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Cladding Tube Deformation Test for Stress Reorientation of Hydrides

ABSTRACT: The phenomenon of stress reorientation of hydrides in fuel cladding tubes was studied with the help of a test technique named as the “cladding tube deformation test” (CTDT). Unirradiated Zircaloy-2 (Zry-2) cladding tube specimens charged with 250 ppm of hydrogen were tested. The test consisted of heating specimens to 400°C and then cooling them down to room temperature at cooling rates of 0.7°C/min and 2°C/min. During the cooling phase, a constant tensile load was applied to specimens with the help of two inner cylinder halves. The stress-strain fields developed in the tube specimen were calculated with the help of the finite element method (FEM). Post-testing metallographic observations revealed considerable amount of hydrides oriented radially as a result of loading. Lengths and relative position of all hydride bands were determined through a semi-automatic image processing technique. Mapping of FEM calculated stress fields on the metallographic section helped to determine a stress threshold value of 72 MPa. Tests performed with a cooling rate 2°C/min showed a considerably lower extent of hydride reorientation. The above test technique was validated by performing internal pressurization tests using pre-hydrided Zry-2 cladding tubes. The external diameter of the tube was tapered by fine turning in order to exert different uniform hoop stress values on different cross sections of the tube. Internal pressurization tests were performed with a maximum temperature of 400°C and a cooling rate of 0.5°C/min. Post-testing metallographic observations on several cross sections of the specimen revealed a stress reorientation threshold of 65 MPa. Percentage of radially oriented hydrides as well as their lengths increased with the stress level until it reached a plateau. Further tests were carried out with the CTDT technique on specimens with H₂ content ranging from 560 ppm to 750 ppm. These results revealed a reorientation stress threshold of around 120 MPa for higher hydrogen content.

KEYWORDS: Zircaloy-2, hydrides, reorientation, fuel, cladding

Introduction

Since early applications of zirconium alloys in water reactors, hydrogen induced embrittlement and cracking have been largely identified as important damage mechanisms [1–4]. The source of damage is hydride precipitates that form as platelets or bands of aligned platelets [5] when the solubility limit of hydrogen in zirconium alloys is reached. The extent of hydride embrittlement depends not only on the quantity of hydrides present but also on its morphology and orientation with respect to the applied stress [6]. In case of fuel cladding tubes, hydrides present perpendicular to the hoop stress direction (radial hydrides) cause a severe loss of ductility in the circumferential direction of cladding tubes [6]. Hydride orientation in unloaded cladding tubes is to a large extent dependent on strain history of the fabrication process [7,8] and is therefore controlled in the cold tube reduction process (pilgering) to be circumferential [9]. However, during reactor operation and subsequent spent fuel handling, a high hoop stress applied to the fuel cladding tube can cause hydrides to precipitate and grow in the radial direction. The high stress field on an incipient crack tip can also cause hydrides to precipitate aligned with the crack growth direction and successive failure of such hydrides can lead to a sub-critical crack growth called delayed hydride cracking (DHC) [10]. The phenomenon of DHC is also of concern to the post-service interim storage of fuel cladding tubes. This is because the blow drying of spent fuel after pool storage can raise their temperature high enough to cause significant dissolution of hydrides in the matrix. On subsequent cooling therefrom, hydride reorientation might propagate an incipient crack through DHC. It is important to study stress reorientation of hydrides in terms of material embrittlement and as a prerequisite to DHC.

Radial reorientation of hydride precipitates in zirconium alloys has been studied over the past four...
decades but has undergone a recrudescence in recent years in the perspective of high burnup operation of fuel cladding tubes [6,7,11,12]. The parameters influencing this phenomenon, should they be related to material microstructure, fabrication processes, or to thermo-mechanical loading conditions, have been thoroughly investigated. A major part of these studies was dedicated to Zr-2.5 % Nb alloys for applications in CANDU pressure tubes [7,13–18]. The techniques used for such investigations have been evolving over this time period. For example, in earlier studies, curved specimens were machined out of pressure tubes and mechanically flattened to obtain a uni-axial specimen suitable for testing in a universal testing machine [13]. Flattening of specimens induced large variations of residual stresses and this was obviously a potential source of error in this type of technique though heat treatments were performed in order to relax residual stresses to a great extent [16]. Investigations have also been performed on tube specimens exposed to internal pressurization [8]. This type of test specimen allows application of loads close to those in real component; i.e., pressure tubes or fuel cladding tubes. However, the amount of material involved in such specimens is high, which is unsuitable for testing on irradiated specimens due to cost, handling, and waste disposal issues.

