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Study of the influence of zirconium hydrides on fatigue crack initiation at volumetric flaws on CANDU 600 reactor pressure tubes

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Abstract. The Zr-2.5%Nb alloy, from which the pressure tubes of the CANDU 600 power plant at the Cernavodă NPP are made, is permanently subjected to a corrosion process during the normal operation of the fuel channels in the reactor. At the same time, the formation of specific flaws on the inner surface of the pressure tube under the conditions of prolonged irradiation and absorption of deuterium from the cooling agent, heavy water, leads to the initiation of precursor centers for cracking under mechanical load, called in the specialized literature type cracking Delayed Hydride Cracking (DHC). Among the phenomena that can contribute to the initiation and triggering of the DHC mechanism is the phenomenon of fatigue under mechanical load under the conditions of the formation of defects such as "bearing pad fretting flaw" or "debris fretting flaw". These defects are the most important defects revealed by periodic inspections of CANDU fuel channels, as they are the precursors to DHC propagating cracks. Therefore, an important objective in the studies carried out internationally is to analyze and model the consequences of the fatigue phenomenon of the pressure tube, which may contain this type of flaw, under the conditions of hydrogen accumulation, the formation of zirconium hydride platelets and their reorientation in the thermo-mechanical field.

The paper presents part of the experimental studies regarding the performance of thermo-mechanical treatments for the reorientation of zirconium hydride platelets under high mechanical stresses in the Zr-2.5%Nb alloy, the micro-structural analysis and the definition of the HCC metric for characterizing the morphology of the hydrides located in the tube wall of CANDU pressure, carrying out analyzes by the finite element method of the mechanical stress field developed at the top of the defects of the "bearing pad fretting flaw" or the complex defect "bearing pad fretting flaw with debris fretting flaw". Finally, based

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on the data obtained from the mechanical fatigue tests on samples with the mentioned type defects, the fatigue lifetime was determined in the format of the corresponding Basquin-type equation, „the amplitude of the alternating stress versus the number of cycles to failure". The results of the work are useful in the structural integrity analysis of the CANDU fuel channels.

Keywords: CANDU pressure tube, DHC cracking, zirconium hydrides, mechanical fatigue.

1. Introduction

The CANDU pressure tube, made of Zr-2.5%Nb alloy, mechanically behaves like a thin-walled tube, considering the geometric dimensions: length of 6400 mm, inner diameter of 104 mm and wall thickness of 4.2 mm. Under normal operating conditions, the pressure tubes are subject to an internal coolant pressure of about 10 MPa, and the temperature varies from 260°C at the inlet of the D_2O agent (inlet) to 310°C at the outlet of the coolant through the opposite end of the pressure tube (outlet).

The probability that a flaw will be present on the inner surface of the CANDU pressure tube during normal operation is quite high due to corrosion phenomena, vibrations induced by the flow of the agent through the fuel bundles, and precipitation of hydrides by the continuous absorption of deuterium in the zirconium matrix. These phenomena are coupled with the generation of striations (blunt flaws) on the inner surface of the pressure tube by the hard impurities trapped between the pad of the fuel bundle and the pressure tube, or even by these pads when the axial displacement of the bundle train occurs during the process of loading - unloading with fuel. Pressure tube operating experience revealed that approximately 8% of inspected tubes were affected by defects greater than acceptable limits, and of these 60% were produced by hard impurities in contact with the pressure tube or beam skids fuel [1]. The most relevant defects to consider when analysing the integrity of the pressure tubes in the CANDU 600 fuel channels are:

- The defects caused by the friction of the pads of the fuel elements, in areas other than the area of the end-fitting region, called "Bearing Pad Fretting Flaw-BPFF" type defects;
- Defects caused by the friction of hard impurities with the inner surface of the pressure tube are called "debris flaws" defects (Debris Fretting Flaw - DFF).

