Development of in-situ Delayed Hydride Cracking tests using neutron imaging to study the H redistribution in Zr-2.5%Nb

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Development of in-situ Delayed Hydride Cracking tests using neutron imaging to study the H redistribution in Zr-2.5%Nb

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Abstract. Delayed Hydride Cracking (DHC) is a failure mechanism that occurs in Zr alloys under certain conditions of hydrogen concentration, temperature and stress gradient. In service, hydrogen produced by corrosion reaction can be incorporated in Zr alloys and if the solid solubility is exceeded, hydrogen precipitates as zirconium hydride. The presence of a stress concentrator, such as a crack, generates the hydrogen diffusion and precipitation to the high stress zone beginning the DHC process. In this work, in-situ DHC tests in air at 250°C were performed at ANTARES, the neutron imaging facility of the FRM-II reactor. Samples of Zr2.5%Nb produced from extruded tubes and pressure tubes were studied using a stress rig specially modified to perform DHC tests in the neutron beam. H redistribution during mechanical testing was followed in-situ by registering the changes in neutron transmission. The results were compared with the images obtained by light optical microscopy after the tests. The results highlight the capabilities of neutron imaging to analyze time-dependent H distribution during DHC crack growth.

1. Introduction

Zr alloys are widely used in the nuclear industry due to the combinations of excellent mechanical properties, corrosion resistance and low neutron absorption cross-section for internal structural components of nuclear power reactors. In particular, Zr alloys that contain Nb in a concentration range from 0.5 to 2.5% (wt. %) exhibit the best corrosion performance for high burn-up fuel claddings [1]. The CANDU reactor (CANadian Deuterium Uranium) is a Pressurized Heavy Water Reactor in which the large steel pressure vessel found in the light-water reactor is replaced by individual Pressure Tubes (PT) that contains the fuel bundle. PTs are made of the Zr-2.5%Nb (wt. %) alloy. During the service of the nuclear power plant, PTs are in contact with heavy water at high temperatures (250-300°C) and pressures (HTHP) in an environment of intense neutron radiation.

In service, the structural integrity of the Zr-2.5%Nb pressure tube may be affected by a steady pick-up of deuterium due to a corrosion reaction [2] that modifies the mechanical properties by reducing the ductility and crack-grown resistance. As deuterium (D) is chemically equivalent to hydrogen (H), under certain thermo-mechanical conditions, H and D incorporation may lead to called delayed hydride cracking (DHC).

DHC is a degradation mechanism consisting of an accelerated crack propagation that originates from the precipitation and reorientation of brittle hydrides in front of a stress concentrator such as a crack or a flaw in the material surface [3]. Brittle hydride platelets precipitate normal to tensile stress when the local terminal solid solubility (TSS) is exceeded [4]. Crack growth occurs when hydride platelets formed in front of the crack tip reach a critical length. Under load, the stress gradient produced by the crack is the driving force for H diffusion to the crack tip. The accumulation of hydrides ahead of the crack tip during a DHC test, due to H diffusion and precipitation, has not been studied experimentally. As neutrons are strongly attenuated by hydride materials, neutron imaging is an adequate technique to follow H redistribution during DHC tests.

In the present work, H redistribution and accumulation near the crack tip during DHC tests in air at 250°C in two different Zr-2.5%Nb materials (extruded tubes and PTs) was studied by in-situ neutron imaging. For this purpose, a dedicated test rig was developed to perform the DHC test on the neutron beam, with the requirement of the test specimen being as close as possible to the neutron detector, to improve spatial resolution. Additionally, the degree of hydride re-orientation was assessed by optical microscopy after the DHC tests.