Modern calculation techniques such as finite element method (FEM) make it possible to develop and practice non-standard mechanical test methods. An example of such a technique used to study hydride reorientation in cladding tubes has been given in a paper published recently [19]. The experimental effort in this case is shifted from that of specimen fabrication and handling to the analysis of complex stress strain fields developed in the specimen during testing. Moreover, numerical image processing techniques can be used to assess, with a higher level of precision, the size and relative location of each and every hydride particle or band. The combination of both leads to a test technique giving more precise results while requiring a small amount of material and at the same time reducing machining and handling efforts on the specimens to the minimum.

This paper presents such a technique called the cladding tube deformation test. The work presented here gives an account of the technique and of results obtained on hydride reorientation in un-irradiated Zry-2 cladding tubes of the type LK3 used in boiling water reactors. The results obtained were validated with the help of internal pressurization tests on the same material under the same thermal cycle. The results were also compared to the ones published in the literature.

**Cladding Tube Deformation Test Setup**

The test setup is shown in Fig. 1(a). The test specimen is a 12 mm long cutup of a fuel cladding tube. Load is applied on the specimen with the help of two cylinder halves with 45° wedge shaped ends. The loading mechanism is schematically shown in Fig. 1(b). Cylinder halves are made to pass through the cladding tube specimen and the ensemble is mounted into a specimen holder with reciprocating 45° wedge shaped surfaces. Specimen holder is mounted into a universal testing machine within a furnace so that required thermo-mechanical loading cycles can be applied to the specimen. Both cylinder halves and specimen holder are made of a commercially available Ni-base superalloy MarM247CC (Ni-10Co-10W-8.5Cr-5.5Al-0.7Mo-3Ta-1.4Hf wt %). This material was chosen for its excellent creep properties below 600°C. The thermo-mechanical loading cycle applied during the test is schematically shown in Fig. 1(c). A specimen is heated from room temperature (RT) to a maximum temperature ($T_{\text{max}}$) so as to dissolve a significant part of hydride precipitates in the specimen. The specimen is then cooled down to room temperature at a slow cooling rate ($dT/dt$). During the cooling phase, a constant load ($F$) is applied to the specimen through the cylinder halves. Hydride bands usually aligned in the circumferential orientation of the tube specimen as observed on a transversal section are reoriented radially in the zones subjected to stress levels greater than the threshold stress. One of the aims of this test was to determine the threshold stress for hydride reorientation and to compare it with other studies in the literature. The type of loading mechanism in CTDT brings about bending of the cladding tube specimen and also some bending of the cylinder halves. It generates a 3D stress field that needs to be calculated through FE modeling of the test setup. The other objective of the study was to validate the CTDT technique by simple-to-interpret tests.

**Tube Internal Pressurization Test Setup**

The CTDT is a non-standard technique with a non-uniform stress distribution calculated through FEM. The results from CTDT must be confirmed and validated with the help of another test technique leading to
a uniform tangential (hoop) stress on a transversal cross section of a tube specimen. It was therefore chosen to perform validation tests on pre-hydrided Zry-2 tubes using internal pressurization. In the case of pressurized tubes, the hoop stress is also the maximum principal stress. Therefore, the comparison of the stress states between the CTDT specimen and the pressurized tube specimen should be done on the basis of maximum principal stress. The specimen dimensions are shown in Fig. 2. The outer diameter of the specimen is tapered by fine turning in order to obtain a variation of stress in the axial direction of the specimen. The ends of the axial segment with constant wall thickness were used to clamp the specimen.
The specimen was pressurized with oil with the help of an electrical feedback control system. It was then enclosed within an electric heating chamber and the temperature was regulated and controlled with the help of two thermocouples attached to the middle of the gage length. Two tests were carried out, one with 75 bar internal pressure (7.5 MPa) and the second one with 135 bar (13.5 MPa). The specimen was not axially restricted. A hoop stress range from 55 MPa to 170 MPa was covered with these two tests.

Material and Test Specimens

The test material used in this study was un-irradiated and pre-hydried Zry-2 cladding tube material. The material was provided by two different sources. The CTDT specimens were provided by Sandvik Materials Technology (SMT) on behalf of Westinghouse Sweden, as 12 mm long cladding tube cutups. These specimens were cut from a liner tube of the type LK3 (internal diameter 8.63 mm, external diameter 9.84 mm, original wall thickness with a Zr-0.25Sn liner 605 μm) in fully re-crystallized condition with a grain size of ASTM 13. Before cutting specimens for hydriding, the inner liner was removed by flush pickling. The tube wall thickness after the removal of the liner was 445 μm. It should be noted here that the final wall thickness was not previously specified and it resulted from the flush pickling process meant to fully remove the liner and the interface zone and to obtain a smooth inner surface finish. The specimens were hydrided by SMT in a sealed system with horizontal glass tube. The hydriding took place at 400 °C in vacuum. The holding time was 2 h followed by slow cooling to room temperature. Two groups of specimens were made as a function of their hydrogen content. The first group contained around 250 ppm of H₂, while the other contained different hydrogen contents ranging from 560 ppm to 750 ppm.