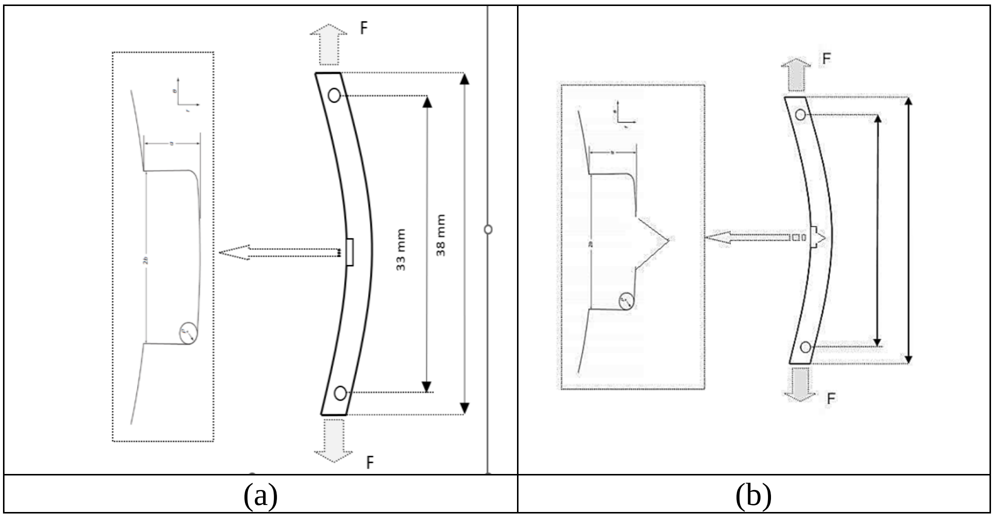
The Zr-2.5%Nb alloy, from which the pressure tubes in the CANDU 600 power plant are made, is permanently subjected to a corrosion process during the normal functioning of the fuel channel in the reactor. When the equivalent hydrogen exceeds the solubility limit, zirconium hydride platelets appear in the pressure tube. In the case of the pressure tube, these are delta hydrides ($ZrH_{1.6}$), which occur as a result of thermal and mechanical stress conditions in the normal operation of the CANDU fuel channels [2]. Due to the brittleness of the hydride plates, a certain brittleness is induced in the zirconium matrix, the host, which loses its original ductility and properties, including those related to the fracture toughness of the material. It should be mentioned that the degree of fragility of the material, in the situation where the hydride plates have formed, is also related to their possible reorientation under certain thermo-mechanical conditions (thermal cycling under continuous mechanical stress). The formation of specific defects on the inner surface of the pressure tube under the conditions of prolonged irradiation and absorption of deuterium from the cooling agent, heavy water, leads to the initiation of precursor centers of cracking under constant mechanical load, called Delayed Hydride Cracking in the specialized literature (DHC).

The Canadian specification imposed a maximum concentration of up to 0.23 atomic per cent (25 ppm) of hydrogen in the fuel channel pressure tubes until 1993. In practice, the tubes contain on average 0.095 atomic % (10.3 ppm), although values of 0.16 % (18 ppm) have also been observed. This last value represents the precipitation concentration of the hydride platelets at 217°C. Assuming an absorption rate of 0.005 atomic per cent per year (1 ppm/year) at the inlet end of the coolant in the pressure tube, then at 250°C the hydrides will be present after approximately 26 years of operation. It should be noted that the hydride front formed in the material when the coolant enters the pressure tube will move under the mechanical stress gradient towards the areas of high stress in the mandrel area [3]. In the specialized literature [4], finite element modelling of the changes induced by hydrogen precipitation in zirconium alloys has already been done. The effect of mechanical stress on hydrogen solubility has been studied in many profile articles, especially for zirconium alloys, which are of great importance for nuclear energy [5, 6, 7]. The problem of hydrogen solubility is usually put in terms of equilibrium thermodynamics for solid solutions, following the equilibrium conditions for the Gibbs potential, taking into account the intensive quantities (partially molar) [8].

Among the phenomena that can contribute to the initiation and triggering of the DHC mechanism should also be considered the phenomenon of fatigue under constant mechanical load under the conditions of the formation of "bearing pad flaw" or "debris fretting flaw" defects. These defects are the most important defects revealed by periodic inspections of CANDU fuel channels, defects from which DHC propagating precursor cracks are initiated. That is why it is very important to know, analyze and model the fatigue phenomenon of the pressure tube, which can contain this type of defect, under the conditions of hydrogen accumulation, the formation of zirconium hydride plates and their reorientation in the thermo-mechanical field.

2. Carrying out experimental tests of controlled hydration and reorientation of zirconium hydrides in the Zr-2.5%Nb alloy

The samples made of Zr-2.5%Nb alloy were processed in the circumferential direction of the pressure tube (Figure 2.1), having the C-type shape.