2. Experimental method

DHC tests use cantilever beam (CB) samples obtained from PTs. CB samples are notched and then pre-cracked by fatigue at the notch to form the initial crack. The tests are usually carried out in air at 250-350°C, with the main parameters being the velocity of crack propagation (v) and the stress intensity factor at the crack tip (K_I) [3]. Two CB specimens were produced respectively from a Zr2.5%Nb extruded tube produced by Ati Wah Chang and a PT produced from a similar extruded tube in Ezeiza Atomic Centre for the life extension project of a CANDU nuclear power plant at Embalse, Argentina. CBs specimens of 3 mm thickness and 60 mm length are typically used in the DHC test [5] and were produced in PPFAE-CNEA by machining, being the width of the samples determined by the wall thickness of each tube used, ~6mm for extruded tubes and ~4mm for PT.

Before the notched and pre-cracked, H was incorporated into the specimens by cathodic charge technique in 0.1 M H_2SO_4 aqueous solution at 80°C for 24 h followed by a homogenization treatment. After that, samples had H concentrations of 70±10 wt. ppm consisting of circumferential hydrides and H in a solid solution. An increase of 5 wt. ppm in H content in α-Zr, when precipitated, corresponds to a change of the hydride volume fraction of 0.0003. For a sample of 3 mm thickness, this correspond to a change in neutron transmission in the order 0.0003 [6].

Typical DHC tests are performed within a tubular furnace, whilst the bending moment is applied by a long arm loading [5]. Here, we designed a dedicated DHC test rig (Figure 1) in order to obtain a small device capable of being mounted in the tensile machine that was available at the ANTARES instrument [7]. This test rig was developed by the *Departamento de Tecnología del Circonio* (DTC) in CNEA, Argentina. The design converts the tensile load P indicated in Figure 1 into a four-point bending moment M due to the rotation of the upper and lower grip on the corresponding rotation pin, converting axial stress to circumferential stress conditions on the notch. The upper and lower grips of the samples have two cylindrical heaters cartridges (50 W electrical power, 5 mm diameter, 35 mm length), and their temperature was measured by resistance temperature detectors Pt 1000 connected to a Lake Shore 340 thermal control unit. The initial free space between the grips was 10 mm, with the notch located in the centre of the neutron image. The test rig was mounted on the tensile machine using extension bars of 150 mm length to avoid the heating of the load cell and crosshead (Figure 2). The maximum distance between the sample and the scintillator plate was 20 mm obtaining a pixel size of ~45 µm. Grips were made of aluminium while the cover plate of the sample and pins were machined from Zr2.5%Nb alloy. These materials were selected to minimize activation of the test rig after the measurements.

The test procedure consisted of a thermal treatment where samples were heated at 300°C with 10°C/min, maintained at this temperature for 30 min, and then cooled to 250°C at 5°C/min when it was maintained until the end of the test. Before applying a load, the sample was kept for 30 min to homogenize the temperature on the sample and grips. After that, loads of 90 N and 80 N were applied to the extruded tubes and PTs, respectively, fixing the crosshead in that position until the end of the test. The heating and loading procedures were adapted in these experiments to the availability of beam time, and different times were used for the two specimens based on previous experience [5].

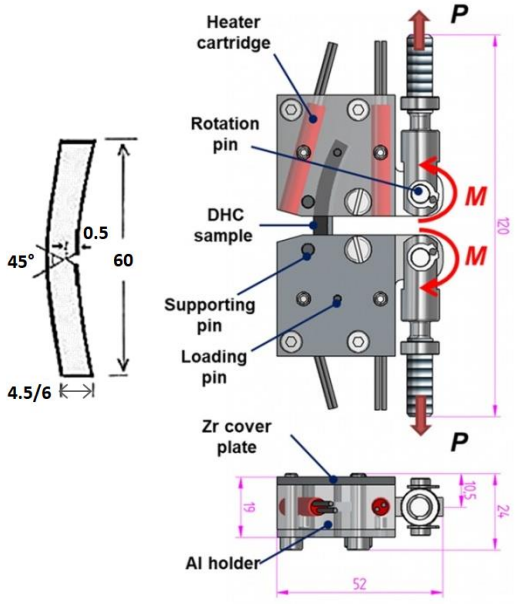

Figure 1. Scheme of the DHC specimen used (dimensions in mm) and the test-rig developed by the DTC, CNEA, Argentina.