The specimens used in the internal pressurization tests were provided by Westinghouse Columbia in the form of cladding tubes of 110 mm length. These tubes were cut from un-irradiated cladding tubes of the type LK3 with an inner liner (internal diameter 8.3 mm, external diameter 9.6 mm). The initial wall thickness of these tubes was therefore 650 μm including the liner, which was not removed, unlike the CTDT specimens. The specimens were hydrided using the same procedure as that for CTDT specimens, above, from SMT. The final hydrogen content of these specimens was around 250 ppm; however, a rim of hydrides was formed at the interface between the inner liner and the outer Zry-2 component. Due to the formation of the rim the real concentration of hydrogen in the Zry-2 cladding bulk would be less than 250 ppm. The precise relative concentration of hydrogen in the liner and in the Zry-2 bulk cladding is not known. The micrographs of Fig. 3 show the orientation and morphology of hydrides precipitates in as-received conditions. It can be noticed that hydride precipitates are almost exclusively aligned in long circumferential bands in all specimens. The bright strip in Fig. 3 represents the inner liner. Though the tube specimens used in the internal pressurization tests were delivered with inner liner, the hydrde rim associated with them was ignored since the objective of these tests was to determine the stress threshold leading to hydrides reorientation in the bulk of the specimen.

Test Conditions

A similar type of thermo-mechanical loading cycle was applied to specimens in all tests (Fig. 1(c)). The difference between the CTDT and the internal pressurization tests being that of loading mechanism; applied force in the former case and applied pressure in the latter. Table 1 gives an overview of all specimens tested in the present study.

Finite Element Modeling of Test Specimens

Separate 3D FE calculations were carried out for every test condition and in the case of CTDT, for every combination of load, geometry of cylinder halves, and temperature. Commercial FE code ANSYS version 10 was used for this purpose.

FE Modeling of CTDT

Only one eighth of the test setup including cladding tube specimen and cylinder halves was modeled taking benefit of the symmetry with respect to XZ, YZ, and XY planes (Fig. 4). Both cladding tube
FIG. 3—Hydride precipitates in as-received specimens: (a) 250 ppm CTDT specimen, (b) 750 ppm CTDT specimen, and (c) 250 ppm internal pressurization specimen.

TABLE 1—Overview of thermo-mechanical loading conditions used in all tests and that of the results.

<table>
<thead>
<tr>
<th>Test Type</th>
<th>$H_2$ Content, ppm</th>
<th>$T_{max}$, °C</th>
<th>Specimens Tested</th>
<th>Dwell at $T_{max}$, min</th>
<th>Cooling Rate, °C/min</th>
<th>Load, N</th>
<th>Reorientation Threshold Stress, MPa</th>
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<td></td>
<td></td>
<td></td>
<td></td>
</tr>
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<td>70</td>
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<tr>
<td></td>
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<tr>
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<td>0.5</td>
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<td>560</td>
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<td>0.5</td>
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<td>140</td>
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<tr>
<td></td>
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<td>0.5</td>
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<td>0.5</td>
<td>290</td>
<td>135</td>
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<td>75 bar</td>
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<td>1</td>
<td>30</td>
<td>0.5</td>
<td>135 bar</td>
<td>65</td>
</tr>
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</table>
specimen and cylinder halves were modeled using eight-node brick elements. Cylinder halves were attributed temperature dependent elastic-plastic mechanical properties and thermal expansion coefficients of MarM247CC.

For the contact between cladding tube and the cylinder halves, a 3D deformable-deformable contact was modeled. In order to simulate bending of the tube specimen and an eventual bending of the cylinder halves, the type of contact was chosen to be such that slipping between the two was possible and load was rendered zero when a separation occurred between any two contacting elements. Intuitively, an important parameter for the modeling work was the friction coefficient between Zry-2 and MarM247CC. The friction coefficient has been found to have a large impact on the results of other cladding tube testing techniques such as the ring tensile test [20] or conical mandrel test [21]. At the level of load transmitted through the contact in a CTDT (290 N maximum), the resulting friction force is expected to be small. Moreover, a high temperature resistant lubricant Molycote® was used between the CTDT specimen inner wall and the cylinder halves. The presence of a sufficient amount of the lubricant at all steps of the tests was controlled with the help of preliminary interrupted tests. It was further checked through FE calculations that for a range of friction coefficients (between 0 and 0.2), there was a negligible impact of friction on resulting stresses. For all these reasons, a frictionless contact was assumed between the CTDT specimen and cylinder halves.

