Measurement of DHC Velocity in CANDU Pressure Tubes

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ABSTRACT

This paper presents the experimental results obtained as part of the IAEA Co-ordinated Research Project on “Hydrogen and Hydride Induced Degradation of the Mechanical and Physical Properties of Zirconium-based Alloys”. Two tubes made from cold-worked Zr-2.5Nb were tested: one tube was made using material that had been melted four times, while the second, typical of Unit 1 of Cernavoda, Romania, was made earlier from material that had been double melted. Measurements of DHC velocity have been performed in the axial direction of the pressure tubes using compact toughness specimens in a standard procedure. The objective of the tests was to discern any difference in cracking behaviour between tubes made from different starting materials.

The crack propagation rates were measured on samples containing 28 to 60 ppm hydrogen in the temperature range 144 to 250 °C. The applied load was kept constant during the tests, corresponding to an initial stress intensity factor of $K_I = 15 \text{ MPa} \cdot \sqrt{\text{m}}$. The crack length was monitored using the Potential Drop method. Both materials behaved in a similar manner. The DHC axial crack velocity was about $9 \times 10^{-8} \text{ m/s}$ at 250 °C and had a temperature dependence that followed an Arrhenius-type law with an activation energy of about 48 kJ/mol. These results show that DHC behaviour of Zr-2.5 Nb pressure tubes is not affected by ingot melting practice.

KEY WORDS: Zr-2.5Nb alloys, CANDU reactor, Delayed Hydride Cracking

INTRODUCTION

The CANDU-PHWR (Canada Deuterium Uranium – Pressurized Heavy Water Reactor) uses natural uranium, in the form of pellets of UO₂ contained in Zircaloy-clad fuel bundles, and heavy water as moderator and heat-transport medium. The design is based on about 400 individual channels, which hold the fuel bundles, comprising pressure tubes of Zr-2.5Nb alloy joined to 403 stainless steel end-fittings that operate in an environment of heavy water at elevated temperature (250 to 300°C) and internal pressure (10 MPa) in a fast neutron flux ($\approx 10^{17} \text{n/m}^2\text{s}$). The design allows for dimensional changes that are the consequences of the irradiation and thermal and mechanical stresses imposed by these operation conditions. The structural integrity and operating life of the Zr-2.5Nb pressure tube may be affected by the gradual pick-up of hydrogen, as deuterium, that modifies the mechanical properties by reducing ductility and crack growth resistance; certain thermomechanical conditions may lead to delayed fracture of this alloy. This mechanism, called Delayed Hydride Cracking (DHC), was the cause of leakage in some Zr-2.5Nb pressure tubes of CANDU plants at Pickering and Bruce [1, 2] and Russian RMBK reactors at Kursk and Chernobyl [3] and may contribute to the large axial splits experienced in the Zircaloy fuel cladding in light water reactors [4, 5].

The mechanism of DHC describes the propagation of a flaw when zirconium hydrides are present in the component. When a tensile stress is applied, hydrogen accumulates by diffusing up the stress gradient at the flaw tip where a hydride forms. The hydride grows as more hydrogen accumulates and, if the critical conditions of hydride size, maximum stress and critical strain are reached, the hydride cracks, and the flaw extends by the length of the hydride. The process is then repeated [6].

The Cernavoda Nuclear Power Plant - Unit 1, Romania, is a CANDU-PHWR. Its Zr-2.5Nb pressure tubes were fabricated in the early 1980’s and we needed to confirm that their properties were similar to those of tubes made more recently. Early pressure tubes were made from ingots, comprising a mixture of sponge zirconium and recycled zirconium scrap material, that were melted twice while current pressure tubes are made from ingots that have been melted four times to ensure that the fracture toughness remains high. [7, 8]. In this paper we compare the DHC properties of a typical Cernavoda pressure tube with one made using quadruple-melted material. The experimental methods were developed at the Institute for Nuclear Research, Pitesti, Romania, and at Chalk River Laboratories, Canada [9], as part of an IAEA Co-ordinated Research Programme.
EXPERIMENTAL PROCEDURES

Material Tested

This paper presents the experimental results obtained as part of the IAEA Co-ordinated Research Project on “Hydrogen and Hydride Induced Degradation of the Mechanical and Physical Properties of Zirconium-based Alloys”. Two tubes made from cold-worked Zr-2.5Nb [10] were tested: one tube was made using material that had been melted four times, while the second, typical of Unit 1 of Cernavoda, Romania, was made earlier from material that had been double melted. The α-phase grains were elongated platelets, with an aspect ratio of about 1:5:50 in the radial, transverse and longitudinal direction, respectively, and surrounded by partially decomposed β-phase. The longitudinal tensile strengths at 300°C of the two types of tubes are 530 MPa for the four times melted material and 521 MPa for the double melted one.