(a) (b)

Fig. 2.1. Sketch of type C samples with defects: (a) “bearing pad” type, (b) complex type (“bearing pad with debris fretting”).

The controlled hydration of the samples processed from the CANDU pressure tube alloy, Zr-2.5%Nb, was carried out by the method of electrolytic deposition of a film layer of hydrogen, followed by an appropriate thermal treatment, necessary for the diffusion and homogenization of hydrogen in the sample. This method, also used in the specialized literature, especially in the experiments used for the controlled hydration of the Zr-2.5%Nb alloy, was developed in the Metallography laboratory at RATEN Pitești Nuclear Research Institute, first for the Zircaloy-4 alloy. Later, the method was also applied to the Zr-2.5%Nb alloy. The deposition of the hydride layer on the raw samples is carried out in an electrolytic bath containing a solution of H_2SO_4 (0.1%). The samples of Zr-2.5%Nb, polished and pickled, play the role of the cathode, the anode is made of a 5 mm thick lead plate. In the electrolytic bath, a working temperature of 90°C is maintained, and the holding time is 8 hours. Following this treatment, a uniform layer of hydride is obtained, which is subsequently subjected to a thermal homogenization treatment that leads to the diffusion of hydrogen in the volume of the sample.

The current density used was 100 mA/cm^2. To ensure a homogenization of the solution, the heating plate was equipped with a magnetic stirrer. The duration of hydrogenation was chosen to obtain the desired final hydrogen concentration in the sample. The hydride heat treatment consisted of heating the samples in a vacuum chamber at 425°C for 2 hours. Taking into account the variation of the hydrogen concentration in the pressure tubes at the CANDU reactor, in this work the hydrogenation of the samples was carried out in the range of 30-60 ppm, with an absolute error of +/-5 ppm. To highlight the hydrides and determine their orientation, established metallographic techniques were used, namely: the samples were polished and chemically attacked for pickling (solution 45% nitric acid, 45% distilled water, 10% hydrofluoric acid, t = 20 seconds). The hydrides were highlighted through a specific chemical attack (immersion in a solution of 25 ml of lactic acid, 25 ml of nitric acid, and 3-4 drops of hydrofluoric acid, for 10 seconds), the microstructural examination being carried out in a light field, using a metallographic microscope.

To obtain the hydride reorientation phenomenon, specific thermal cycling tests were performed. Thus, an experimental setup was used on the ADAMEL creep machine to allow a thermal cycling treatment under the controlled load of hydrate samples with "patina trace" type defects. To obtain the reorientation of the hydrides in the defective area, thermal cycling treatments were carried out under constant mechanical load. The working sequences on the ADAMEL creep machine were as follows:

- The hydrated and instrumented sample was mounted in the clamping device characteristic of type C samples;
- To dissolve the hydrides, the samples were heated at 300ºC for 24 hours. This was followed by cooling to 185ºC at the rate of 1ºC per minute and the holding time at this temperature was 3 hours. After this, the cooling was continued at 1ºC per minute until the temperature of 90ºC. It is then heated to 250ºC at a rate of 1ºC per minute and maintained for one hour at this temperature.

This scenario represents the first thermal cycle. Cooling from 250ºC to 185ºC and 90ºC follows the sequence of the first cycle. With the help of suitable software, the test parameters (force, temperature, time, electric potential difference) are acquired. The number of thermal cycles varied between 9 and 25 cycles under constant mechanical load. From the preliminary tests carried out on samples with concentrations higher than 30 ppm of hydrogen, the threshold mechanical load for triggering the reorientation of localized hydrides was obtained for the temperature range 280 - 350ºC. Thus, a threshold mechanical tensile stress of about 120 MPa was obtained for this temperature range.

3. Analysis of the reorientation phenomenon of localized hydrides on Zr-2.5%Nb alloy samples

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, having different orientations in the radial-transverse plane. The hydride plates "reoriented" following the heat treatment, are those hydride plates that have the direction of orientation located at an angle that varies from the transverse direction (45º-90º), as defined in Figure 3.1.

