DEVELOPMENT OF THE HYDRIDE CONTINUITY COEFFICIENT METRIC TO CHARACTERIZE ZIRCONIUM HYDRIDES AT FLAWS IN CANDU PRESSURE TUBE

ALEXANDRA JINGA[1], VASILE RADU[1], MARIOARA ABRUDEANU[2], DENISA TOMA[1], MIRELA MATEI[1], VALENTIN OLARU[1], AMALIA POPA[1]

[1] Institute for Nuclear Research (RATEN ICN) Pitești, Romania
[2] Nat. Univ. of Science and Techn. Politechnica Bucharest, Pitesti University Center, Romania
e-mail: alexandra.jinga@nuclear.ro

Abstract

The consequences of hydrogen absorption in CANDU pressure tubes, Zr-2.5%Nb alloy lead to complex phenomena. Therefore, the evaluation of the formation of hydride platelets requires an adequate analysis. The samples, made from Zr-2.5%Nb alloy, were subjected to thermal treatments for hydrogen absorption and hydride precipitation, observing the influence of thermo-mechanical parameters on the reoriented hydrides morphology near the mechanical stress concentrators, which develop at the tip of volumetric flaw. The finite element method has been used to analyze the field of mechanical stresses and strains developed in the area of blunt flaws. The parameters that determine the reorientation of the localized hydrides were analyzed: the mechanical stress intensity factor K_I , the mechanical stress concentration factor K_C , the root radius of the flaw, and the flaw depth. To fully characterize the microstructural morphology of the zirconium hydrides in the material, the "Hydride Continuity Coefficient (HCC)" metric was developed.

Keywords: *Zr hydrides, Hydride reorientation, Zr-2.5%Nb alloy, HCC metric, FEA analysis*

Introduction

For CANDU pressure tubes, Zr-2.5%Nb alloy, when the limit at a certain given temperature, the Zr-2.5%Nb alloy precipitates in the form of zirconium hydride (ZrH_2), which behaves brittle in the ductile matrix of the alloy. In situations where no external mechanical stress is applied, or at very low mechanical stress, ZrH_2 platelets precipitate and align in the axial-transversal plane of the pressure tube. These hydrides are referred to as "circumferential hydrides" and their behaviour does not greatly affect the pressure tube stress resistance under normal circumferential mechanical stress in operation. If the mechanical stresses are high enough, exceeding a certain threshold, then the zirconium hydride platelets can reorient in the radial-axial plane, perpendicular to the direction of mechanical stress application. These hydrides are called "reoriented hydrides" and their presence may involve failure mechanisms because they produce a decrease in the ductility as well as the fracture toughness of the pressure tube.

Experimental programs were thus started to determine the conditions for DHC initiation from volumetric defects with different tip depths and radii, hydrogen content, thermal cycles and pressure tube material conditions in the CANDU reactor [1-6]. Overall, there is insufficient data on the fracture toughness models of the Zr-2.5%Nb material to reproduce its dependence on hydrogen (H_2) concentration and hydride morphology, especially for high hydrogen concentrations and hydride morphology specific to some realized conditions of CANDU reactor operation.

This fact requires an extensive research program to obtain additional information and to understand the fracture toughness of cold-worked, irradiated and non-irradiated Zr-2.5%Nb material, as a function of H_2 concentration and hydride morphology, to be representative of operating conditions in the reactor.

This paper presents some experimental tests that followed the phenomenon of the formation of hydride platelets in the matrix of the Zr-2.5%Nb alloy, their reorientation at the volumetric flaw tip placed on the dog-bone type samples, which were subjected to a specific thermal treatments for H_2 absorption and hydride precipitation. For the analysis of the mechanical stresses developed at the flaw tip in the dog-bone sample, the finite element method was used to analyze the field of stresses and mechanical deformations developed in the area of defects on pressure tube specimens. The specimens were obtained from rings cut in the circumferential direction of the CANDU pressure tube, with different hydrogen concentrations, undergoing initial heat treatment under constant load to achieve hydride reorientation at the tip of the flaw.

In this work, the Hydride Continuity Coefficient (HCC) metric was used, being the parameters that influence the HCC values for complex flaws of the "Bearing Pad Fretting Flaws" and "Debris Fretting Flaws" type. The results were processed according to the methodology from the Canadian Standard CAN-CSA N285.8. [7].