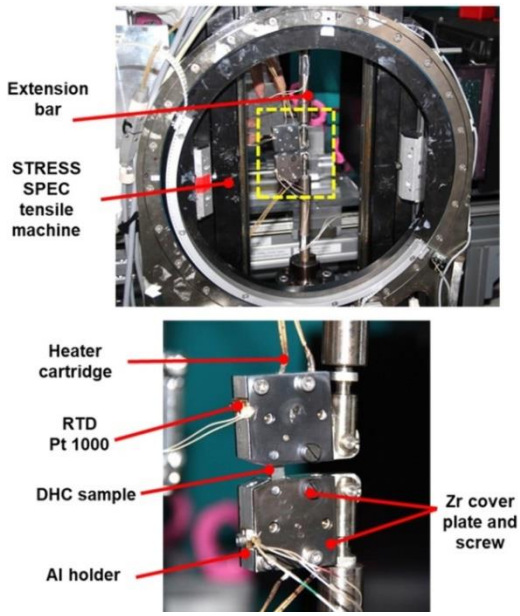

Figure 2. Experimental arrangement mounted in the DHC in-situ test on the neutron beam.

Neutron imaging (NI) experiments were performed at ANTARES, the neutron imaging facility of the FRM II reactor [8]. The samples were analysed using a collimation ratio L/D of 500. Images were acquired using a Gd_2O_2S scintillator plate of 20 µm thickness with an acquisition time of 30 s by an ANDOR cooled CCD camera with field-of-view (FOV) of 2048 x 2048 pixels, 16 bit. Images were

averaged in stacks of 30 images obtaining a time step of 15 min. Before that, the images were corrected considering the flat field and the dark current of the neutron detector systems. The experimental arrangement is shown in Figure 2.

The time evolution of H content within the samples was evaluated by following the changes in the neutron transmission T, obtained after performing flat and dark current corrections to the images. The change in H content at different ROIs (regions of interest) of the specimen was estimated using a previous calibration obtained by Buitrago *et al.* [5] for the ANTARES facility, as the same experimental setup was used for the present experiments [9]. It is important to note that a recent work by Al-Falahat *et al.* [10] found that changes in the temperature modified the neutron transmission in crystalline samples. An increase in the temperature generates a decrease in the elastically scattered intensities together an increase in the inelastically scattered intensities that can affect the experiments of energy-selective neutron imaging. In-situ DHC experiments were performed without a selective wavelength, allowing the use of the calibration curve determined at room temperature [8]. Moreover, the neutron transmission evolution was studied at the same temperature (250°C) using a beam monitor outside the specimen close to the notch.

3. Results and discussion

3.1. Extruded tube

A load of 90 N was applied to the specimen at 250°C, and images were taken for 4 hrs whilst the change in the load was monitored until the load was released. Figures 3(a) and (b) show the optical micrograph of the sample before and after the test, respectively, where the crack growth occurred during the test is visible. The detail of the crack near its tip shown in Figure 3(c) probes that the crack has indeed grown by DHC, as its direction is always perpendicular to the circumferential hydrides, indicating that it propagated along stress-reoriented radial hydrides.