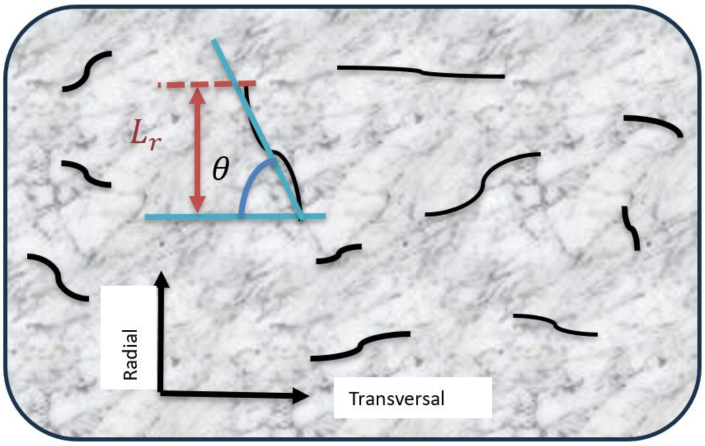


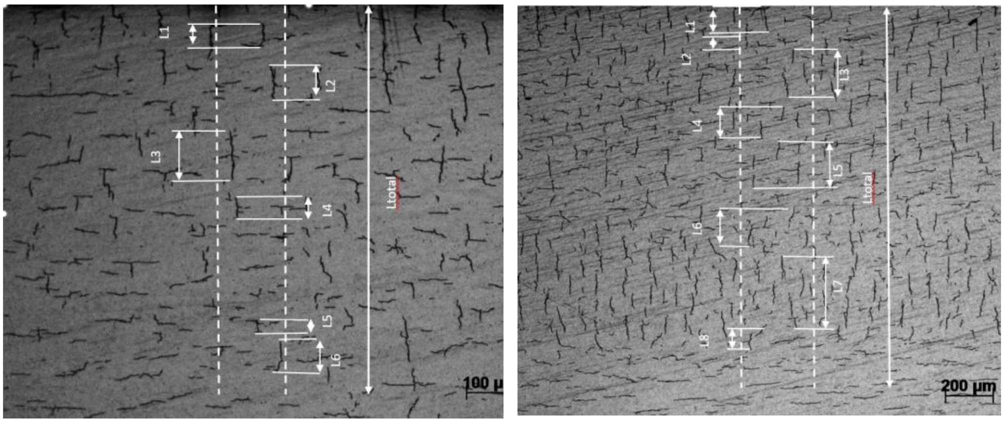
Fig. 3.1. The orientation of the hydride plates in the Zr-2.5%Nb alloy in the radial-transverse section.

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 metric (HCC), based on the analysis of the micrographs in which the zirconium hydride plates are highlighted. In short, the stages of determining this coefficient are:

- An analysis surface is chosen in the shape of a rectangle, whose dimensions are approximately 0.1 mm x 1.0 mm, its orientation being in the radial direction;
- Inside this r;
- Rectangle, the projections of the length of the hydride plates in the radial direction are made,
- The projected lengths L_k of each projection in the radial direction (the length of the rectangular area) are read, as well as the total length of the examined rectangle L_{total};
- The HCC coefficient is calculated with the relation:

$$HCC = \frac{\sum_{k=1}^{n} L_k}{L_{total}} \tag{3.1}$$

The phenomenon of reorientation of the hydride plates in the Zr-2.5%Nb alloy is related to the fact that in the material the hydrogen concentration must exceed the solubility limits at a specified temperature. A more rigorous analysis of the characterization of the reorientation of the hydride plates by the HCC coefficient can be done when the hydrogen concentration reaches higher values, such as the situation of the samples having 55-60 ppm. For these samples, several variants of thermal cycling under constant load were carried out, between 9 and 25 thermal cycles, at a mechanical stress of about 120 MPa, so that the condition of the threshold stress is met. With the increase in the number of cycles, it can be observed (Figure 3.2) that the platelets are oriented very clearly in the radial direction, their length increases in this direction, and consequently, the HCC value becomes higher.