Metallographic analysis and Scanning Electron Microscopy (SEM) complete the characterization of Zr-2.5% Nb specimens subjected to experimental tensile tests. HCC metric characterization is completed regarding the phenomenon of reorientation of the ZrH_2 located in Zr-2.5%Nb alloy.

Quantification of hydride morphology to analyze the phenomenon of reorientation of localized hydrides

To analyze the reorientation phenomenon of the hydrides located on Zr-2.5%Nb alloy samples, hydrided and subjected to a thermal cycling treatment under constant mechanical load, micrographs were made on the samples in the radial cross-section. The hydrides are visualized as small filaments (Fig. 1), having different orientations in the radial-transversal plane.

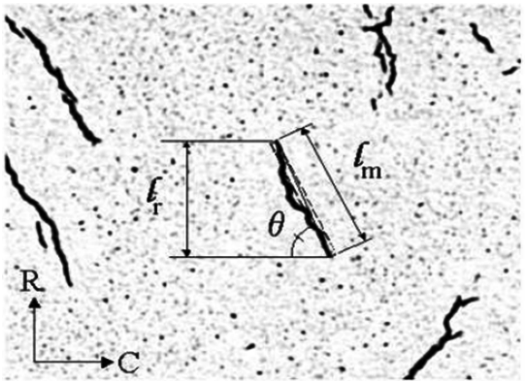


Fig. 1. The orientation of the hydride plates in the Zr-2.5%Nb alloy in the radial-transverse section [2]

The hydride plates "reoriented", following the heat treatment, are those hydride plates whose direction of orientation is located at an angle that varies from the transverse direction $\theta \in (45° - 90°)$, as defined in Fig. 1.

In the situation where a more precise quantification of the reorientation effect of the hydrides in the Zr-2.5%Nb alloy is desired, a dimensionless quantity is defined, called the Hydride Continuity Coefficient (HCC), based on analysis of the micrographs in which are highlighted hydride plates. In short, the stages of determining HCC are:

- Choose an analysis surface in the shape of a rectangle, with dimensions of approximately *0.1 mm x 1.0 mm*, its orientation being in the radial direction;
- Inside this rectangle, the length of the hydride plates is projected in the radial direction;
- Read the projected lengths L_k of each projection in the radial direction (i.e. the length of the rectangular area), as well as the total length of the examined rectangle L_{total};
- The HCC coefficient is calculated with the relation:

$$HCC = \frac{\sum_{k=1}^{n} L_k}{L_{total}} \quad (1)$$

It should be noted that there are some restrictions when evaluating L_k projections in the radial direction, namely:

a) very small segments (order of microns) are eliminated;

b) segments overlapping in the examined area are considered only once, by the common length of the projection in the radial direction.

In the literature [8], several parameters have been defined to measure/characterize the morphology of zirconium hydrides in the material volume (also called metrics) to describe its effect on the initiation and propagation of cracks.

Experimental Activity

The experimental tensile tests were performed on the hydride specimens at different hydrogen concentrations to evaluate the mechanical properties of the Zr-2.5% Nb alloy. Several steps were followed to perform the tests.

*Step 1.*The specimens were prepared in a typical "dog-bone" shape from the circumferential direction of the CANDU pressure tube, Zr-2.5% Nb alloy (Fig. 2).



Fig. 2. The "Dog-bone specimen"

Step 2. The specimens were subjected to thermal treatment of localized hydrides under constant mechanical stress in the concentration range 40-80 ppm, corresponding to the pressure tube conditions of operation in the reactor (Fig. 3).

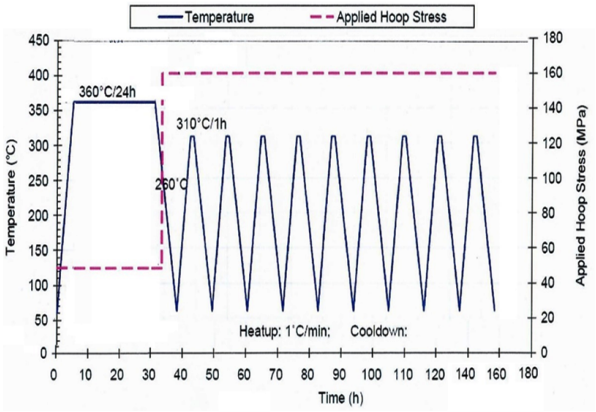


Fig. 3. Thermal scenario applied for hydride reorientation on dog-bone samples

Step 3. After the stage of hydrogen content determination, the experimental investigations are based on the tensile tests. These are usually used to evaluate the mechanical properties of the Zr-2.5% Nb alloy using the Walter+Bai mechanical test facility (Fig. 4) at room temperature.