Since the loading is force controlled, the FE stress calculations are independent of the mechanical properties of materials used. However, since the extent of bending of the CTDT and the cylinder halves modifies the gap between these two, which would alter the bending moment transmitted to the lateral CTDT specimen walls to a small extent, it is therefore of some importance to use suitable mechanical properties for both the CTDT specimen and cylinder halves. Mechanical properties for the CTDT specimen used into the FE code were generated in-house in the framework of PSI’s participation in the NFIR III program. These properties were obtained through burst tests on 200 ppm H₂ charged pressurized tube specimens in the hoop direction. Besides, it was found that in the elastic range, there was negligible difference of tensile hoop mechanical properties between 0, 250, and 400 ppm charged fuel cladding tubes. That is why the uncertainty in stress values induced while using 200 ppm H₂ Zry-2 mechanical properties for 250 ppm H₂ CTDT were estimated to be negligible.

Calculations were carried out in three steps. In the first step, a small load of 5 N was applied to the specimen through cylinder halves and a static calculation was carried out. The purpose of this step was to simulate initial contact between the cylinder halves and tube specimen in order to later apply the $T_{\text{max}}$ under static equilibrium conditions. In the second step, a uniform temperature equal to $T_{\text{max}}$ was applied to the whole setup. A second static calculation was run in order to calculate the relative displacement of the cylinder half and tube specimen due to the difference of thermal expansion coefficients of their respective materials. In the third step, the full load was applied to the specimen through cylinder halves, and static
calculation was carried out with automatic time stepping. Since no creep deformation was expected, the stress distribution obtained at the third calculation step was taken to be the one applied during the entire cooling phase. This hypothesis was checked after all tests by comparing the diameter of the test specimen before and after the test. No permanent deformation was detected.

Figure 4 shows a FE contour plot of the CTDT test specimen under an applied load of 290 N at $T_{\text{max}}$ of 400°C. As can be seen, the term “hoop stress” loses its significance in the case of the CTDT specimen because of a non-uniform 3D stress field. There is an across-the-wall gradient of maximum principal stress at the point of lateral bending of the specimen, from high stress on the inner wall side to lower on the outer wall side. The maximum principal stress gradient on the top of the specimen, at the point of contact between the specimen and the half cylinder, is from the outer to the inner wall sides. The direction of maximum principal stress, not drawn in the contour plot of Fig. 4, is parallel to the tube circumference in all the region of positive maximum principal stresses. The maximum principal stress is therefore analogous to the hoop stress in the regions of positive maximum principal stresses; i.e., on the inner wall side on the bending point and on the outer wall side on the point of contact between the CTDT specimen and the cylinder halves.

**FE Modeling of Internal Pressure Tube Specimen**

The externally tapered and internally pressurized tube specimen was also modeled in 3D. A two step simulation was carried out. In the first step, a constant temperature equal to $T_{\text{max}}$ was applied to the specimen. It was supposed that the coefficients of thermal expansion of the cladding tube and the clamping material were the same so that heating would not alter the clamping force. Then the 25 mm long end sections of the specimen were blocked in radial displacement. The axial degree of freedom of the specimens was maintained. In the second step, an internal pressure was applied and static calculations were carried out. Due to the tapered external diameter, the hoop or maximum principal stress increased along the axis of the specimen to attain its maximum value in the filet.

**Metallographic Analyses**

For CTDT specimens, the metallographic sections corresponded to the middle transversal cross section of a specimen. Consequently, only one cross section per specimen was observed. Micrographs were taken at different angular locations on the metallographic section, scanning the entire section. Typically, ten micrographs per cross section were considered for further analyses. For the specimens from the internal pressurization tests, five transversal metallographic sections were made per specimen, corresponding to different hoop (or maximum principal) stress levels. After mounting, metallographic samples were ground and polished. They were then chemically etched with a 90%HNO$_3$+10%HF solution to reveal the hydride precipitates. All samples from all specimens were observed with the help of an optical microscope. Electronic images of all observations were numerically recorded for further analysis. Figure 5 shows an example of micrographs obtained on tested specimens. Figure 5(a) shows a micrograph of a CTDT specimen containing 250 ppm of hydrogen and located on the middle cross section. The micrograph contains some hydride bands in the circumferential orientation of the specimen like the ones shown in as-received condition in Fig. 3(a). It also contains some hydride bands oriented perpendicular to the circumferential hydrides close to the inner surface of the specimen. Figure 5(b) contains a micrograph taken on the 94 MPa transversal cross section of an internal pressurization test specimen. The location, morphology, and angular position of each single hydride or hydride band were determined.

