Journal of the Korean Nuclear Society
Volume 32, Number 1, pp.1~9, February 2000

DHC Characteristics of M11 Pressure Tube in Wolsong Unit 1

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(Received April 28, 1999)

Abstract

Delayed hydride cracking (DHC) velocity and threshold stress intensity factor for DHC (K_{IH}) tests in the radial direction on M11 pressure tube material in Wolsong unit 1 were carried out following the Atomic Energy Canada Limited (AECL) standard test procedure in order to identify the effect of undercooling on DHCV and to acquire the K_{IH} data. The results showed that K_{IH}' s were 8.8 ± 0.8 MPa$\sqrt{m}$ in the back offcut and 11.4 ± 0.7 MPa$\sqrt{m}$ in the front offcut. The fact that K_{IH} in the front offcut is about 20% higher than that in the back offcut is attributed to the microstructural difference between the materials of the front and back ends. K_{IH}' s in M11 pressure tube appeared to be higher than the values from the tubes made of double melted ingot reported earlier. This can be interpreted by the fact that very small amounts of Chlorine (Cl) and Phosphorus (P) are contained in the ingot and that the content of the harmful elements in the M11 pressure tube is equivalent to that made of a quadruple melting process. DHC velocities at 250℃ in the front offcut in the radial direction are measured to be $5 \sim 8 \times 10^{-8}$ m/s. The results show that the prior thermal history change the DHC velocity significantly. This effect was confirmed by the experiment of undercooling prior to the DHC tests.

Key Words : delayed hydride cracking (DHC), pressure tube, threshold stress intensity factor (K_{IH}), DHC velocity DHCV), cantilever beam specimen, brittle fracture, Zr-2.5%Nb

1. Introduction

Pressure tubes in CANDU reactors have suffered primary coolant leakage accidents due to delayed hydrogen cracking (DHC) which is related to the hydrogen concentration, the applied stress, the presence of flaws, and the precipitation of hydrides at the crack tip. Pressure tubes in CANDU reactors absorb hydrogen formed by corrosion from coolant and precipitate the hydride later when the hydrogen content exceed the terminal solid solubility during operation [1, 2].

There may be several methods to mitigate DHC susceptibility [3]. They are:

1) the reduction of the hydrogen ingress rate by lowering the corrosion rate and by Cr-plating between the end fitting and pressure tube in a rolled-joint.

2) the hydrogen ejection using Pd-plating and the addition of oxygen in the annulus gas system.
3) the modification of texture from a circumferential to a radial one.

It has been reported recently that the crack growth rate increases exponentially with the basal pole component and K_{IH} decreases linearly with the basal pole component in the direction normal to the cracking plane. The behavior of K_{IH} variation could be explained by the uniformly dispersed aggregate composite theory using the basal pole component as a brittle aggregate and using the remaining fraction as a ductile matrix [4].

There has been no coolant leakage accident in Wolsong unit 1 during operation since 1983. However, three of the 380 pressure tubes in Wolsong unit 1 have been retubed to evaluate the condition of pressure tubes and to monitor degradation of materials to ensure the integrity of the pressure tubes for the safer operation. Some of these materials have been tested destructively to obtain the fracture toughness and so on. The K_{IH} and DHC velocity on irradiated M11 pressure tubes were measured at Chalk River Laboratories (CRL) in 1995 and reported in a Korea Electric Power Corporation (KEPCO) internal report.

Even though Wolsong unit 1 has operated over 14 years, there has been no study on the DHC behavior of the unirradiated Wolsong pressure tube materials in Korea up to now. Thus, the DHC tests on M11 pressure tube were carried out in order to acquire the characteristic DHC data of Wolsong pressure tube and to identify the effect of under-cooling on DHC velocity, and the K_{IH} procedure used in this study are described.

2. Experimental

The materials used in this study were the front and back offcuts of M11 pressure tube in Wolsong unit 1, which were provided by KEPCO to AECL-CRL for K_{IH} measurement for comparison with an irradiated one. Cantilever beam (CB) specimens were machined from the front and back offcuts, 9 and 6 specimens, respectively. There might be slight microstructural differences and tensile properties between the front and back offcut pieces due to the 6 m long tube geometry. The specimen dimension is shown in Fig. 1.