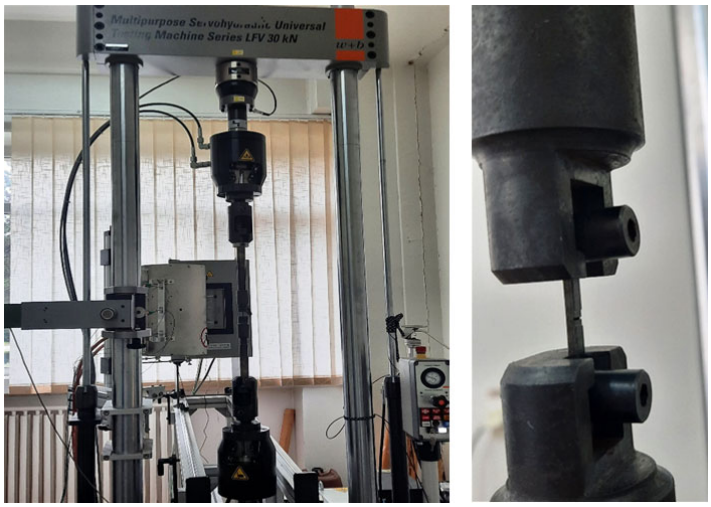


Fig. 4. Walter+Bai installation (left); Detail of the sample mounted for testing (right)

After tensile testing, the specimens were examined by SEM and optical microscopy to identify the fracture characteristics in the localized hydride reorientation area.

Analysis of the state of mechanical stress-strain state in the dog-bone sample by the finite element method

To determine the field of mechanical stresses developed at the top of the flaws practised on the dog-bone sample, the VisualFEA software (based on finite element method) was used. To achieve above mentioned goal, the project sketch of the sample in Fig. 1 was considered.

In the case of the thermal treatment applied to obtain the phenomenon of reorientation of the hydrides located at the tip of the complex defect practised on the surface of the dog-bone sample, the applied load was constant. The nominal mechanical stress generated in the sample by this constant stress was $\sigma_n = 62.5$ MPa.

Figure 5 shows the principal stress field σ_{1p}, developed at the top of the flaw with a depth of 0.5 mm and a radius of $\rho = 0.05$ mm (at the top), which determines the reorientation of the hydrides at the top of the volumetric flaw.

The figure also shows a detail of the mechanical stress field limited to the area adjacent/near the tip of the flaw with a depth of 0.5 mm and the profile of the principal stress σ_{1p} dependence on the distance measured from the tip of the flaw. It can be seen that stress values higher than 120 MPa are found in a 1.5 mm zone measured from the top of the defect in the transverse direction of the sample, the zone that will constitute the area where the hydrides will reorient during the heat treatment. It should be noted that in the specialized literature, the value of 120 MPa is specified to achieve ZrH_2 reorientation in a Zr alloy.

Figure 6 shows the main stress field σ_{1p}, developed at the top of the flaw with a depth of 0.7 mm, which determines the reorientation of the hydrides located at the tip of the volumetric flaw.

To study the influence of the ZrH_2 located at the tip of the artificial flaws practised on the dog-bone samples, after obtaining the reorientation effect, tensile tests according to ASTM E3 [9] at room temperature, were also carried out. After tensile testing, the samples were examined by SEM and optical microscopy to identify the fracture characteristics in the area of localized hydride reorientation.