**Statistics on Hydride Precipitates**

All hydride particles on all the micrographs of all specimens were counted individually. Their location and angle with respect to the specimen cross section were determined. With the knowledge of their orientation angle, the relative percentages of radial and circumferential hydrides were determined. All hydrides or hydride bands, the angle of which was within $\pm 30^\circ$ of specimen’s hoop direction, were considered as circumferentially oriented. The ones within $\pm 30^\circ$ of specimen’s radial direction were counted in the category of radial hydrides. With the knowledge of precise location of a hydride on a specimen’s cross
section, the stress value occurring on it was determined. Another feature of interest was the length of radial hydride bands. Its evolution with the applied stress during cooling was also determined.

Micrographs were analyzed in a semi-automatic way with the help of an image processing software: Scion Image. This technique consists of selecting all metallographic details, hydrides and others, on a micrograph based on a gray scale threshold. Then the selected details are modeled with ellipses in order to determine their lengths, widths, inclination angles, etc. Figure 6 shows different steps of the hydride analysis procedure. The first step was density slicing (Fig. 6(a)), in which a gray level micrograph was rendered binary. Since radial and circumferential hydrides have the same gray level, in case of their intersection, the resulting particle has a random shape and inclination angle. The second step was particle contouring (Fig. 6(b)), in which the boundaries of all selected particles are highlighted. In order to avoid the problem raised by intersection of radial hydrides with circumferential ones, the radial hydrides or bands of radial hydrides were, in addition, contoured manually. The third step was particle analysis (Fig. 6(c)). In this step, particles outlined in the previous steps were counted and their lengths and orientation angle with respect to the micrograph frame were determined. Since the position of every micrograph on the specimen cross section was known, the orientation angle of each and every hydride particle or band with respect to specimen’s cross section could be determined. Two more filtering procedures were applied in order to eliminate all useless details present on a micrograph due to the etching process. All counted particles with a length-to-width ratio less that 2.5 were eliminated and were not considered as hydrides. Moreover, all particles less than 15 μm in length were also eliminated. The justification of the above two limits was driven from intensive observations and statistical distributions of hydride length and morphology. In order to avoid risk of repetition in counting, the radial hydrides counted through automatic contouring were ignored and only the ones counted through manual contouring were taken into account for statistics. Circumferential hydrides, however, were only automatically treated. At each step, the resemblance of original micrograph and its numerical model were verified.

**Determination of Stress Threshold for Hydride Reorientation**

Stress threshold for hydride reorientation was determined at first on CTDT specimens and then confirmed with results from internal pressurization tests. This procedure is explained in Fig. 7. The individual
location \((x_i, y_i)\) and individual length \(l_i\) of all manually contoured hydride bands were determined. Fitting the edge of a specimen in a micrograph with a straight line equation, the distance \(d_{i,\text{center}}\) between the middle of the \(i\)th hydride band and the specimen’s edge was calculated. Finally the distance \(d_{i,\text{end}}\) of the farthest end of the \(i\)th hydride band from the specimen’s edge was calculated as: \(d_{i,\text{end}} = d_{i,\text{center}} + l_i/2\). This distance was then fed into the across-the-wall maximum principal stress profile calculated by FEM at the

FIG. 6—Different steps for statistical analysis of hydrides using image processing: (a) density slicing, (b) particle contouring, and (c) particle numbering and measurement.
location of the micrograph. The maximum principal stress applied at the farthest end of a hydride band was hence determined. This procedure was repeated for all hydride bands of a micrograph and the minimum of stress values occurring on all the hydride bands of a micrograph was retained. Ten micrographs per specimen were analyzed and all the minimum stress values obtained through all micrographs were averaged to obtain the hydride reorientation threshold value.

Results

Stress Threshold for Hydride Reorientation

The stress thresholds for hydride reorientation, as determined on CTDT specimens, are summarized in Table 1. It can be noticed that the stress threshold values are dependent upon the hydrogen content of the specimen. For 250 ppm, this value is 72 MPa as an average of three specimens. The deviation from the average value is small. For higher values of hydrogen content, the stress threshold rises. For instance, for the group of single specimens with hydrogen content ranging from 560 ppm to 770 ppm, the stress threshold values ranged from 115 MPa to 140 MPa. There is at present no evident correlation between the stress threshold values and hydrogen content for this particular group of specimens, especially as tests at a given hydrogen level could not be repeated. The general trend, however, is that of a higher stress threshold for higher hydrogen content.

Two tests performed with a faster cooling rate of 2°C/min could not be analyzed properly since a lower extent of radial reorientation was observed in these specimens as compared to the ones cooled at <1°C/min. As a consequence, the observed micrographs were statistically very poor in radial hydrides. These tests may be repeated later with a much higher applied load in order to enhance the effect of stress in the precipitation process.