Hydrides were formed at the surface about 20μm thickness electrolytically in an acidic solution, and, then, the hydrided materials were homogenized for 96 hours at 300℃ to 60 ppm. Then, the hydrogen content was analysed by the vacuum extraction method after removing all the surface hydride from the sample. Although there were some hydrides on the CB specimen surface, they did not affect the test results because soaking was carried out below the TSS temperature for the specimen. The soaking treatment was selected at 307℃ for 1 hour in the case of 60ppm hydrogen,.

A schematic test system is shown in Fig. 2. The load reducing method was used to determine K_{IH} at 250℃ according to the AECL procedure. Crack propagation was detected by an acoustic emission (AE) sensor and no cracking for a 24 hour criterion at a constant load was used to determine the K_{IH}. AE counts were used to calculate the in-situ crack length at each crack propagation. Once the load is applied according to the initially required K_I, then, the crack start to propagate with AE counts. The load is decreased with the crack propagation if there is no adjustment of load since the cantilever is lowered with cracking. The required K_I is maintained by the action of stepper motor controlled by computer. AE counts were monitored by computer, and then the estimated in-situ crack length could be calculated using the number of count and the AE constant. Again, in order to reduce K_I the required load for a given

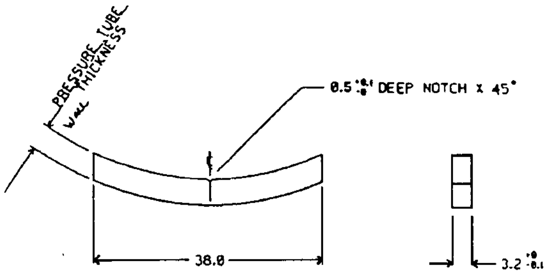

Fig. 4. Schematic Drawing of Cantilever Beam Specimen(CB)

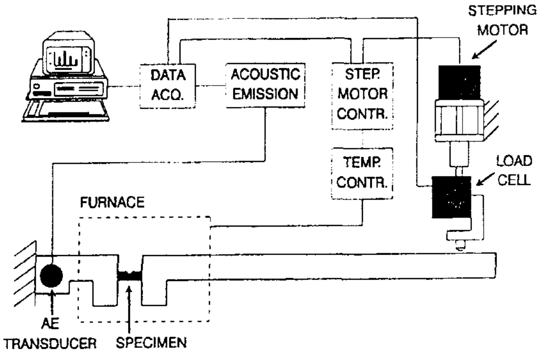

Fig. 2. Schematic Diagram of Cantilever Beam Specimen Testing System

crack length was calculated by formulae(1) described below, and then the reduced load was applied by a stepper motor controlled by a computer system through the load cell. The AE constant used to operate the test system was 3×10^{-9}m/count and the applied load was reduced by 3% when the crack length reached about 3μm. The variations of the applied load, K_I, and AE counts during K_{IH} test were shown in Fig. 3.

The real crack length should be measured and the real K_I during the test should be calculated again after breaking the specimen, and then, K_{IH} was determined at the end, because it was impossible to know the in-situ crack length during a test,

The applied K_I is calculated by the following equation [2]:

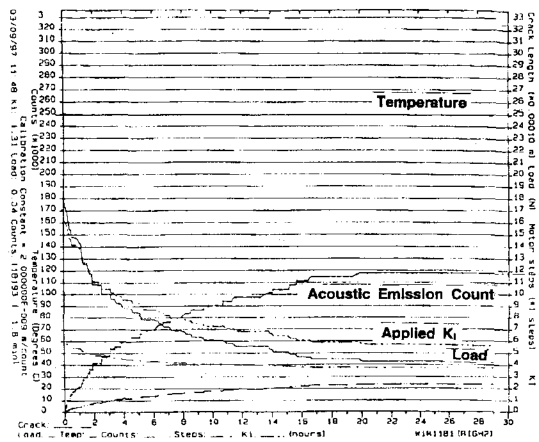

Fig. 3. Acoustic Emission Counts, Load, and K_I Variation During K_{IH} Test

$$K_I = 4.12\, M(X^{-3} - X^3)^{0.5}/BD^{1.5} \qquad (1)$$

where M is the bending momentum (grip weight x length of center of gravity of bar + applied load x distance from notch to loading point), B is the breadth of specimen, D is the depth (radial thickness), A is the crack length, and X=1-A/D.