(a) (b)

Fig. 3.2. Morphology of hydride reorientation: (a) sample having the concentration of 60 ppm, 10 thermal cycles, HCC=0.38; (b) sample having a concentration of 60 ppm, 20 thermal cycles, HCC=0.81

Although there are significant variations in the HCC coefficient values calculated on the micrographs of the experimental samples at a certain number of cycles, it can still be found that there is a very clear increase in HCC when the number of cycles is increasing. That is why, in the work, the dependence of the HCC coefficient on the number of cycles was evaluated, without taking into account the hydrogen concentration. The dependence obtained for the HCC metric depending on the number of thermal cycles N_{CT} has the expression:

$$HCC = -0.001 \cdot N_{CT}^2 + 0.088 \cdot N_{CT} - 0.389 \qquad (3.2)$$

It is considered that this relationship can describe the phenomenon of reorientation of hydrides in the pressure tube in the radial direction when the hydrogen concentration is in the range of 30-60 ppm, a range that is relevant for pressure tubes that have a long operation in the reactor.

4. The experimental tests on the initiation of fatigue cracking

One of the structural integrity requirements of CANDU pressure tubes, Zr-2.5%Nb alloy, regarding volumetric defects, consists in their assessment at the initiation of fatigue cracking. It is taken into account that during an evaluation period (the time interval between two periodic inspections of the fuel channels), the initiation of a crack from the analyzed defect must not take place. The general requirements for evaluating the integrity of pressure tubes are described in the clauses of the Canadian standard CAN/CSA N285.8 [i] and are addressed only to pressure tubes made of the Zr-2.5%Nb alloy, which was also used at the CNE plant Cernavoda.

The fatigue crack initiation characterization is carried out in terms related to the fatigue crack initiation curve, described by the Basquin relationship, as the relationship between the maximum amplitude of the effective alternating stress, S_a, and the allowed number of mechanical stress cycles, N, namely:

$$S_a = S_0 \cdot N^{-\alpha} \tag{4.1}$$

with:

S_a - the maximum value of the amplitude of the von Mises alternating elastic stress;

S_0 - constant that usually depends on the minimum radius of curvature of the defect and the HCC metric;

N - the number of cycles of mechanical stress;

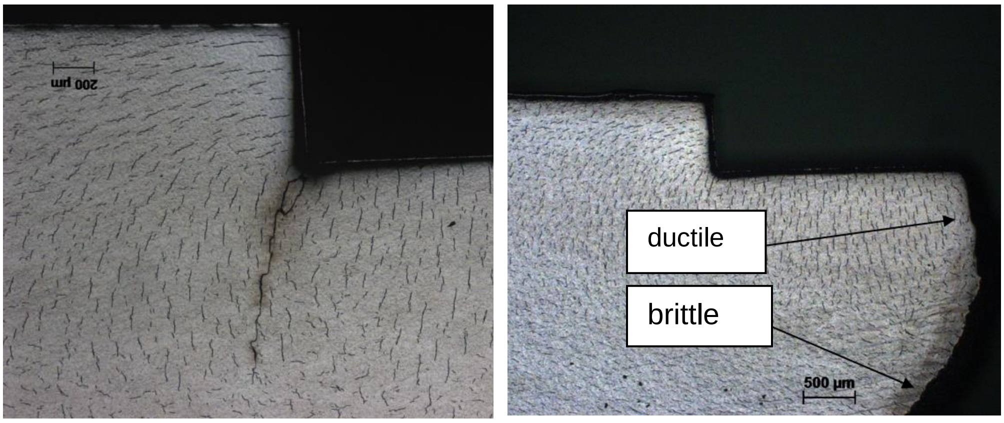
α - material constant.

The experimental tests were carried out at the Walter+Bai mechanical testing facility (Figure 4.1), which is instrumented to ensure the desired frequency of mechanical cycling, the mechanical stress range of the sample (F_{min}-F_{max}), as well as the accounting of the number of cycles that are perform until the moment the crack initiates.

Fig. 4.1. Instrumented Walter+Bai mechanical testing machine for the acquisition and highlighting of fatigue characteristics.

Mechanical fatigue tests were performed at room temperature. The cycling frequency was set at a value of f=2 Hz, and the range of forces applied to the sample during mechanical cycling was F=0.1-1.5 KN, so that, taking into account the dimensions of the tested sample, a range of nominal mechanical stresses of $\Delta\sigma_n$=70-100 MPa.

From the metallographic analyses carried out on the samples with "bearing pad" defect, it was found that the crack propagation occurred in the area of the maximum stress concentrator at the corners of the defect, through the area with reoriented hydrides (Figure 4.2. (a)).



(a) (b)

Fig. 4.2. Crack initiation (a) and rupture (b)
in mechanical fatigue tests on samples with bearing pad-like defects.

