

## Effect of Neutron Irradiation on the Delayed Hydride Crack Velocity in CANDU Pressure Tube Materials

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### 1. Introduction

Since the pressure tube in Pickering CANDU reactor cracks in 1974, the delayed hydride cracking (DHC) has been intensively studied. It is a stable crack growth mechanism in zirconium alloys. Hydrogen accumulates at a stress raiser. If sufficient hydrogen is present, hydrides form and, and if the stress is high enough, hydride fracture and the crack advance. DHC is the important factor to govern the safety of pressure tubes in CANDU reactor. During reactor operation the accumulation of neutron irradiation on tube effects the behaviors of DHC to be accelerate crack velocity. It is very important to understand the effect of neutron irradiation on the tube material during reactor operation to protect the tube breakage. In this study the changes in the DHCV of the tubes which arise from exposure to irradiation are reviewed.

### 2. Experiments

All test specimens were fabricated to curved compact tension shape with 17mm in width (17mm-CCT) from the coolant inlet, middle and outlet in M-11 tube operated at Wolsong-1 reactor and the unirradiated one, which is double melted Zr-2.5Nb. Fatigue pre-cracking was performed to 1.7mm in crack advance with the method of  $\Delta K$  decrease. Hydrogen charging to the specimen in the amount of about 50 ppm was done by letting hydrogen diffuse into the metal from the hydride layer produced electrically.[1] The soaking temperature was 275 °C. The static universal testing machine, direct current potential drop (DCPD) system, furnace and data acquisition system were set up in hot cell area to perform DHCV test. The load to apply the specimen is determined in the compliance with ASTM E399 to 12 MPa·m<sup>1/2</sup> in K-value. The crack increment amount during test was continuously measured using DCPD system with 6 Ampere in input current. After finishing test the specimen was heat-tinted during 1 hour on 250 °C and fractured. The fractured surface was investigated by highscope system and SEM function of EPMA.

### 3. Result and Discussion

#### 3.1 Crack Growth Behaviors

Fig. 1 shows the typical variations of load, load line displacement and dropped voltage during test. In figure the dropped voltage start to increase behind applying load, so called incubation time. This phenomenon is

guessed to be time for hydrogen to accumulate crack tip and formation a hydride. Incubation time in the irradiated specimen shows to be shorter than in the unirradiated one. After the starting to change it increase uniformly with time. It means that the DHC advances with invariable velocity after the crack being initiated.

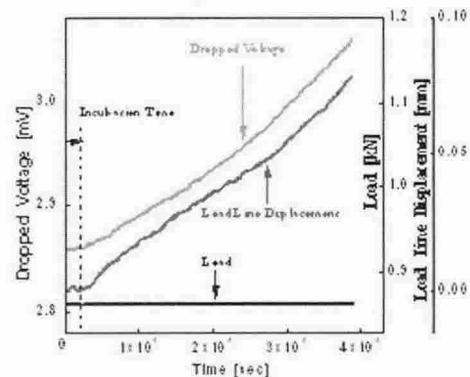


Fig. 1 Typical load, load line displacement and drop voltage behavior during the test

#### 3.2 Determination of activation energy

Fig. 2 shows the logarithm DHCV plot for the irradiated and the unirradiated specimens according to the inverse of the absolute test temperature. As expected the DHCV in both materials increased exponentially with proportional to the temperature and the velocity in the irradiated one was faster 4~6 times than in the unirradiated one. According to Simpson and Pulse [2, 3] the DHCV can be expressed in the form of the thermal activation energy proposed by Arrhenius. From the least square linear fit in Fig. 2 the velocity equation likes as  $V = 5.1 \times 10^{-3} \exp(-47,933/RT)$  for the unirradiated materials,  $V = 3.9 \times 10^{-2} \exp(-50,514/RT)$  for the irradiated ones. From the slope linear fit, the activation energy of DHCV was determined to be about 48 kJ/mol and 50.5 kJ/mol for the unirradiated and the irradiated materials respectively.

#### 3.3 Effect of Irradiation

To investigate the DHCV variation according to the irradiation amount these tests and Sagat's results [4] are plotted in Fig. 3, which divided into fluence with operating positions. From the figure the DHCV has a trend to increase rapidly at the initial stage of irradiation

up to ( $\sim 1 \times 10^{25}$  n/m<sup>2</sup>) and saturate to constant value the accumulation of neutron irradiation amount.

