

# INVESTIGATION OF DELAYED HYDRIDE CRACKING MECHANISM IN THE CANDU PRESSURE TUBE

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## ABSTRACT

The paper presents the testing methodology on the CANDU pressure tube specimens (Zr-2.5% Nb alloy) to determinate the fracture mechanics parameters. The crack initiation and subsequent DHC propagation tests were performed on compact-tension (CT) specimens prepared from pressure tube off-cuts. Previously, the CT specimens were subjected to the specific method to increase the hydrogen concentration up to 50 ppm. This process consists of two stages: the first one is the electrolytic disposal of a uniform zirconium hydride layer and, the second one is to apply a homogenization thermal treatment causing the diffusion of hydrogen into the zirconium matrix from the hydride surface layer. The testing temperature was around 280oC, which is the temperature of interest for the pressure tubes operating in CANDU 6 reactor.

**Key words: fracture mechanics parameters, KIH, CT specimens, DHC mechanism**

## Introduction

Zirconium alloys are used in nuclear reactors because of their combination of high strength, high corrosion resistance, and low neutron absorption cross-section. Their most demanding applications in nuclear reactors are as fuel cladding and in CANDU, RBMK, and other Pressurized Heavy Water (PHW) reactors as pressure tubes containing the fuel bundles. It is important for the safe and economic operation of these reactors that these components maintain their integrity throughout their design life. However, during their residence in the reactor these components are subject to aging mechanisms resulting from thermal - and pressure driven changes, fast neutron bombardment, and corrosion at the water/metal interface, the latter resulting in a small fraction of the released hydrogen produced during the corrosion reaction being absorbed in the zirconium alloy. When the hydrogen concentration in the material exceeds the Zr-H solvus composition, zirconium hydrides are formed. These hydrides, which are less ductile than the surrounding metal matrix, can have deleterious effects on the mechanical properties of these components when present at sufficiently high volume fraction. Their deleterious effects are enhanced by increases in yield strength and decreases in fracture toughness of the zirconium material. These changes are produced as a consequence of the production of dislocation loops and other microstructural changes during fast neutron bombardment in the reactor core [1].

The paper presents the crack initiation and subsequent DHC propagation tests performed on Zr-2.5% Nb alloys from CANDU pressure tubes. Its objective is focused on the experimental DHC tests and to develop the fracture mechanical testing methodology on the specimens cut from the back and front end of the pressure tubes from Cernavoda NPP. This methodology will be used for implementing the fracture mechanics tests in the Post-Irradiation Examination Laboratory on irradiated material cut from removed pressure tubes in Cernavoda NPP. Also, the experimental results will be used also to build-up a database in order to perform some structural integrity assessment activities based on the in-service inspection results.

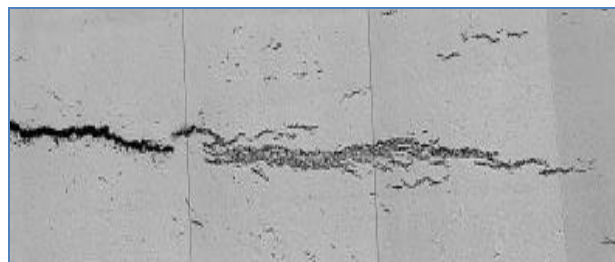
### Overview on the Delayed Hydride Cracking mechanism in the CANDU pressure tubes

Delayed Hydride Cracking (DHC) involved the accumulation of hydrides to some critical size at a region of elevated tensile stress, the fracture of this hydrided region up to its leading edge and the repetition of this process. In pressure tube material, DHC is largely limited to the radial-axial plane of the tube. Optical micrographs of arrested DHC cracks show that hydrided regions formed at a crack can be idealized as plate shaped, extending in the crack growth directions with thickness much smaller than their in-plane lengths. The hydrides platelets have an irregular aspect, darker in contrast with zirconium matrix (Fig. 1). Lying on the pressure tube's radial-axial plane these plate shaped (often tapered) hydrided regions are also referred to as radial hydrides. Observations of fracture surfaces at low magnifications often showed periodic rows of ridges (striations) extending in rows parallel to the crack front. The fracture surfaces between these striations were of brittle appearance, being flat with cleavage-like river patterns. The lengths between these striations were strongly temperature dependent.



**Fig 1.** Micrograph showing hydrides in Zr-2.5%Nb pressure tube

The interpretation of the experimental observations is that the hydrided region grows to some critical length from the crack tip, fracturing along its length up to its leading edge (Fig. 2). The fracture of the hydrided region causes an abrupt increase in length of the macroscopic crack, which is arrested at the leading edge of the hydrided region by the ductile matrix. The striations are the physical evidence of this arrest and the distance between each row of striations, called the striation spacing, represents the critical fracture length of the hydrided region while the striation length itself is associated with the plastic zone of the macrocrack at its point of arrest.