Mapping of FE Results on Micrographs

In order to visualize the effect of a threshold stress on the reorientation of hydrides, the FE calculated stress distribution was mapped on the micrographs taken at different angular locations of an observed metallographic section. Figure 8 gives a comparison of FE calculated applied stresses and hydride reorientation in case of a 250 ppm H2, CTDT specimen. For the sake of simplicity, the value of stress threshold determined previously is used to render the contour plot of Fig. 8 binary, with a darker shade corresponding to stresses below 72 MPa. It can be seen from this comparison that the orientation of hydrides follows the maximum principal stress distribution in quite a precise manner. When the maximum principal stress

FIG. 7—Procedure for determining the stress threshold for hydride reorientation in CTDT. The stress occurring on the farthest from the wall edge of a hydride band is determined through comparison with across-the-wall maximum principal stress profile.
is above the threshold on the inner side of the specimen wall, hydrides are reoriented radially on the inner side. When the stress does not go beyond the threshold value, no radial hydrides can be seen. Finally, when a stress above the threshold is applied on the outer side of the specimen’s wall, hydrides reorient radially on the outer side. The location and depth of radial hydrides correspond well with the threshold stress. Moreover, all radially reoriented hydride bands are situated in the positive maximum principal stress region of the specimen where the maximum principal stress is parallel to the tube circumferential direction (analogous to a hoop stress). Hydrides are therefore reoriented perpendicular to the maximum principal stress direction. The same behavior was observed on all the CTDT specimens tested in this study.

Hydride Reorientation During Internal Pressurization

Five micrographic sections were analyzed for each one of the specimens that had been exposed to internal pressurization. The applied stress was uniform on one cross section but varied from section to section due to the external taper of the specimens. The results of the internal pressurization tests were analyzed in the following way: the percentage of radial hydrides was calculated on every metallographic section, on the basis of five micrographs per section. Counted hydrides were mostly either radial or circumferential. Hydrides falling out of these two categories were relatively small in number and were ignored, thus avoiding the risk of including non-hydride details in the analysis. The radial hydride percentage was calculated by dividing the total number of radial hydrides counted on five micrographs of a section by the sum of radial and circumferential hydrides. This percentage is plotted as a function of applied stress in Fig. 9. Secondly, the length of all radial hydride bands was determined and plotted as a function of applied stress in Fig. 9. Both radial hydrides’ percentage and the maximum length follow a similar trend as a function of applied stress. Two plateaus can be observed at low and high stress values. Both increase sharply as a function of stress from lower to upper plateau. Given also are a few micrographs in the transition zone. The “zero” stress state corresponds to the as-received specimens.

Discussion

Hydride Reorientation Threshold Stress

In order to determine a stress threshold for hydride reorientation, a criterion for the beginning of reorientation must be defined. Moreover, it is well known that there exists a certain proportion of radial hydrides even in the as-received specimens.

It is observable in Fig. 3 and quantified in Fig. 9 that about 5 % of the hydrides in the specimens used for internal pressurization tests are radial prior to testing. The maximum length of as-received radial hydride is around 30 μm. Both maximum length and percentage of radial hydrides increase with increasing applied stress up to 110 MPa, where a plateau is reached. In the literature, different authors have taken different criteria for defining the onset of hydrides radial reorientation. For example, using internal pressurization tests on stress relief annealed Zry-4 specimens, Chu et al. [8] have defined an observable radial to circumferential hydrides fraction of 5 % as criterion. Although their test matrix does not include tests at low stress levels, these authors, based on model calculations, determine a stress threshold of around 55 MPa for Zry-4 specimens charged with 250 ppm of H₂. On the other hand, a criterion used by Singh et
al. [18] when studying hydride reorientation in flattened Zr-2.5 %Nb tube specimens, was that 50 % of the micrograph surface should be filled with radial hydrides. These authors obtain a stress threshold value of 129 MPa for Zr-2.5 %Nb specimens containing 100 ppm of H₂ and heated up to a T_{\text{max}} of 350°C. In another study presenting tests on Zry-4 cladding tube specimens, Daum et al. [19] obtain a hydride reorientation pattern quite similar to the one described in this paper on CTDT specimens (Fig. 8). Their criterion for hydride reorientation threshold stress is the stress acting on an angular location of the cladding tube transversal section where the transition between zones of radial to circumferential hydrides takes place. These authors obtain a hydride reorientation stress threshold of 80 ± 10 MPa irrespective of H₂ content with T_{\text{max}} equal to 400°C. However, the criterion of angular position is very sensitive to the precision of angular mapping between metallographic section and the FE contour plot. This is the reason we chose to determine the stress threshold based on into-the-wall depth of radial hydride bands.