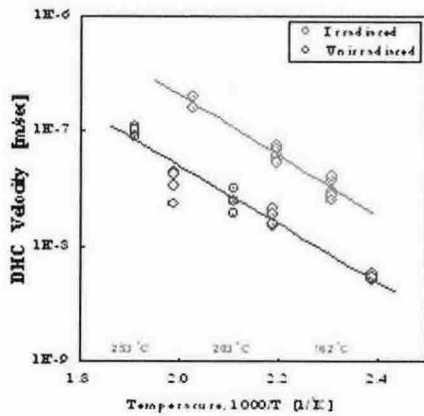


Fig. 2 Temperature dependence of the DHCV in the irradiated and the unirradiated pressure tube materials.

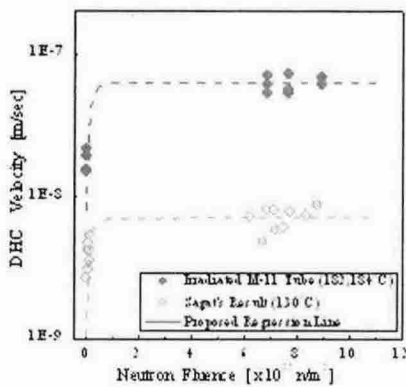


Fig. 3 Dependence of DHCV on irradiation fluence

It is known that a neutron irradiation on Zr-based material enhances the yield strength and the terminal solubility of hydrogen. [5,6] To prospect the velocity of the irradiated materials from the unirradiated ones the effect of two parameters are reviewed to introduce two scale factors. From the Fig. 4 the DHCV in the irradiated materials prospect well introducing  $C_d$ ,  $C_{ys}$  in the proposed scaled factor.

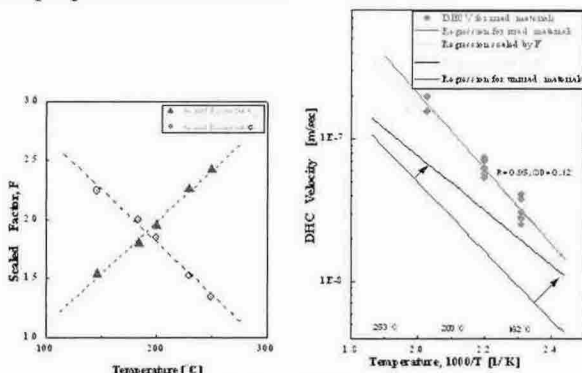


Fig. 4 Scaled factor variations (left) and the proposed DHCV (right) scaled by the yield strength and the dissolved hydrogen contents.

3.4 Fracture Surface Investigations

Using the highscope and the shielded EPMA fractured surfaces are investigated to review the characteristic of crack increment. The fractographic observation in Fig. 5 indicated that, in the regions between striations, crystallo-graphic facets are present, confirming the brittle fracture of the hydride platelets. This suggests that, when the hydrides regions reaches a critical size, the hydrides crack by cleavage.

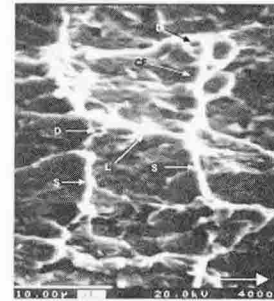


Fig 5 Visual (left) and SEM fractograph of the fractured surface by DHC

4. Conclusion

Using the pressure tube materials operated in Wolsong-1 CANDU reactor during 13 yrs, the DHCV behaviors were reviewed on the point of the effect of neutron irradiation. The DHCV equation of the irradiated materials is proposed and the variation trend of velocity with irradiation fluence. Introducing the scale factors of yield strength and dissolve hydrogen content the prospective model for the velocity in the irradiated materials was proposed and the characteristics of crack increment on fractured surface were investigated.

REFERENCES

[1] A. D. Lepage, W. A. Ferris and G. A. Ledoux, "Procedure for adding hydrogen to small sections of zirconium alloys", FC-IAEA-03, 1998.  
 [2] R. Nuttal, M. P. Pulse and L. A. Simpson, "Mechanisms of hydrogen induced cracking in hydride forming metals", Metallurgical Transaction, Vol. 8A, p. 1553, 1977.  
 [3] M. P. Pulse, "Effect of crack tip stress states and hydride matrix interaction stresses on delayed hydride cracking", Metallurgical Transaction, Vol. 21A, p. 2905, 1998  
 [4] S. Sagat, S. E. Colemann, M. Griffith and B. J. S. Willkins, "The effect of fluence and irradiation temperature on delayed hydride cracking in Zr-2.5Nb", ASTM STP 1245, p. 35, 1994.  
 [5] S. B. Ahn, Y. S. Kim and J. K. Kim "Tensile behavior characteristics of CANDU pressure tube material degraded by neutron irradiations", Journal of KSME, Vol. 26A, p. 188, 2002.  
 [6] A. McMinn, E. C. Darby and J. S. Schofield, "The terminal solubility of hydrogen in zirconium alloys", ASTM STP 1354, p. 173, 2000.