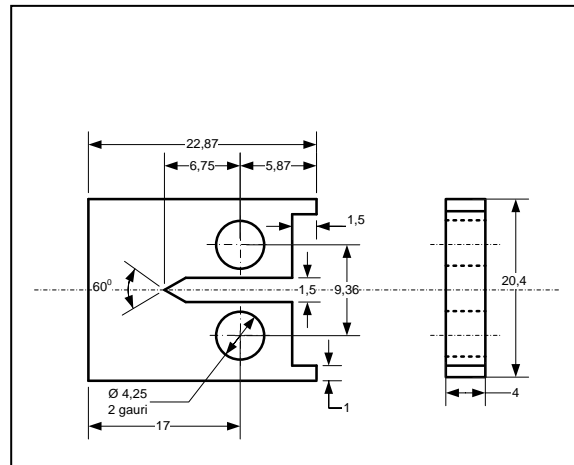


**Fig. 2.** Crack growth by DHC mechanism in Zr-2.5%Nb

The rate of growth of the DHC crack suggests that the process is diffusion driven. The presence of a ductile stretch zone (the striation) after each growth event suggests that the first step for continuation of the process is the nucleation of new radial hydrides. Although there may be, by chance, transverse hydrides located at the new location of the crack tip, these hydrides are not in favorable orientations to cause embrittlement of the crack front. The requirement for the nucleation of new, reoriented (radial) hydrides after each microgrowth step then means that the concentration of hydrogen at the flaw must be at a concentration that is at least as great as the solvus composition for nucleation of new radial hydrides [2].

## Experimental Activity

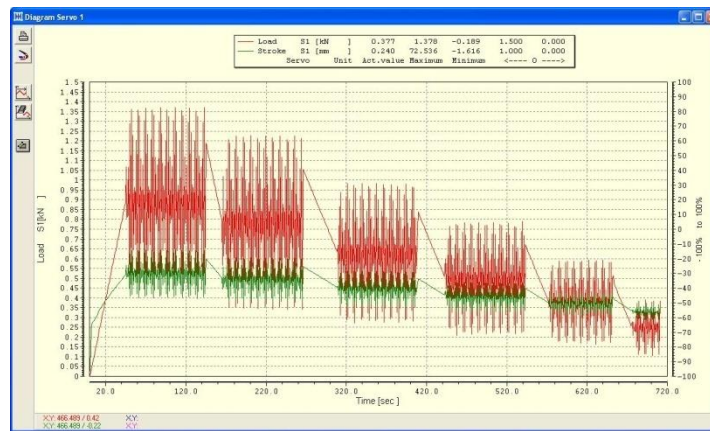
**Sample preparation.** The fracture mechanics tests were performed on CT (Compact Tension) samples cut from Zr-2.5% Nb pressure tubes. The geometry of the samples used in fracture mechanics tests, carried out to determine the parameter  $K_{IH}$ , must meet specific standards for fracture mechanics tests : ASTM E 399, ASTM E 647 , ASTM E 1820. The samples are obtained from the axial direction of the pressure tube. The sketch of the CT sample is shown in Figure 2.



**Fig. 2** The CT specimen for DHC test

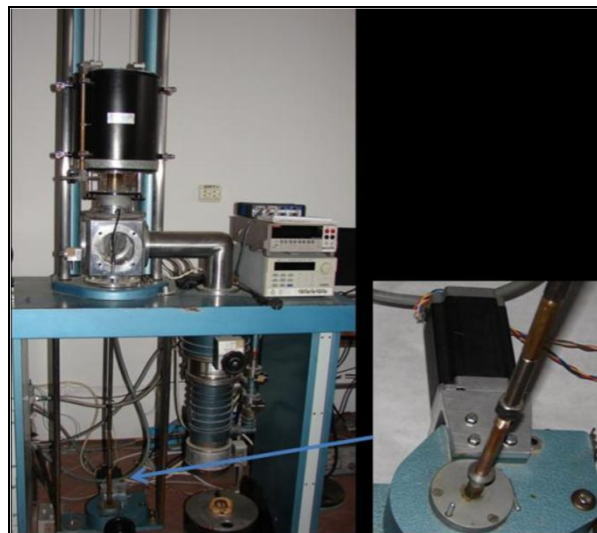
To obtain  $K_{IH}$  parameter on the CT specimen, one of the requirements is to have a small fatigue crack at the CT tip flaw. Thus the tip crack play the role of stress concentration factor and further will promote the hydride reorientation process during the test. Also, the fatigue crack constitutes the start point from which the cracking initiates through DHC mechanism, according to E-399 ASTM.