The DHC velocity in the radial direction was measured at 180~260℃ using the same CB specimens used in K_{IH} determination before breaking the specimen. The DHC velocity was calculated by crack length divided by cracking time. The crack length was measured by the normalization of a cracking area by breadth. The effect of undercooling on crack velocity was tested in some specimens.

3. Results and Discussion

The hydride distribution in the plane normal to the longitudinal direction in a M11 back offcut specimen is shown in Fig. 4. Most hydrides are precipitated in the longitudinal-circumferential plane as platelets.

The (0002) direct pole figures for the front and

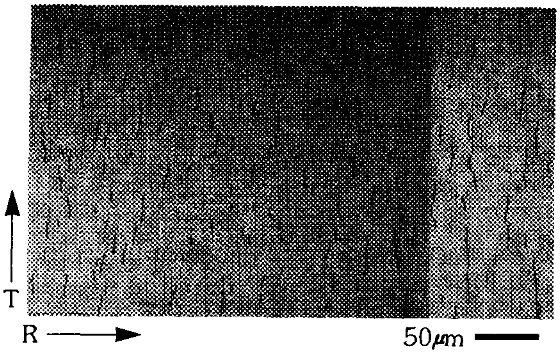

Fig. 4. Hydride Morphology in Wolsong Uint 1 M11 Pressure Tube

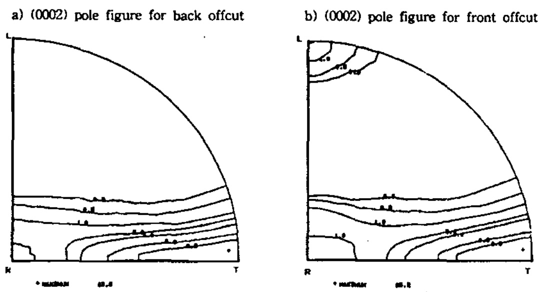

Fig. 5. (0002) Direct Pole Figures in Wolsong Unit 1 M11 Pressure Tube

back offcuts are shown in Fig. 5 a) and b), respectively. The basal poles are concentrated in the transverse direction and distributed in the radial-transverse plane in 50° range. These results are consistent with the literature [5].

The fracture surface in a CB specimen is shown in Fig. 6. It is possible to distinguish the machined notch, pre-fatigue crack, and DHC crack, and the first crack below the pre-fatigue crack is generated during the K_{IH} test while the second crack is generated during the DHC velocity test. It is clearly shown that the crack frontier is not flat. This means that the load enduring length at the crack tip is longer than the specimen width, and this may result in higher K_{IH} due to a larger load during tests.

