lowest tensile stress. This results in a gradient of the amount of the precipitated hydrides between the stressed region and the unstressed region though the dissolved hydrogen concentration is kept constant between the unstressed and stressed regions. This leads the stressed region to have the increased hydrogen concentration compared to that of the unstressed region as observed by Kammenzind (2000). Consequently, the transfer of hydrogen only under thermal cycling as reported by Kammenzind (2000) occurs due to the enhanced precipitation of the hydrides by a tensile stress in the stressed region at the cooling stage of the thermal cycling.

4.2 Dependence of Hydrogen Concentration on DHCV of Zr-2.5Nb

According to the new DHC model, since the driving force is the ΔC or the supersaturated hydrogen concentration over the TSSD at a test temperature, as shown in Fig. 2, DHCV should have a hydrogen-concentration dependency at the charged hydrogen concentration equal to and less than the TSSP at the test temperature and will become independent of the hydrogen concentration in excess of the TSSP. To ascertain the feasibility of our new DHC model, DHCV of the Zr-2.5Nb tube was determined at 182 °C with the charged hydrogen concentration changing from 10 ppm to 50 ppm. Fig. 6 shows a DHCV dependence for the Zr-2.5Nb tube on the supersaturated hydrogen concentration. As expected, the DHCV increased with the concentration of supersaturated hydrogen and leveled off to a constant. From the results shown in Fig. 6, we conclude that ΔC governs the DHCV as suggested by the new DHC model.

5. DISCUSSIONS

5.1. Supersaturation of Hydrogen

Since DHC is associated with hydrides, it will be governed by the factors affecting precipitation of the hydrides such as the concentration of supersaturated hydrogen or ΔC , cooling and tensile stress. The best way to make the supersaturation of dissolved hydrogen in zirconium is cooling: cooling from the peak temperature to the test temperature, as shown in Fig. 1, is required to create the supersaturated hydrogen for the DHC. Experimental evidence to this was provided in Fig. 1 where the TSSP temperature generally corresponded to the DHC initiation temperature at the hydrogen concentration in excess of 20 ppm or at the TSSP temperature in excess of 180 °C. This fact explains why the DHC cannot occur in zirconium alloys subjected to no thermal cycle at test temperatures higher than 200 °C (Ambler, 1978, Shek, 1988). The fact that the DHC initiation temperatures generally correspond to the TSSP temperatures at the hydrogen concentration in excess of 20 ppm suggests that an applied tensile stress does not affect the starting temperature where the hydrides can precipitate but causes their enhanced precipitation at the stressed region. In contrast, at as low a hydrogen concentration as 7 ppm, the DHC occurred with little cooling as shown in Fig. 1, indicating that the stress induced transformation of γ -hydrides to δ -hydrides likely causes the transfer of hydrogen towards the crack tip from the bulk region (Nath, 1973, Root, 1996).

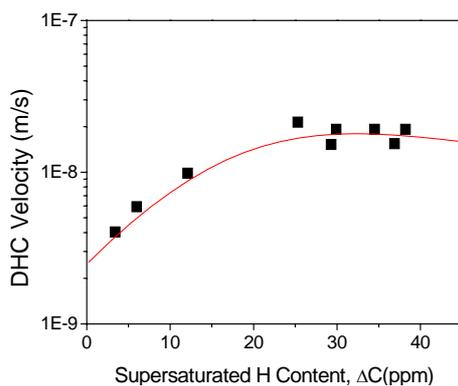


Fig. 6. DHCV at 182 °C of the Zr-2.5Nb tube as a function of the supersaturated hydrogen concentration over the TSSD.