It can be observed in Fig. 9 for the pressurized tube specimen, that at 55 MPa the radial-to-total hydride percentage is around 6 %; i.e., almost at the same level as in as-received specimens. At 65 MPa, the same fraction is 11 %, i.e., 6 % higher than under the as-received conditions. Intuitively, 65 MPa can be taken as a threshold value for hydride reorientation in 250 ppm H₂ Zircaloy-2 specimens heated to a T_{\text{max}} of 400°C. At 74 MPa, this proportion is 17 %. Nevertheless, as mentioned earlier in this paper, the pressurized tube specimens contain an inner liner that absorbed hydrogen preferentially in such a way that the real concentration of hydrogen in the cladding bulk is in fact less than 250 ppm. There is clear evidence in the literature on hydride reorientation that the percentage of observed radial hydrides is inversely proportional to the hydrogen content under the same T_{\text{max}} and applied stress conditions. For example, Shanahan and Hardie [17] showed, for Zr-2.5 %Nb alloy containing from 100 ppm to 300 ppm of hydrogen, heated up to a T_{\text{max}} of 400°C and then cooled down slowly under stress, that the 100 ppm specimen exhibited the highest percentage of reoriented hydrides followed by 150, 200, 250, and 300 ppm specimens in descending order. Chu et al. [8] also presented results confirming this observation for hydrogen charged Zry-4 specimens. It can therefore be envisaged that the observed radial hydride percentage in the pressurized tube specimens is a little higher than what is expected for a 250 ppm specimen under the same conditions. Inversely speaking, the stress level of 74 MPa should correspond to a radial hydride percentage less than 17 %, as observed in this paper. Consequently, the threshold stress for radial reorientation of hydrides in 250 ppm charged pressurized tube specimens should be slightly higher than 65 MPa.

Comparing this result with the stress threshold values obtained through CTDT Table 1, it can be
concluded that for 250 ppm H$_2$ specimens, a mean stress threshold value of 72 MPa obtained through CTDT is already slightly into a stress range where some significant proportion of radial hydrides begins to be observed.

Reorientation Threshold Stress for Specimens With Higher H$_2$ Content

Testing on specimens with higher hydrogen concentration revealed a higher stress threshold as compared to the samples with 250 ppm H$_2$ (Table 1). Although Daum et al. [19] have found the stress threshold to be independent of hydrogen content, Chu et al. [8] found it to increase with increasing H$_2$ content for the same maximum test temperature.

In fact, the stress reorientation of hydrides is a precipitation driven process [8]. Factors influencing the precipitation of hydrides in zirconium matrix affect the reorientation process as well. These factors are maximum test temperature, hydrogen content, cooling rate, applied stress during cooling, and number of loading cycles. In addition, the hysteresis of terminal solid solubility curves (TSS) in either heating (TSSD) or cooling (TSSP) [22] must be taken into consideration. In our study, the concentration of hydrogen dissolved at 400°C is around 200 ppm, the rest staying undissolved in the matrix. Consequently, 80 % of the hydrogen was dissolved in specimens with 250 ppm of H$_2$, while only 30 % of the hydrogen content in samples with 650 ppm H$_2$ was dissolved upon heating to 400°C. The remaining 70 % increased the relative proportion of circumferential hydrides. The dissolved 200 ppm of H$_2$ would start to precipitate at around 330°C, corresponding to its TSSD temperature [22]. The applied stress would influence the precipitation of hydrides between 330°C to RT in a non-linear way, dependent upon the diffusion rate of hydrogen at different temperatures [8]. If the $T_{max}$ was raised beyond 400°C, the extra dissolved hydrogen would nucleate hydrides on new sites under the influence of an applied stress. Consequently, the threshold stress shall decrease. According to the model developed by Chu et al. [8], for 600 ppm H$_2$ SRA Zry-4 specimens heated up to 400°C and cooled down under applied stress at 1°C/min, the stress threshold, taken at 5 % radial hydrides fraction, is equal to 100 MPa. Among the tests performed in the present study, the closest to 600 ppm hydrogen content is 620 ppm and the threshold stress value obtained through CTDT on this specimen is 117 MPa (Table 1); i.e., slightly higher but nevertheless in the same range as the value predicted by the model presented in [8].

Maximum Length of Radial Hydride Bands

The maximum length of hydride bands is an important aspect to consider when it comes to the effect of radial hydrides on mechanical properties of cladding tubes. This is because long hydride bands provide a function of applied stress, measured from internal pressurization tests in this study. For a 250 ppm H$_2$ content, this length appears to plateau at around 75 μm beyond 110 MPa of applied stress. Comparing this result to the maximum length of hydride bands measured on the CTDT specimen, it is, moreover, found that the maximum radial hydride bands length falls in the range 70 μm to 90 μm (Fig. 7). This finding confirms the existence of a stress dependent plateau for the maximum radial hydride band length for a given H$_2$ content.