When the fracture occurs through such a zone (Figure 4.2. (b)), it can be noted that after the brittle zone where the crack front is perpendicular to the direction of the applied load, the crack propagates to the rest of the section where the hydrides are in the direction circumferential in a plane located at 45° to the direction of mechanical isolation. This fact denotes the ductile character of the area where the plates remained unoriented, i.e. in the circumferential-axial direction of the pressure tube.

In the case of mechanical fatigue tests on samples with complex defects, the initiation of cracking is carried out at the tip of the V-shaped defects (type "patina trace plus impurity trace") where the reorientation of the zirconium hydride platelets was achieved (Figure 4.3).

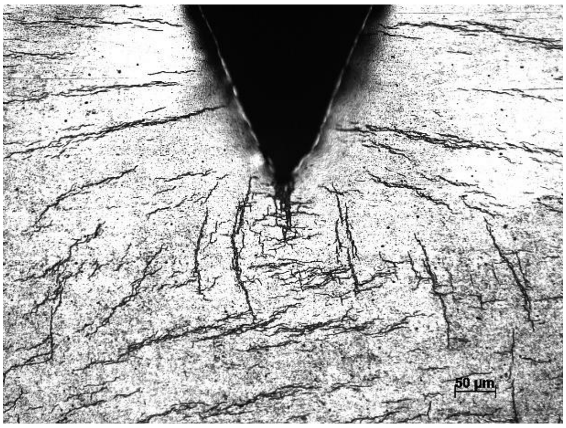


Fig. 4.3. Metallographic images: hydride accumulation
and fatigue crack initiation at the tip of complex flaws.

As can be seen from the analysis of Figures 4.2 and 4.3, the HCC metric in the fatigue crack initiation zone has very high values, namely in the HCC=0.50-0.80 range.

5. Basquin equations for the initiation of fatigue cracking at the defects practiced on Zr-2.5%Nb hydride alloy samples

From mechanical fatigue tests on specimens with "bearing pad fretting flaw" defects, the Basquin relation for the mean fatigue crack initiation curve is obtained:

$$S_a = 675 \cdot N^{-0.17} \tag{5.1}$$

For samples with complex defects of the "bearing pad fretting flaw with debris fretting flaw" type, the obtained Basquin equation is:

$$S_a = 6872 \cdot N^{-0.46} \tag{5.2}$$

In Figure 5.1, the Basquin equations (5.1) and (5.2) are represented, as well as the fatigue curve according to the Canadian standard CAN/CSA N285.8. It should be noted that this fatigue curve stipulated in the standard is the result of fatigue tests on type C samples with only defects in V-type.

It can be seen that the predictions of the two equations are more conservative than the rating curve mentioned by the Canadian standard. Since their predictions are slightly different for the range of values of the number of mechanical stress cycles, an average fatigue curve called the "conservative Basquin curve", of the form was obtained in the work:

$$S_a = 1027 \cdot N^{-0.25} \tag{5.3}$$

The predictions of equation (5.3) are also represented in Figure 5.1, and within a 95% confidence limit, the upper and lower limits of the prediction are also represented. At the same time, the fatigue crack initiation prediction obtained in the work (hatched area in Figure 5.1), can be considered representative of the two types of defects and it is conservative to the Canadian standard CAN/CSA N285.8. It should also be mentioned that this prediction remains valid for hydrogen concentrations in the 30-60 ppm range, as well as for hydrides reoriented at the tip of the mentioned defects having an HCC metric in the range of HCC=0.50-0.80.

In the relevant Canadian scientific literature, it is recognized that it is necessary to treat the curves specific to each type of defect (bearing pad fretting flaw, debris fretting flaw or complex flaws) separately.