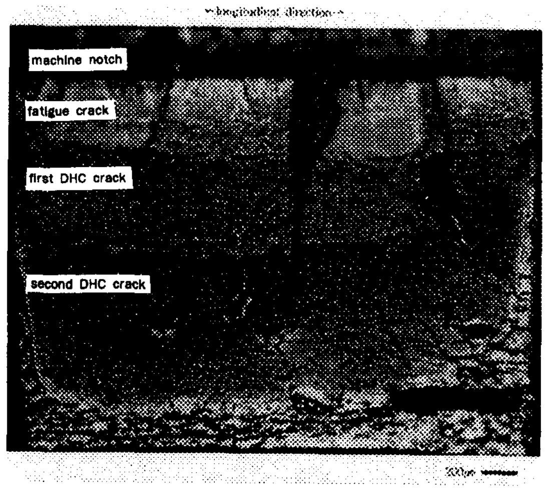

Fig. 6. Optical Micrographs for Fractured Surface of Cantilever Beam Specimen

K_{IH} is measured as 8.8 ± 0.8 MPa$\sqrt{}$m from 6 specimen in the back and 11.4 ± 0.7 MPa$\sqrt{}$m 9 specimen in the front offcuts. These values are higher than that of pressure tube made of the double melted ingot reported earlier. This seems to be due to the low content of Cl and P compared to pressure tubes made of double melted ingot. It is confirmed that the contents of Cl and P in M11 pressure tube were 2-3 ppm and 6-7 ppm, respectively. However those of Cl and P in O-08 which is made of recycled scrap according to the document were 0.1 ppm and 5 ppm, respectively, while the content of Cl in double melted pressure is in the range of 5-15 ppm. Therefore, the chemistry of M11 pressure tube is almost equivalent to a quadruple melted one in the aspect of content of harmful elements. Furthermore, it has been proven that pressure tubes made by the quadruple melting process have a higher toughness due to the lower content of Cl in the pressure tube [3, 6].

The fact that tubes having higher fracture toughness show higher K_{IH} suggests that there is a close relation between the fracture toughness and

Table 1. The Basal Pole Component, Dislocation Density, and Nb Content in β-phase in Front and Back end Offcuts of M11 Pressure Tube for Wolsong Unit 1

components \ offcuts		Front End Offcut			Back End Offcut		
specimen direction		R	T	L	R	T	L
basal pole component		0.289	0.633	0.076	0.342	0.612	0.045
dislocation density, $[10^{14}/m^2]$	<a> type	1.55		3.14	1.44		3.13
	<c> type	0.28	1.15		0.3	0.66	
integral breadth, $[10^{-3}A^{-1}]$	$10\bar{1}0$			3.49			3.55
	$11\bar{2}0$	2.97			2.98		
	0002	3.04	4.34		2.84	3.65	
Nb content of β-phase [%]			53			67	

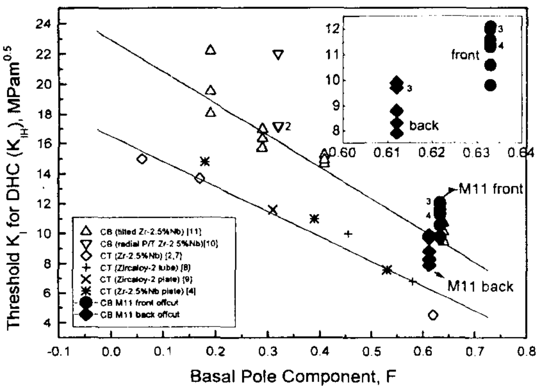

Fig. 7. Comparisons of K_{IH} in M11 Pressure Tube Materials with the Various Zr-alloy

K_{IH}. It is understood that the fracturing process by DHC is not a perfectly brittle one, since the K_{IH} variation with basal pole component (F) could be explained by the rule of mixture of ductile matrix and brittle hydride [4].

The K_{IH}' s for the M11 pressure tube were plotted against the basal pole components (F) in Fig. 7 together with the results for Zr-2.5%Nb and Zircaloy-2 materials using the compact tension (CT) specimens and the CB specimens. The texture dependency of K_{IH} was properly interpreted by the rule of mixture using the basal pole component, F, as a volume of brittle hydride and the remaining portion, (1-F), as the volume of ductile matrix.

The K_{IH}' s values for CT specimens were selected in the lower bound values for comparison in the literatures [2, 4, 7-11]. The K_{IH}' s measured from the CB specimens were always higher than that of CT. This fact may be closely related to the nature of fracture process during DHC, because the cracking direction in CT and CB specimens are the longitudinal direction and the radial direction, respectively. This means that the grain orientations at the crack tip varies significantly in the CB specimens compared to the CT specimens during DHC, as shown in Fig. 5. Therefore, the cracking processes may be disturbed and this seems to be the reason why the K_{IH} in the CB specimens are always higher than that in the CT specimens.