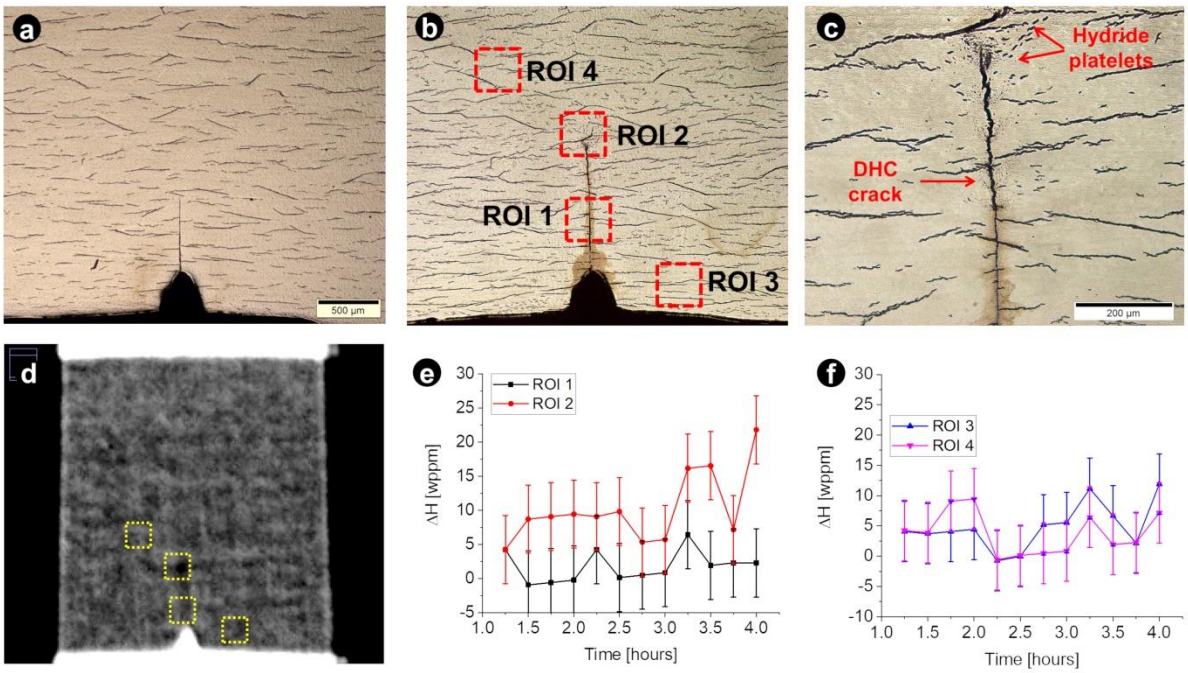

Figure 3. Results on extruded tubes sample: Optical micrographs (a) before the test, (b) after the test, (c) detail of the crack tip after the test, (d) neutron image at the end of the test, (e) and (f) change in H content during the test in the ROIs identified in (b) and shown in (d).

Figure 3(d) shows the neutron transmission image of the sample after the test. The pattern observed within the samples reflects the changes in neutron transmission due to the presence of the circumferential hydrides that cannot be individually resolved by the neutron images. That is related to the actual spatial resolution of the setup (~50 μm) and the attenuation observed in each pixel gauges a 3 mm thickness of material along the beam, hence traversing several hydrides. However, it must be noted that the pattern is mostly horizontal, mimicking the direction of the circumferential hydrides observed in Figures 3(a), (b), and (c).

Four ROIs of 315 x 315 μm^2 were defined to follow their H content during the test, as shown by the dotted squares in Figures 3(b) and (d). The use of ROIs ies required to achieve the sound statistics required to resolve the small changes expected in the transmission. ROI1 is located in the pre-crack region and ROI2 in the final crack tip, whilst ROI3 and ROI4 are relatively far from the crack. Figures 3(e) and 3(f) show the evolution of H content evolution. We observe that whilst ROI1, ROI2 and ROI3 display nearly constant values of H content, ROI2 at the crack tip, presents a significant increase of ~12 wt. ppm after 4 hrs, and a rising trend is observed among the statistical noise. Error bars presented in Figs. 3(e) and (f) correspond to the uncertainty in the H content found in the determination of the calibration curve [6]. Although this amount is close to the minimum detection signal in the present experiment, this is confirmed by a relatively large black dot observed in ROI2 in the neutron image of Figure 3(d), and by a cluster of small hydride platelets in the optical micrograph of Figure 3(c).