The pre-cracking process consists of cycling test on which the load has been gradually reduced while maintaining the stress ration is kept constant,  $R \cong 0.28$ . The fatigue pre-cracking process should be done carefully to diminish the probability of development plasticity at the crack tip.

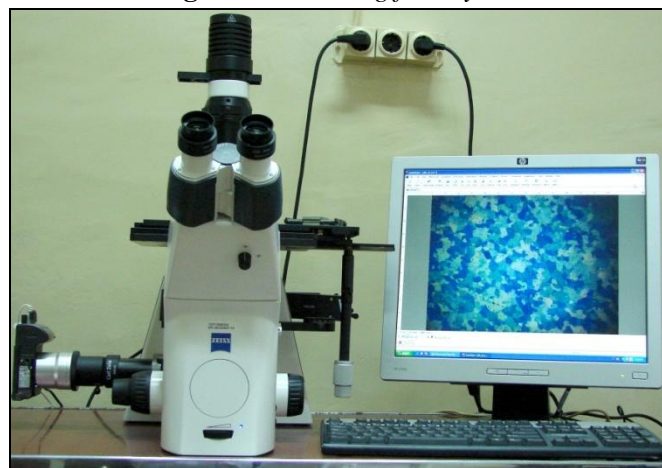


**Fig. 3** Stress versus time during pre-cracking fatigue process

**The testing and analyses facilities.** Figure 4 shows a photo with tensile creep machine where the experimental fracture tests were performed, in order to obtain the values of threshold stress intensity factor,  $K_{IH}$ , which characterize the initiation of DHC.



**Fig. 4** DHC testing facility



**Fig. 5** Optical microscope

The fracture surface and crack propagation measurements were performed by using metallographic examination with the optical microscopy (Fig. 5).

**The experimental methodology used for obtaining  $K_{IH}$  parameter**

The mechanical test to obtain the threshold stress intensity  $K_{IH}$  requires a constant load applied to the specimen simultaneous with a thermal cycling in a specific range. The load monitoring is realized by means of dedicated software developed at RATEN ICN Pitesti. This software is able to visualize and to adjust the testing parameters while the test is going on.

The initial applied load, before test starting, is to develop a stress intensity value of  $K_I=18.5 \text{ MPa}\sqrt{\text{m}}$ . At each step of  $50 \mu\text{m}$  crack advance, the software automatically decrease the load with 3% from previous load. This process is repeated during 24 hours, until the cracking is stopped. In this last stage, the test is stopped and the specimen is removed from the furnace in order to perform its examination. During the test the crack propagation is monitored by the potential drop (PD) method. A specimen ready for test is shown in Figure 6.



**Fig.6** The CT specimen wired for PD method

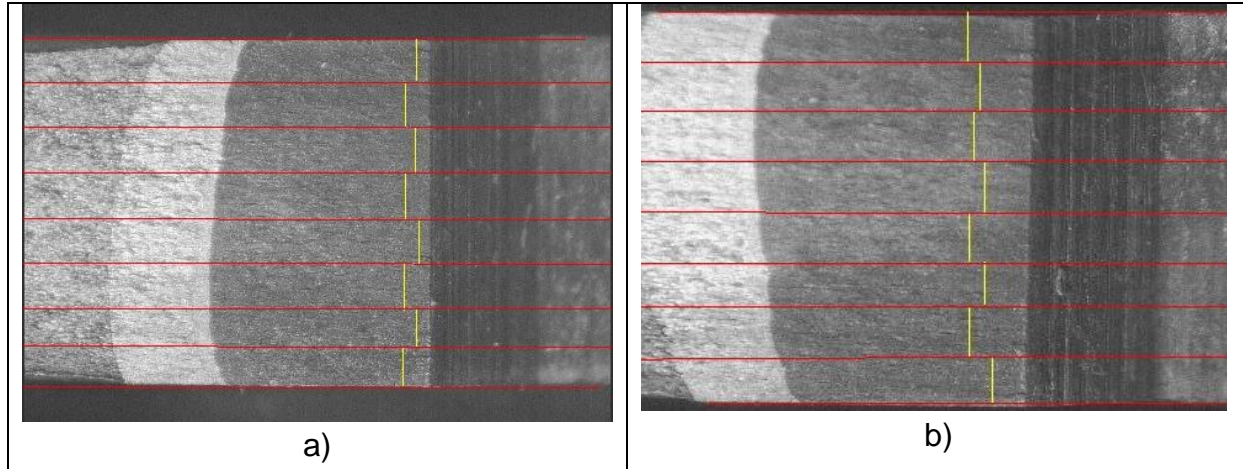
A window with parameters monitored during the test are displayed in Figure 7.