Works Performed

The experimental programme consisted of four steps:

• Hydriding rectangular Zr-2.5Nb samples, cut from pressure tube rings, to a controlled hydrogen concentration;
• Preparing Curved Compact Tension (CCT) specimens;
• DHC crack propagation under constant loading;
• Analysis of the fracture surfaces to evaluate DHC velocity.

Hydrogen was added to each specimen using a method based on the diffusion of hydrogen into the zirconium matrix from a surface layer of hydride produced electrolytically in 0.1 molar H₂SO₄ solution at 65°C [11]. The diffusion annealing process, carried out at a specified homogenization temperature and time depending on the required hydrogen concentration, the solubility limit (TSS) and the diffusion coefficient, ensured a uniform distribution of hydrides in the sample (Figures 1 and 2).

The hydrogen concentrations were chosen to represent possible hydrogen concentrations picked up by the pressure tubes during their service lifetime, 28 to 60 ppm being the investigated hydrogen range.

The CCT specimens with the width dimension, W, of 17 mm (Figure 3) were cut from the pressure tube circumference, retaining the tube curvature, the direction of the crack propagation being axial on the plane with its normal parallel to the transverse direction of the tube. This sampling procedure takes into account the maximum stress direction of the pressure tube during its operation in the reactor, and is also the most likely plane of DHC occurrence. All the specimen dimensions are in the proportions recommended by the ASTM Test for Plane-Strain Fracture Toughness of Metallic Materials (ASTM E-399), [12], except for the thickness value, B, of 4.2 mm, which is the tube wall thickness. The machined notch was extended about 1.5 mm by fatigue to provide a sharp starter-crack.

The crack propagation experiments have been carried out in a creep loading system under constant load. The furnace attached to the testing device achieved the thermal schedule proposed for the test programme [13]:

• Heating at about 3 - 5°C/min to the peak temperature (to ensure the dissolution of hydrides), and soak for 1 hour;
• Cooling down to the test temperature and soaking half an hour before loading;
• Applying the load corresponding to a $K_I$ of about 15 MPa$\cdot$m$^{1/2}$;
• Removing the load after the crack length had extended by about 1.5 mm;
• Cooling the furnace slowly to room temperature to heat-tint the crack surfaces.

The hydrogen concentration was chosen so that TSS was just exceeded at the test temperature. As a function of the hydrogen concentration added to each specimen, four kinds of thermal schedules were applied for both types of pressure tubes samples:

• Room temperature to 315°C to 250 °C then DHC testing at 250°C and slow cooling for samples hydrided to 58 to 63 ppm hydrogen; 15 specimens of tube RX094 and 3 specimens of tube 429 were tested;
• Room temperature to 297°C to 227 °C then DHC testing at 227°C and slow cooling for samples hydrided to 43 to 48 ppm hydrogen; 10 specimens of tube RX094 and 8 specimens of tube 429 were tested;
• Room temperature to 285°C to 203 °C then DHC testing at 203°C and slow cooling for samples hydrided to 35 to 40 ppm hydrogen; 3 specimen of tube RX094 and 5 specimens of tube 429 were tested;
• Room temperature to 250°C to 144 °C then DHC testing at 144°C and slow cooling for samples hydrided to 28 ppm hydrogen; 6 specimens of tube RX094 were tested.

The crack initiation and propagation were monitored by means of the Potential Drop (PD) method, which is suitable for the CCT sample geometry [14]. The principle of the method consists of a disturbance of the electric field at the crack tip during its extension, causing a measurable redistribution of the electric force lines. This redistribution is reflected by a potential change across the specimen. Since the crack extension is linearly related to the electrical resistance change, the method permits knowing the crack length at any moment. The data acquisition and processing software, specially developed for this type of testing, allows the display of the current graphs of the measured variables: current, voltage, thermal schedule (Figure 4) and the crack length (Figure 5).

![Fig. 4. Typical thermal schedule for the DHC tests.](image1)

![Fig. 5. Potential drop and inferred crack length versus time](image2)
Cracking started after a short incubation time. Cracking time was taken as the difference between the duration of loading and the incubation time. The length of crack was evaluated as the area of DHC crack (Figure 6) divided by the specimen thickness. DHC crack velocity was calculated by dividing the crack length by the cracking time.

RESULTS AND COMMENTS

The mean axial crack velocities determined on both types of tested material at the four temperatures are summarized in Table 1.