To compare the observed increase in H content at the crack tip, we mention that Colldeweih *et al.* [11] perform neutron imaging experiments to characterize Zircaloy-2 samples obtained from fuel cladding, after DHC tests using a 3-point bending test at 360°C during 4 hrs. They used a high-resolution setup (neutron microscope) on a 2 mm thick specimen containing the notched region. For samples containing ~200 wt. ppm H, they observed an increase in H in the range of 100-500 wt. ppm in a region 120 μm away from the crack tip. In the present work in Zr-2.5%Nb, we have found a smaller H content but averaged over a larger volume. On the other hand, Tondro & Abdolvand [12] simulated H transport near a V notch tip during DHC at 330°C using a coupled diffusion–CPFE model (crystal plasticity finite element) for the α-phase of Zr-2.5Nb, for an initial homogeneous content of 100 wt. ppm using the typical texture of PT. They found average H increases in the crack tip in the order of 10 wt. ppm.

3.2. Pressure tube (PT)

A load of 80 N was applied to the specimen at 250°C, and images were taken for 13 hrs whilst the change in the load was monitored until the load was released. Figure 4(a) shows a neutron image obtained after the in-situ test, whilst Figure 4(b) shows an optical micrograph of the region identified by the yellow dotted line, where it becomes clear that crack growth in this specimen has been more complex than in the previous case. Two 270x270 μm^2 ROIs are identified in both images: a blue dotted ROI in the pre-cracked region, and a red dotted ROI by the crack tip observed at the end of the test, whilst the black dotted square outside the specimen identifies the region used as a monitor. Figure 4(c) shows the time evolution of the neutron transmission for the two ROIs. A steady increase in the transmission is observed for the pre-crack ROI (blue dots), which may be due to a small opening of the crack during the test. On the other hand, a small reduction in transmission is observed for the crack-tip ROI, which is related to an increase in H content of ~8 wt. ppm, as observed in the red ROI of Figure 4(b).

Figure 5 shows additional details about the test of the PT specimen. Figure 5(a) shows the evolution of the load (blue line) and temperature (red line) during the test. Whilst the temperature remains extremely constant after application of the load (at 0 hs), as the crack grows, a steady drop in the load is observed in the registered load. It must be mentioned that although the applied load was lower than in the previous case, the stress intensity factor at the crack tip was larger due to the smaller thickness of this specimen (4 mm vs. 6 mm). Because of this higher load, at specific times during the

test, the crack grew along circumferential hydrides, as indicated by the numbers in Figures 5(a) and (c), rather than along radial hydrides as required by the DHC process. In the load, this manifested as small increases in the overall decaying curve.

On the other hand, the increase in H content observed at crack tip ROI was fully confirmed by the optical micrograph shown in Figure 5(d), where small re-oriented hydrides ahead of the crack tip are seen.

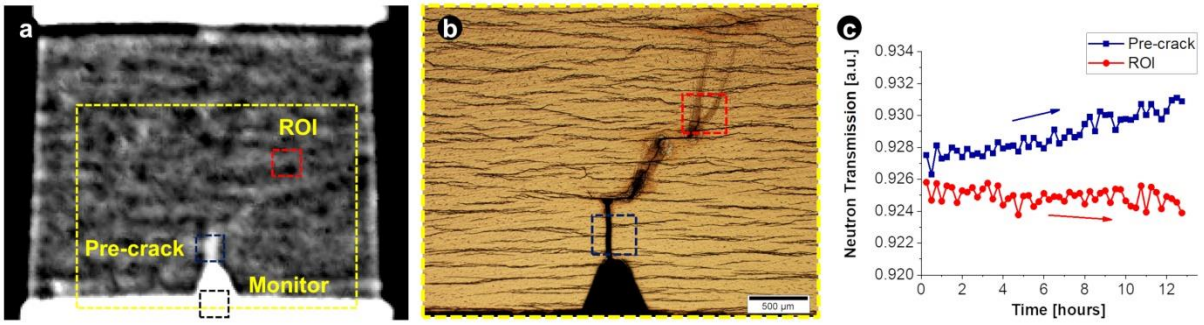

Figure 4. Results on PT samples: (a) Neutron transmission of the samples to finish the test, (b) optical micrograph before the test, (c) time evolution of the neutron transmission in the zone labelled in (b).