**Fig. 7** Windows with evolution of main parameters  $K_{IH}$  test



To infer the  $K_{IH}$  value, the last load step is considered, which the step is when the crack does not propagate for 24 hours. On the metallographic capture (Fig. 8) the crack length has been evaluated by means of the “nine segments method”. This allows obtaining the  $K_{IH}$  value according the E-399 ASTM.



**Fig. 8** Măsurarea propagării fisurii prin metoda „celor 9 segmente” pe proba B55: a) fața A a probei, b) fața B a probei

The stress intensity factor, used for K<sub>I</sub>H assessment is given by the following relationship:

$$K_I = \frac{P_Q}{B\sqrt{W}} f\left(\frac{a}{W}\right) \quad (1)$$

With

$$f\left(\frac{a}{W}\right) = \frac{\left(2 + \frac{a}{W}\right)}{\left(1 - \frac{a}{W}\right)^{\frac{3}{2}}} \left[ 0.866 + 4.64 \cdot \left(\frac{a}{W}\right) - 13.32 \cdot \left(\frac{a}{W}\right)^2 + 14.72 \cdot \left(\frac{a}{W}\right)^3 - 5.60 \cdot \left(\frac{a}{W}\right)^4 \right] \quad (2)$$

Here the meanings of involved parameters are:

$P_Q$ = applied load (N);

$B$  = specimen thickness (m);

$W$  = specimen width (m);

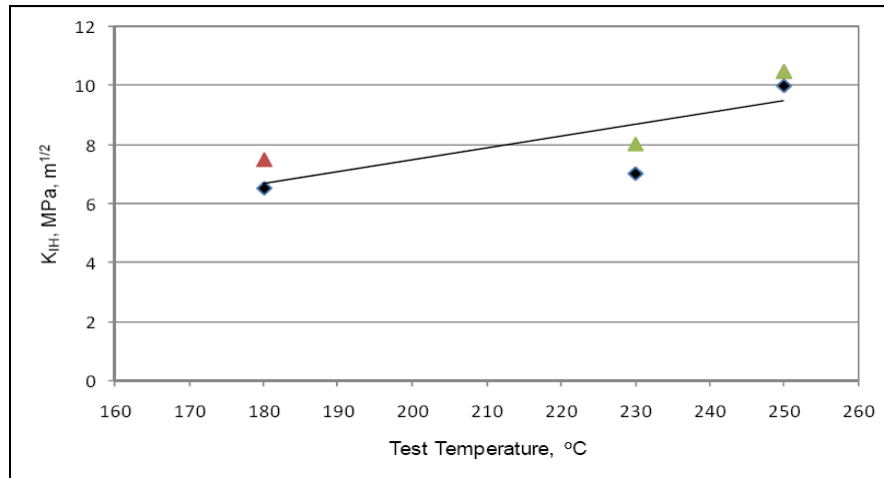
$a$  = crack length (m)

By using the above equations in present work two values of threshold stress intensity factor  $K_{IH}$  were obtained at 280 °C:  $K_{IH}=13.1 \text{ MPa}\cdot\sqrt{\text{m}}$  and  $K_{IH}=19.6 \text{ MPa}\cdot\sqrt{\text{m}}$ . For a temperature range between 180-250 °C, the reference [2] declared values displayed in Figure 9.

To obtain the J integral, the following relation is used:

$$J = \frac{K_I^2}{E} \quad (3)$$

with  $K_I$  given by equation (1), and E is Young modulus.



**Fig. 9** Values of  $K_{IH}$  versus temperature Zr-2,5%Nb [2]

One can see that the results from the present paper are in a rather good agreement with those mentioned in cited reference.

## Conclusions

The main paper outlines are:

- The short overview of Delayed Hydride Cracking (DHC) mechanism is given; DHC is the main damaging mechanism for the CANDU pressure tubes made from Zr-2.5%Nb;
- The experimental methodology to obtain the threshold stress intensity factor  $K_{IH}$ , which characterize the crack initiation by DHC in Zr-2.5%Nb alloy. In the present work two values of threshold stress intensity factor  $K_{IH}$  were obtained at 280 °C:  $K_{IH} = 13.1 \text{ MPa}\cdot\sqrt{\text{m}}$  and  $K_{IH} = 19.6 \text{ MPa}\cdot\sqrt{\text{m}}$ . The results are in a good agreement with those mentioned in the scientific literature;
- This methodology will be used for implementing the fracture mechanics tests in the Post-Irradiation Examination Laboratory on irradiated material cut from removed pressure tubes in Cernavoda NPP. Also, the experimental results will be used also to build-up a database in order to perform some structural integrity assessment activities based on the in-service inspection results.

## References

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- [2] IAEA-TECDOC-1410, Delayed hydride cracking in zirconium alloys in pressure tube nuclear reactors, Final report of coordinated research project 1998-2002, IAEA, October 2004