The K_{IH}' s in the front offcut are about 20% higher than those of the back offcut, even though there are little differences in dislocation densities between front and back offcut, as shown in Table 1. This means that K_{IH}' s affected by the small microstructural variations.

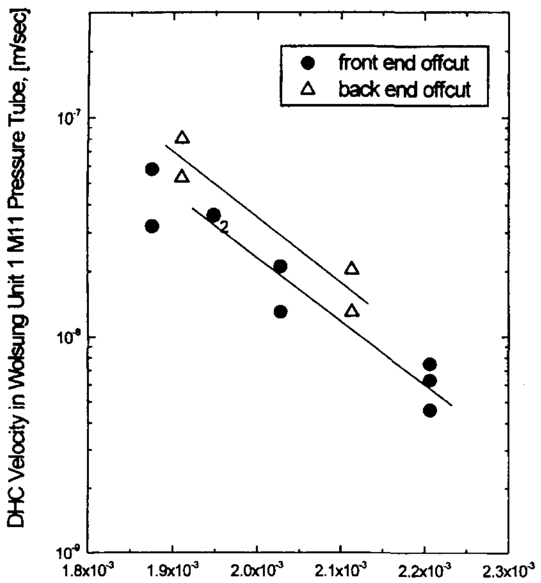

Fig. 8. DHC Velocity with Reciprocal Temperature in M11 Pressure Tube

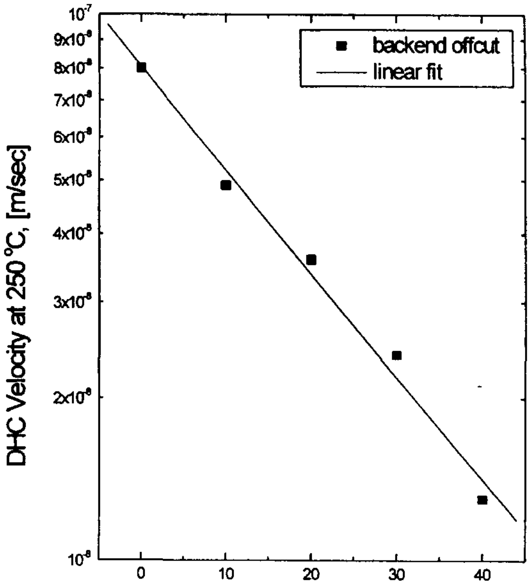

Fig. 9. Effect of Undercooling on DHC Velocity at 250℃ in M11 Pressure Tube

The K_{IH} in an irradiated M11 pressure tube is slightly lower than that of the back end, although the number of K_{IH} data for irradiated M11 pressure tubes is not sufficient enough and the location of the K_{IH} specimen in an irradiated M11 is neither at the back nor front end. This suggests that irradiation does not change K_{IH} significantly and that the selection of K_{IH} for flaw assessments should be carefully taken according to the position of flaw and according to the proper history of a pressure tube.

The reason for selection a testing temperature at 250℃ is that the maximum stress is applied at 250℃ under normal operation in the inlet of coolant, and hydride precipitates at the inlet since the solubility of hydrogen in pressure tube materials increases with temperature. If the testing temperature is lowered, then the testing time becomes significantly longer than that at 250℃. This seems to be related to the longer time needed to precipitate a hydride of critical length.

The standard deviation measured in this study seems to be acceptable, and it is reasonable that the standard deviation is reduced with increases in the number of specimens. This standard deviation seems to reflect material to material variation, gripping error, and so on. The hydrogen content may affect DHC cracking behavior, and, for example, the cracking plane becomes nonuniform with an unnecessarily high hydrogen content. The AECL-CRL is currently developing a new test procedure and test system to reduce the standard deviation below 0.5 $MPa\sqrt{m}$.

Crack velocities in the radial direction were plotted with reciprocal temperatures in Fig. 8. The crack velocities at 250℃ in the front end offcut were measured to be $5\sim8\times10^{-8}$ m/s . The crack velocity in the back offcut is slightly higher than that of the front end offcut. This phenomenon seems to be due to the same reason as K_{IH}.