Table 1. Summary of DHC Results: Average Crack Velocity (standard deviation) x 10\(^{-8}\) m/s

<table>
<thead>
<tr>
<th>Test temperature (°C)</th>
<th>Material</th>
<th>250</th>
<th>227</th>
<th>203</th>
<th>144</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Tube RX094</td>
<td>8.83 (0.62)</td>
<td>4.31 (0.49)</td>
<td>2.50 (0.25)</td>
<td>0.56 (0.025)</td>
</tr>
<tr>
<td></td>
<td>Tube 429</td>
<td>9.56 (0.34)</td>
<td>5.27 (0.56)</td>
<td>2.52 (0.31)</td>
<td>-</td>
</tr>
</tbody>
</table>

Although t-tests (at the 0.05 probability level) on the data from tests at 250°C and 227°C show that the values of crack velocity in each tube are not derived from the same population, the values do agree within 25%. Such a difference between tubes is considered to be low when compared with the usual range of values from nominally similar tubes which can be as high as a factor of four [9].

The DHC phenomenon is a strongly temperature-dependent mechanism, because the movement of hydrogen to the crack tip depends on the diffusion of hydrogen and the solubility limit, both of which are thermally activated processes. The relationship between crack velocity and temperature follows an Arrhenius dependence, i.e.:

\[ DHCV = C \cdot \exp\left(\frac{-Q}{RT}\right) \]

with \(C\) = experimental constant;
\(Q\) = the activation energy (J/mol);
\(T\) = absolute temperature (K);
\(R\) = universal gas constant, \(R = 8.314 \text{ J/mol/K}\).

The influence of temperature on the DHC crack velocity in both Zr-2.5Nb pressure tubes, using the experimental results presented above, is shown in Figure 7. The small difference between the temperature dependencies of DHC velocities of the two tubes reinforces their similarity. The analysis of the mechanical properties is in accordance with this conclusion. The two types of tubes are presenting similar values of the yield and tensile strengths, in the limits of the experimental errors range (Table 2).

Table 2. Summary of Mechanical Properties Results on Zr-2.5Nb tubes (250°C)

<table>
<thead>
<tr>
<th>Material</th>
<th>Mechanical Properties (MPa)</th>
<th>Transverse Tensile Strength</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Transverse Yield Strength</td>
<td>Mean Value</td>
</tr>
<tr>
<td>Tube RX094</td>
<td>567</td>
<td>16.5</td>
</tr>
<tr>
<td>Tube 429</td>
<td>585</td>
<td>12.3</td>
</tr>
</tbody>
</table>
The correlation of the DHC Velocity (DHCV) values and transverse yield strengths, used in Eq. (2) proposed by Oh [15], is a confirmation of our results for the present tubes showing similar tensile strengths and DHCV. The susceptibility to DHC cracking of Zr-2.5 Nb materials, hydrided at the same hydrogen level and tested in identical conditions is similar if the mechanical behaviour is identically.

\[ \text{DHCV} = c_1 D_H C_H \exp(c_2 \sigma_Y) \]  

where \( D_H \) is the hydrogen diffusivity, \( C_H \) the terminal solid solubility of hydrogen, \( \sigma_Y \) is the transverse yield strength, \( c_1 \) and \( c_2 \) are constants.

We thus conclude that tubes made from ingots melted twice have similar DHC velocities to those made from ingots melted four times.

The small difference between the DHC behaviour of the two pressure tubes, allows us to process the data together to calculate the activation energy of this phenomenon. To find an accurate value a statistical analysis was applied. The main goal of this analysis was to verify that the experimental dependence of DHC crack velocity as function of inverse temperature followed a linear model \( Y = A + BX \). To test the adequacy of the linear model, we calculate the maximum likelihood estimators of \( A \) and \( B \), we estimate the variance of the normal distribution, and determine the confidence intervals for \( A \) and \( B \) parameters and the confidence band for the entire median. The results of the statistical analysis are presented in Figure 8. The calculated value of the DHC activation energy for Zr-2.5Nb pressure tubes was about 48 kJ/mol, with interval limits between 41.5 and 54 kJ/mol (confidence level 95%). This result is in the range of previous values, although this range is wide: 42 to 72 kJ/mol [9].
CONCLUSIONS

1. A DHC testing procedure has been developed to measure with accuracy and reproducibility the axial crack velocities values induced in hydrided Zr-2.5Nb pressure tubes samples.
2. Tubes made from double melted and quadruple melted ingots had similar DHC crack velocities with similar temperature dependencies.
3. The activation energy of the DHC phenomenon was determined to be about 48 kJ/mol.

REFERENCES


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