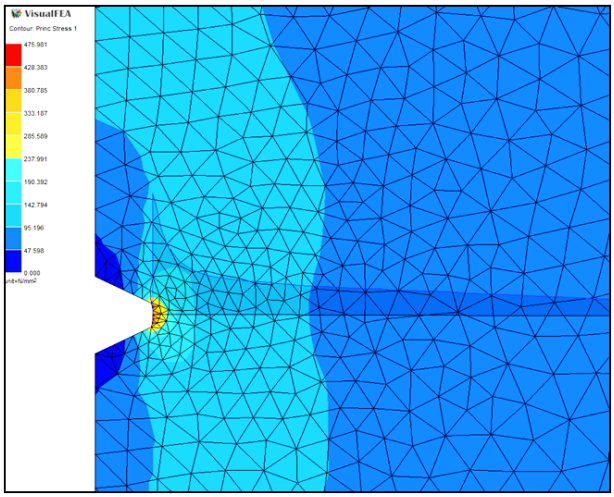


Fig. 5. Detail of the principal stress field limited to area adjacent/near to the tip of the flaw (0.5 mm depth, ρ=0.05 mm)

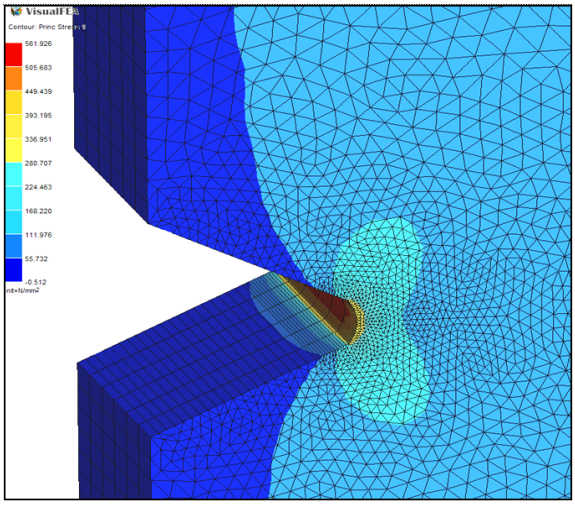


Fig. 6. Detail of the principal stress field limited to area adjacent/near to the tip of the flaw (0.7 mm depth, ρ=0.05 mm)

Results and discussion

To determine the intensity factor of the mechanical stress K_I, in the dog-bone sample, a calculation relationship of the influence function from the reference [7] was used. In this case, the stylized sample sketch from reference [7], shown in Fig. 7, will be considered.

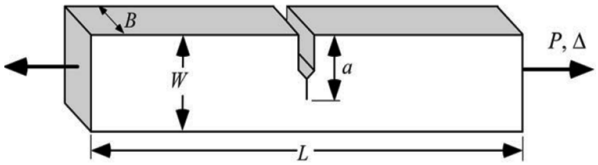


Fig. 7. Stylized sketch of the dog-bone sample for determining the mechanical stress intensity factor K_I

The calculation relation, in this case, for the mechanical stress intensity factor, assimilating the volume defect as an equivalent sharp defect is:

$$K_I = \frac{P}{B\sqrt{W}} f\left(\frac{a}{W}\right) \tag{2}$$

where

$$f\left(\frac{a}{W}\right) = \sqrt{\pi \frac{a}{W}} f\left(\frac{a}{W}\right) = \frac{\sqrt{2\tan\frac{\pi a}{2W}}}{\cos\frac{\pi a}{2W}} \left[0.752 + 2.02\frac{a}{W} + 0.37\left(1 - \sin\frac{\pi a}{2W}\right)\right] \tag{3}$$

with: P – applied traction force; B – sample width; W – sample thickness; a – the depth of the defect.

For the two depths of the defect and at the applied constant mechanical load, the following values are obtained:

- $a = 0.5$ mm

$$K_{I1} = 3.20 MPa \cdot \sqrt{m} \tag{4}$$

- $a = 0.7$ mm

$$K_{I2} = 4.24 MPa \cdot \sqrt{m} \tag{5}$$

The HCC metric for the two defects was analyzed according to the values obtained for the mechanical stress intensity factor K_I. The fracture surface and crack propagation measurements were performed by using the metallographic examination (Fig. 8), along with the HCC metric evaluation area.