Table 2. The Amount of Error on K_{IH} from Farious Uncertainty

kind of uncertainty	average magnitude, [MPa$\sqrt{m}$/unit]	possible magnitude	subtotal, [MPa$\sqrt{m}$]	total amount, [MPa$\sqrt{m}$]
length of center of gravity [1 mm]	0.022	1mm	0.022	0.346
loading distance [1 mm]	0.048	3mm	0.144	
grip weight [1 gram]	0.009	2g	0.018	
carck length [0.1 mm]	0.072	0.1mm	0.072	
load cell output [0.01 N]	0.018	0.05N	0.09	

Crack velocities at 250℃ are plotted with undercooling temperatures after a soaking treatment in Fig. 9. The crack velocities decrease exponentially with the undercooling temperatures. This result suggests that crack velocity is governed by available hydrogen in the solution, and the DHC mechanism is closely related to diffusion and the precipitation of hydrogen in the matrix. It is possible to estimate DHC velocity in the longitudinal direction using that of the radial direction since it is reported that DHC velocity in the longitudinal direction is two times that of the radial direction, based on the wide experimental experiences [12].

Crack length is measured by calculation of the cracking area normalized by the breadth of the specimen, as described above, because a DHC crack does not show linear frontier usually. There may be some errors in measuring the crack length. Although it is very difficult to describe the error of K_I from the crack length measurement, it is possible to calculate that a 0.1 mm error in a crack length measurement induces K_I= 0.061-0.078 MPa$\sqrt{m}$ error on average, which would be dependent on in-situ crack length. The error from a crack length measurement seems to be very significant compared to the effect of other factors. If there is 1.0 mm error in the loading distance measurement, this produces K_I= 0.05 MPa$\sqrt{m}$ error on average. It seems that the measurement error in length would be less than 1.0 mm. If there is a 1.0 mm error in measurement of the length of center of gravity, the error for K_{IH} is calculated to be K_I= 0.022 MPa$\sqrt{m}$ on average. If a 1.0 gram error (0.01 N) occurs in grip weight measurement, this induces K_I= 0.01 MPa$\sqrt{m}$ error in average.

The other may come from the load cell itself. Load cells show different loads sometimes. It is very difficult to identify whether the signal change comes from the real load change or not, although it seems to be related to the ambient temperature changes during the test. Because this force is a major part of the momentum in K_{IH} testing, this error may induce a relatively larger error. If there is a 0.01N (≒1 gram) error, this will induce an error of 0.019 MPa$\sqrt{m}$ on average.

These errors calculated above are sorted out in Table 2. All of these should be added to get a conservative bound although some error could act to be positive or negative each other. Even though some of these could be removed by careful operation, others could be reduced by design change of tester only. Therefore, the magnitudes of standard deviation reported in this study seem to be the summation of the measurement error and material variations.

4. Conclusions

The K_{IH}'s for M11 pressure tube in Wolsong unit 1 were 8.8±0.8 MPa√m from 6 specimens in the back offcut and 11.4±0.7 MPa√m from the 9 specimens in the front offcut. It is confirmed that the M11 pressure tube in Wolsong unit 1 has higher KIH's compared to pressure tubes made of double melted ingot. This effect seems to be due to the fact that M11 pressure tube contains a small amount of Cl and P which are known as detrimental elements for fracture toughness of Zr-2.5%Nb alloy. The DHC velocities in the radial direction were measured to be $5\sim8\times10^{-8}$ m/s at 250℃ in the front end offcut of the M11 pressure tube. The crack velocities decreased exponentially with an increase in undercooling temperatures. It can be concluded that the crack velocity and K_{IH} are very sensitive properties to microstructure and prior thermal history.

Acknowledgement

This work has been carried out in the CANDU Pressure Tube Materials project as a part of the Nuclear R&D program funded by Ministry of Science and Technology in Korea. The authors would like to acknowledge people working in building 375 in AECL-CRL for their help, and KEPCO for providing M11 offcuts for the testing.

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