Conclusions

It is concluded that the cladding tube deformation test has been validated and can be used for studying hydride reorientation in fuel cladding tubes. The following arguments lead to this conclusion:

- The stress threshold value for reorientation of hydrides obtained through CTDT, in un-irradiated Zircaloy-2 specimens, is 72 MPa for 250 ppm H$_2$ specimen tested at a $T_{max}$ of 400°C and cooled down under stress at a rate of $<1°C/min$. This is consistent with results presented earlier [17,19].
- The stress threshold for hydride reorientation seems to increase to around 120 MPa for hydrogen content between 560 ppm and 750 ppm. Some recently published results in the literature support this finding.
- Pressurized tube test results on Zry-2 tubes with liner containing 250 ppm of hydrogen and heated to 400°C lead to a stress threshold value of 65 MPa. However, considering that due to the liner the actual hydrogen concentration in the cladding bulk is lower than 250 ppm, the stress threshold
value could be slightly higher than 65 MPa for a 250 ppm hydrogen specimen. This argument is based on published experimental results showing that under the same temperature and stress conditions, a lower hydrogen content specimen exhibits a higher radial hydride percentage. Consequently, for a 250 ppm hydrogen pressurized tube specimen, a reorientation threshold stress value slightly higher than 65 MPa is expected.

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References


**DISCUSSION**

*Question 1, J.L. Bechade, CEA, France:*—Did you observe any effect of the cooling rate on the reorientation of hydrides?

*Authors Reply:*—Our tests were, in general, performed with a cooling rate of 0.5°C/min. Only two tests were performed at 2°C/min. As it is explained in the paragraph 10.2, there was a drop of the extent of hydride reorientation i.e. the proportion of radial hydride bands versus total hydride bands in the 2°C/min specimens as compared to the 0.5°C/min ones. It made the results statistically too poor to be analyzed properly. Qualitatively, however, the reorientation stress threshold value did not seem to drop in the 2°C/min tests.

*Question 2, Gerry Moan, AECL, Canada:*—Your specimens contained 250 ppm H or greater and the specimens used by other workers also contained 250 ppm H. Is there any significance to the 250 ppm H value?

*Authors Reply:*—For us, the value of 250 ppm of hydrogen is representative of the hydrogen absorbed by BWR Zircaloy-2 fuel cladding tubes in high burnup operation.

*Question 3, Karl-Fredrik Nilsson, Institute for Energy, Netherlands:*—1) Did you consider different temperatures? Is the threshold stress a function of the temperature? 2) Why did you not consider 100 ppm, more relevant than 600 ppm?

*Authors Reply:*—1) The present study in fact focused on the validation of the CTDT and therefore the main test results shown here correspond to a 400°C maximum temperature. Further investigations and some literature work indicate that the threshold stress for reorientation of hydrides is in fact dependent upon the maximum test temperature and the hydrogen content.

2) In this first test series the idea was to measure the hydride reorientation threshold for unirradiated Zircaloy-2 cladding tube material charged to a hydrogen level representative of high burnup irradiated cladding tubes. The purpose of using very high hydrogen content (600 ppm) was to check if there was any evolution of the measured stress threshold with the hydrogen content. The idea was to prepare the ground for future studies.

*Question 4, Paul Cantonwine, GNF, USA:*—Is the cooling rate used reasonable relative to what is expected during a helium backfill?

*Authors Reply:*—Our results indicate that a faster cooling rate reduces the extent of hydride reorientation. In the present paper, a few tests were performed with a faster cooling rate of 2°C/min. The results of these tests showed a hydride reorientation of a much lower extent; so much so that the micrographs were statistically poor for a reliable determination of reorientation stress threshold. On the opposite side, it can be expected that a slower cooling rate under applied stress shall favor the extent of radial reorientation of hydrides.

Hydride reorientation data presented in the literature is usually produced at a cooling rate of less than 1°C/min. It can be expected that a much lower cooling rate shall have an impact on increasing the proportion of radial hydrides by giving ample time for hydrogen diffusion and precipitation on new nucleation sites. The final length of hydride bands is not expected to be affected by a much slower cooling rate because it depends upon the total amount of hydrogen in solid solution. The threshold stress for reorientation also intervening in the energy balance of hydride nucleation shall not be affected by a much slower cooling rate. These hypotheses, however, needs experimental verification, something not yet thoroughly addressed in the literature.