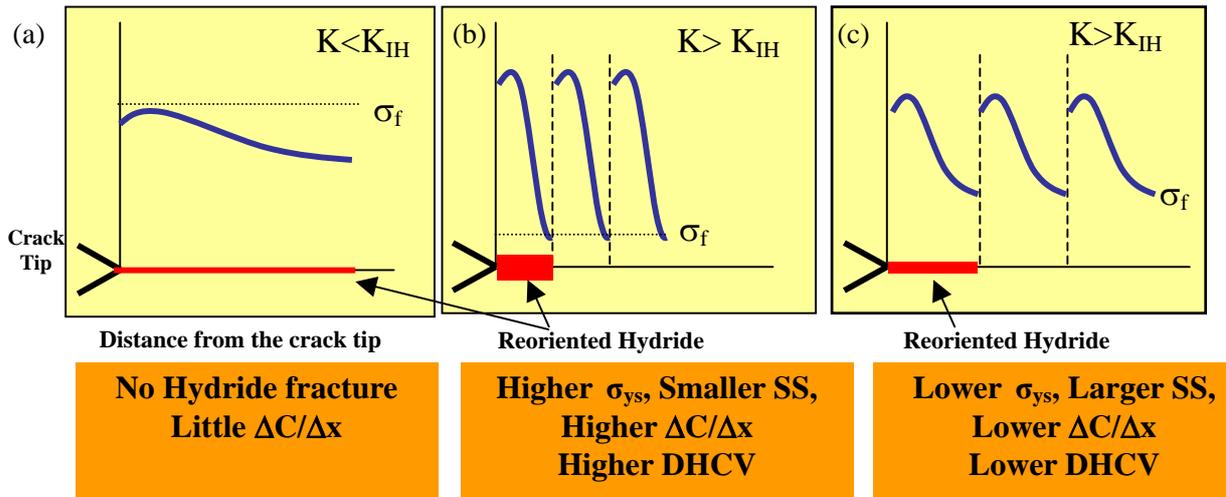


Fig. 7. Dependence of the plastic zone size at the crack tip on fracturing of hydrides and yield strength: (a) wide plastic zone size without fracturing of hydrides, (b) smaller plastic zone due to a higher yield strength and (c) comparatively larger plastic zone due to a lower yield strength.

5.2. Constant DHCV independent of Stress Intensity Factors in Excess of K_{IH}

With the applied stress intensity factor or K in excess of the threshold stress intensity factor, K_{IH} , the maximum concentration of supersaturated hydrogen or ΔC_{max} that the Zr-2.5Nb specimens can have between the crack tip and the bulk region is the TSSP-TSSD at the test temperature as shown in Fig. 2:

$$\Delta C_{max} = \text{TSSP-TSSD} \quad (1)$$

at a constant temperature and hydrogen concentration. In other words, the ΔC or the driving force for the DHC becomes independent of the K as long as it exceeds the K_{IH} at the conditions of constant temperature and hydrogen concentration. However, since the DHCV is governed by a hydrogen concentration gradient, a discussion has to be made not only about the driving force or ΔC , but also the plastic zone, or Δs where the ΔC is developed. Necessary condition for a DHC crack to grow continuously is fracturing of the hydrides precipitated at the crack tip. The gradient of the hydrogen concentration formed at the crack tip under the applied K would decrease gradually with time unless the hydrides fracture as shown in Fig. 7a. With the applied K in excess of the K_{IH} as shown in Figs. 7b and 7c, the hydrides precipitate ahead of the crack tip, grow to the critical length corresponding to the K_{IH} and then fracture. As soon as the crack grows through fracturing of hydrides, the hydrogen concentration gradient will go away. Then, the grown crack waits until the start of the next cycle where the hydrides precipitate, grow and fracture over the newly formed plastic zone ahead of the crack tip, whose size is the same as the former plastic zone size. The plastic zone size is, therefore, determined by the yield strength of the zirconium alloys and the K_{IH} , not by the applied K because the hydrides fracture whenever the applied K reaches the K_{IH} (Choi, 2004):

$$\Delta s = k \left(\frac{K_{IH}}{\sigma_y} \right)^2, \quad (2)$$

where Δs is the plastic zone size, k is a constant, K_{IH} is the threshold stress intensity factor and σ_y is the yield strength. As shown in Eqs. 1 and 2, the Δs and ΔC , governing the hydrogen concentration gradient at the crack tip, becomes independent of the applied K , leading to the constant DHCV independent of the applied K . In essence, the constant DHCV independent of the applied K is attributed to the constant driving force or ΔC independent of the applied K and the constant plastic zone size due to the discontinuous growth of the crack at the constant K_{IH} .

It should be noted that the growth of the crack in the axial direction of the Zr-2.5Nb tube through fracturing of the hydrides accompanies twinning of the zirconium matrix ahead of the crack tip, leading to the formation of a striation line (Kim, 2001). As a result of that, the plastic zone size will correspond to the striation spacing. Consequently, a correlation of the striation spacing and the yield strength can be obtained from Eq. (2): the

striation spacing on the DHC fracture surfaces has a yield strength dependence and full independence of the applied K (Kim, 2004, Choi, 2004).

6. CONCLUSIONS

For the first time, we propose a new DHC model where a driving force for DHC of zirconium alloys is the supersaturated hydrogen concentration or ΔC resulting from hysteresis of the terminal solid solubility of hydrogen in zirconium upon heating and cooling. Experimental evidence to the feasibility of the new DHC model was provided through the model experiments: the applied stress promoted precipitation of the reoriented hydrides in the plastic zone, no DHC occurred in the absence of the supersaturation of hydrogen resulting from the reoriented hydrides precipitated all over the cross-section on cooling, and the DHCV of the Zr-2.5Nb tube increased with the ΔC and leveled off to a constant at the ΔC in excess of TSSP-TSSD at the test temperature. The new DHC model proposed here comprehends the constant DHCV independent of the K accounting for the facts that the ΔC becomes independent of the K and the plastic zone size Δs is governed by the yield strength and the threshold stress intensity factor over which the hydrides precipitated at the crack tip fracture, initiating the DHC.

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