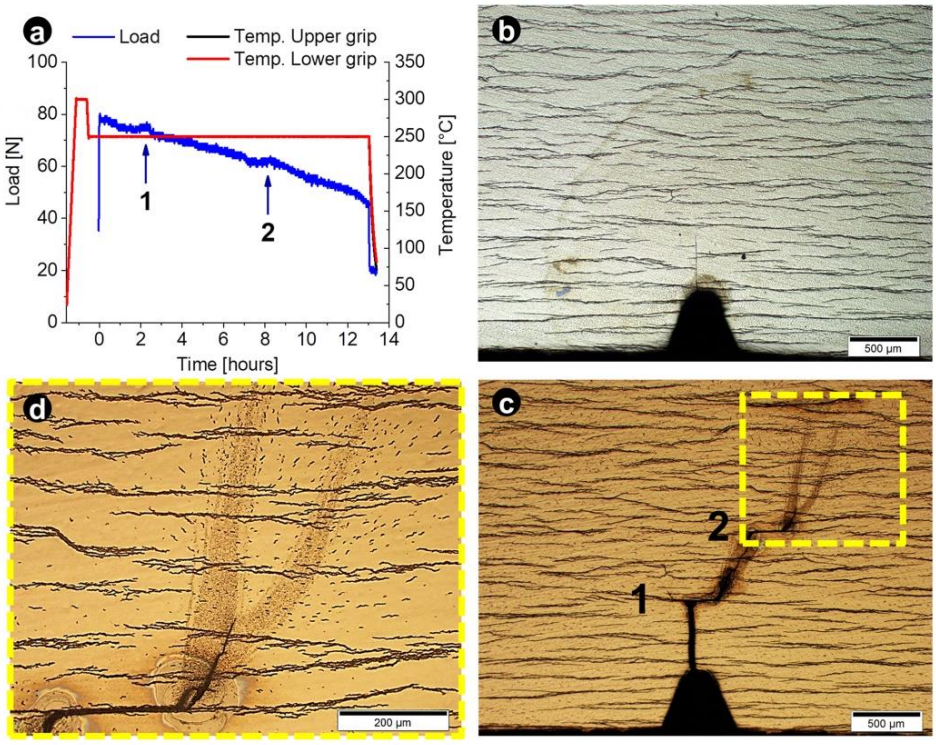

Figure 5. Results on PT sample: (a) Experimental temperature and load curves, (b) optical micrograph (b) before and (c) to finish the test. (d) Detail of the crack tip after the test where small hydride platelets precipitated.

4. Conclusions

In the present work, in-situ neutron imaging experiments during Delayed Hydride Cracking tests at 250°C on cantilever specimens produced from Zr2.5%Nb tubes were performed. For this, a specially designed testing device was developed that allows converting a tensile load produced by a stress rig in a bending moment on the samples, needed to initiate the DHC mechanism. The results show that neutron imaging techniques have the sensitivity to follow the changes occurring in H concentration at the crack tip during a DHC test. The increase of H at the crack tip was in the order of 10-15 wt. ppm, close to the sensitivity of the technique due to the small thickness of the sample used (3 mm).

The choice of 3 mm was adopted because is normally used in the DHC studies on PTs. An increase in the sample thickness will improve the H sensitivity without compromising the mechanical requirements of the DHC test. Hence, specimens of at least 6 mm thickness are recommended for future DHC experiments at neutron imaging beamlines.

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