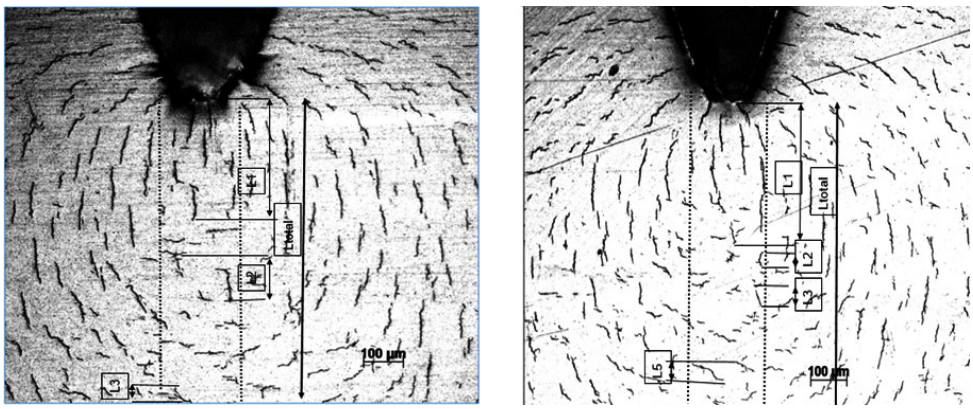


Fig. 8. Metallographic examination of the specimen surface after test:
(left) H_2 concentration 60 ppm, a=0.5 mm, ρ=0.05 mm; (right) H_2 concentration 67 ppm, a=0.7 mm, ρ=0.05 mm

As a first observation, the size of the circular zone, generated at the top of the flaw, where the reoriented hydrides are concentrated, has a size several times larger than that of the process zone.

In the paper, the parameters determining the reorientation of localized hydrides (the mechanical stress intensity factor K_I, the mechanical stress concentration factor K_C, the tip radius, and the depth of the flaw) were analyzed to fully characterize the HCC metric, which defines the microstructural morphology of hydrides in the material.

This analysis resulted in the following:
- For the samples with complex defects, concentration of 40-60 ppm, a=0.5 mm, ρ=0.05 mm, at the parameters K_{I1}=3.20MPa·√m, K_{C1}=7.5, we found HCC= 0.61;
- For the samples with complex defect, concentration of 39-67 ppm, a=0.7 mm, ρ=0.05 mm, at the parameters K_{I2}=4.24MPa·√m, K_{C2}=8.9, we found HCC= 0.66.

It should be mentioned that the initiation of DHC-type cracking occurs at the top of the process zone, and in a subsequent step, the crack propagates through the process zone and outside it. After carrying out the mechanical tensile tests until the fracture of the dog-bone samples, analyses of the fracture surfaces were carried out, to visualize the character and morphology of the surface.

The "ductile-fragile mixture" behaviour is even more evident in SEM analyses of the crack surface (Fig. 9). As a first observation, the size of the circular zone, generated at the top of the flaw, where the reoriented hydrides are concentrated, is several times larger than that of the process zone. It should be mentioned that the initiation of DHC-type cracking occurs at the top of the process zone, and in a subsequent step, the crack propagates through the process zone and outside it.

The SEM examination direction is perpendicular to the crack surface. One may see the voids, typical for the ductile matrix fracture of the Zr alloy, which interferes with the trans-granular fragile fracture characteristics. In the case of 60 ppm H_2 concentration (Fig. 9 - left), the trans-granular fracture planes can even be the planes of some hydrides, which exhibit low cohesion in the case of mechanical stress that develops tri-axially in the collapse zone of the active area of the specimen.

From the SEM analysis of the fracture surfaces, it can be noted that in the zone of reorientation of the hydrides located at the top of the defects in the dog-bone samples, there is a mixed, ductile-brittle character of the fracture front. It is given by the brittle character of the hydride platelets developed at the tip of the flaw and by the ductile character of the Zr-2.5%Nb alloy matrix.