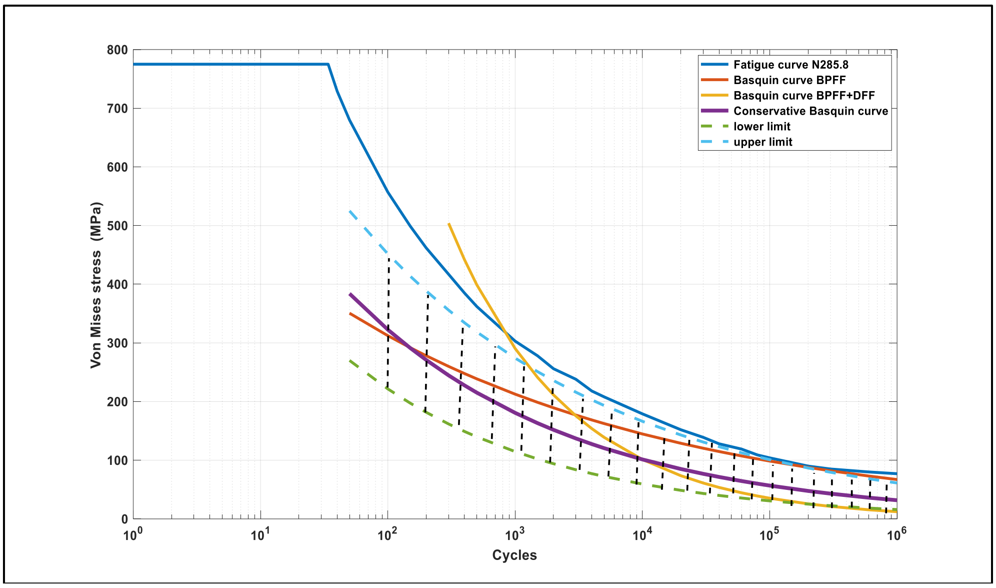


Fig. 5.1 Representation of the Basquin-type fatigue curves obtained in the paper to the Canadian standard CAN/CSA N285.8

Next, the results obtained in the work will be analyzed and discussed as reflected in Figure 5.1.

A fatigue crack initiation curve for an impurity trace-only defect modelled as a "V-flaw" cannot fully correlate with a high-fatigue crack initiation curve for a "bearing pad fretting flaw" or "bearing pad fretting flaw with debris fretting flaw" on the same category of samples. One explanation is that the "bearing pad fretting flaw" type defect has two spots with mechanical stress concentrators, at the two corners of the defect in the cross-section, and thus two locations of localized hydride reorientation, where the HCC parameter has large values. As it was found from most metallographic analyses, fatigue crack initiation occurs almost simultaneously in the two locations, if the radii of curvature are close in value. This is also supported by the metallographic aspect of the reorientation of the localized hydrides, whose densification is visible at the spots of the voltage concentrators and which continues over the entire section of the sample is quite consistent, and the orientation in the radial direction is achieved almost along its entire width.

6. Conclusions

The pressure tubes in the CANDU 600 power plant from CNE Cernavodă are made of the Zr-2.5%Nb alloy, which is permanently subjected to a corrosion process during the normal operation of the fuel channels in the reactor. At the same time, the formation of specific defects on the inner surface of the pressure tube under the conditions of the absorption of deuterium from the cooling agent, heavy water, leads to the initiation of precursor centers for cracking under mechanical load, called in the specialized literature type cracking Delayed Hydride Cracking (DHC). Among the phenomena that can contribute to the initiation and triggering of the DHC mechanism is the phenomenon of fatigue under mechanical load, under the conditions of the formation of a "bearing pad fretting flaw" or "debris fretting flaw".

The paper presents the following activities:

- Characterization of the microstructural morphology of the areas with reoriented hydride samples made from CANDU 600 pressure tube, Zr-2.5%Nb alloy, which were hydrided and subjected to complex thermal treatments with the HCC metric and obtaining an original relation of dependence of this metric on the number of thermal cycles;
- Carrying out mechanical fatigue tests, after which two forms of the Basquin equation were obtained, for the two types of defects, namely "bearing pad fretting flaw" and "bearing pad fretting flaw with debris fretting flaw".
- In the work, an average fatigue curve was obtained, called the "conservative Basquin curve", and within a 95% confidence limit, the upper and lower prediction limits are also obtained.
- Thus, the prediction of fatigue crack initiation obtained in the work can be considered representative of the two types of defects and it is conservative to the Canadian standard CAN/CSA N285.8. This prediction remains valid for hydrogen concentrations in the range of 30-60 ppm, as well as for hydrides reoriented at the tip of the mentioned defects, having the HCC metric in the range HCC=0.50-0.80.

The results of the work are useful in the structural integrity analysis of the CANDU 600 fuel channels from Units U1 and U2 of the Cernavodă NPP plant.

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