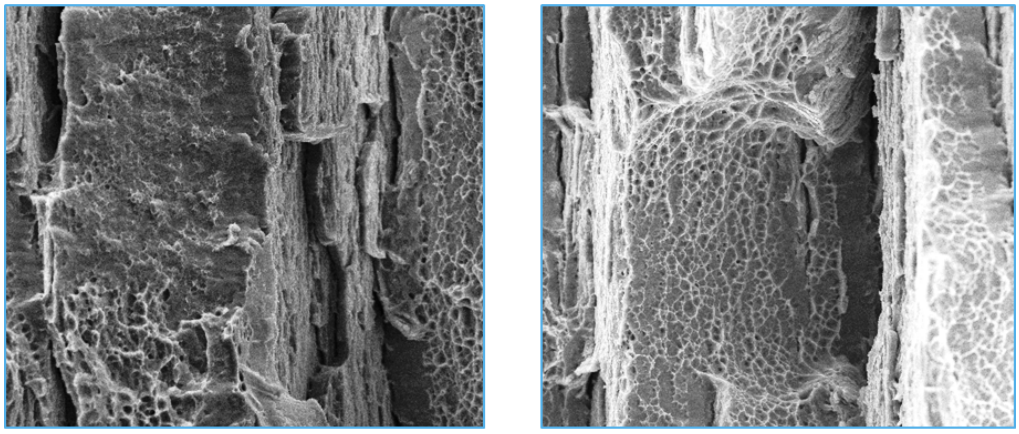


Fig. 9. SEM examination of the cracking hydride surface:
(left) 60 ppm (x1000); (right)) 67 ppm (x1000)

Conclusions

The main objective of the paper is focused on the characterizing hydride reorientation phenomenon at the blunt flaw in the Zr-2.5% Nb alloy in the hydrogen concentration range of 40-80 ppm.

To analyze the mechanical stresses developed at the tip of the defect in the dog-bone samples, the finite element method was used with the help of the VisualFEA software.

The parameters determining the reorientation of localized hydrides, namely the mechanical stress intensity factor K_I, the mechanical stress concentration factor K_C, the tip radius, and the depth of the flaw, were analyzed to fully characterize the Hydride Continuity Coefficient metric, which defines the microstructural morphology of hydrides in the material.

The metallographic and SEM analyses completed the analysis of the reorientation phenomenon of the hydrides located in Zr-2.5%Nb alloy.

The paper approach is foreseen to be implemented in the Post-Irradiation Examination Laboratory on the irradiated material cut from removed pressure tubes in Cernavoda NPP. The experimental results will be also used to build-up a database to perform the structural integrity assessment activities based on the in-service inspection results.

References

[1] Eadie, R.L, Coleman, C.E (1989) ″Effect of stress on hydride precipitation in zirconium-2.5% niobium and on delayed hydride cracking″, *Scripta Metallurgica*, vol. 23(11), pp. 1865-1870.

[2] Ells, C.E. (1968) ″Hydride precipitates in zirconium alloys (A review)″, *J. of Nuclear Materials*, vol. 28(2), pp. 129-151.

[3] Puls, M.P., Shi, S.Q., Rabier, J. (2005) ″Experimental studies of mechanical properties of solid zirconium hydrides″, *J. of Nuclear Materials*, vol. 336(1), pp. 73-80.

[4] Nitu, Al.I., Radu, V., Stoica, L., Toma, D. (2023) ″Characterization of the DHC Crack Parameters in Zirconium Alloys″, *Romanian J. of Physics*, vol. 68(7-8), art. 910, https://doi.org/10.59277/RomJPhys.2023.68.91.

[5] Stoica, L., Radu, V., Nițu, A., Ionescu, V., Toma, D. (2021) ″A New Approach to obtain Ramberg-Osgood Constitutive Equation for Materials used in Structural Components of Nuclear Reactors″, *J. of Nucl. Research and Develop.*, nr. 20-21, pp. 65-68.

[6] Jinga, A., Abrudeanu, M., Radu, V., Stoica, L., Nitu, A., Toma, D. (2023) ″A novel approach for the structural integrity assessment of CANDU Reactor Pressure Tube″, *U.P.B. Sci. Bull.*, Series B, vol. 85(4), pp. 211-222.

[7] Canadian Standards Association (2021) *″CAN/CSA Standard N285.8-21: Technical requirements for in-service evaluation of zirconium alloy pressure tubes in CANDU reactors″*, CSA group, Toroto, Canada.

[8] Simon, P.C.A., Frank, C., Chen, L.Q., Daymond, M.R., Tonks, M.R., Motta, A.T. (2021) ″Quantifying the effect of hydride microstructure on zirconium alloys embrittlement using image analysis″, *J. of Nuclear Materials*, vol. 547(3), art. 152817, https://doi.org/10.1016/j.jnucmat.2021.152817.

[9] American Society of mechanical Engineering (2004) *″ASME Boiler and Pressure Vessel Code, Section XI: Rules for In-service Inspection of Nuclear Power Plant Components″*, ASME Int., USA.