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The influence of stress state on the reorientation of hydrides in a zirconium alloy

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ABSTRACT

Hydride reorientation can occur in spent nuclear fuel cladding when subjected to a tensile hoop stress above a threshold value during cooling. Because in these circumstances the cladding is under a multiaxial stress state, the effect of stress biaxiality on the threshold stress for hydride reorientation is investigated using hydrided CWSR Zircaloy-4 sheet specimens containing ~180 wt ppm of hydrogen and subjected to a two-cycle thermo-mechanical treatment. The study is based on especially designed specimens within which the stress biaxiality ratios range from uniaxial ($\sigma_2/\sigma_1 = 0$) to "near-equibiaxial" tension ($\sigma_2/\sigma_1 = 0.8$). The threshold stress is determined by mapping finite element calculations of the principal stresses and of the stress biaxiality ratio onto the hydride microstructure obtained after the thermo-mechanical treatment. The results show that the threshold stress (maximum principal stress) decreases from 155 to 75 MPa as the stress biaxiality increases from uniaxial to "near-equibiaxial" tension.

1. Introduction

Zirconium-based alloys have been widely used in the nuclear industry as nuclear fuel cladding in nuclear power reactors because of their low neutron absorption cross-section, high temperature corrosion resistance and adequate mechanical properties. However, as a byproduct of waterside corrosion, hydrogen is picked-up by the cladding. While up to 120 wt ppm of hydrogen can typically be in solution in a zirconium alloy during elevated temperature operation [1,2], when the reactor shuts down and the cladding cools, nearly all of the hydrogen precipitates as "in-plane" (circumferential) zirconium hydride platelets, especially when the material is in the cold worked and stress relieved (CWSR) condition. Upon removal of the rods from the spent fuel pool for placement in dry storage, there is a concern that the drying operations, which consist of heating the rods to high temperature, could cause hydrides to dissolve and reprecipitate in a reoriented direction as "out-of-plane" (radial) hydrides with platelet faces parallel to the hoop direction of the cladding tube. This phenomenon has been known for a long time; that is, when cooled under a sufficiently high applied stress the dissolved hydrogen can precipitate as reoriented hydrides perpendicular to the applied stress[1] [3–5]. The occurrence of hydride reorientation depends on the external stress applied to the cladding caused by the initial fill gas and gaseous fission products in the spent nuclear fuel rod.

Although hydride reorientation is influenced by properties inherent to the alloy such as the degree of cold work, grain microstructure, texture, and others [5–10], and by parameters related to the specific thermo-mechanical treatments, such as maximum temperature, dwell time at maximum temperature, cooling rate, and number of thermo-mechanical cycles, the critical parameter is the threshold stress (σ_{th}) [11–18]. Most of the data on the threshold stress for hydride reorientation in zirconium-base alloys available in the literature has been obtained from uniaxial tension tests [6–8,16–24]. However, during the vacuum drying process, the thin-wall nuclear fuel cladding tubes are subjected to internal gas pressure and/or pellet clad mechanical interaction (PCMI) that create stress biaxiality ratios (σ_2/σ_1) ≥ 0.5, much higher than that for uniaxial tension ($\sigma_2/\sigma_1 = 0$) [25]. Although the data are

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[1] In the literature, various definitions have been employed to define the external stress to cause hydride reroeintation. The definitions in this study are given in the experimental and computational procedures section of this paper, see pages 12 and 13.

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available from studies using internally-pressurized tubes [12,13,15] that induce stress states similar to vacuum-drying, no systematic study of the effect of stress state on the threshold stress has been performed. Existing data does suggest that there may be an effect of stress state on the threshold stress for hydride reorientation. For example, data from unirradiated, cold worked and stress relieved (CWSR) Zircaloy-4 shows higher threshold stress values under uniaxial tension ($140 < \sigma_{th} < 200$ MPa) than under biaxial stress state ($\sigma_2/\sigma_1 = 0.5$) when $20 < \sigma_{th} < 115$ MPa [11,13,15,19,20,23,24].

The purpose of this study is to systematically examine and quantify the role of stress state on hydride reorientation. In this work the threshold stress to reorient hydrides is determined using specifically designed test specimens that induce stress states ranging from uniaxial tension ($\sigma_2/\sigma_1 = 0$) to near-equibiaxial tension ($\sigma_2/\sigma_1 = 0.8$). The threshold stress is obtained in each case by matching the stress states calculated by finite element analysis of the test specimens under load to the hydride microstructures (and specifically the presence of reoriented hydrides) at that specific location. The study uses Zircaloy-4 sheet containing nominally ≈180 wt ppm hydrogen as a model material subjected to identical two-cycle thermo-mechanical treatments that bound conditions of vacuum drying of spent nuclear fuel. The data thus obtained isolates the stress state effect from other parameters that may affect hydride reorientation.

2. Experimental and computational procedures

Because of experimental advantages of the use of a flat sheet compared to thin-wall tube, this study utilized flat 0.67 mm thick Zircaloy-4 sheet obtained from Teledyne Wah-Chang in the cold-worked stress relieved (CWSR) condition. As described elsewhere [26], the CWSR Zircaloy-4 sheet exhibited a strong crystallographic texture. Importantly, the Kearns factors [3] of the CWSR sheet ($f_N = 0.59$, $f_R = 0.05$, and $f_T = 0.31$ in the normal, longitudinal/rolling, and transverse directions of the sheet, respectively) are similar to those reported for unirradiated CWSR Zircaloy-4 cladding tubes (0.58, 0.10, and 0.32, respectively) [27], where the hoop direction of the tube corresponds to the orientation transverse to the rolling direction of the sheet. Therefore, the texture of the Zircaloy-4 sheets used in this study is similar to the typical texture of Zircaloy-4 cladding tubes [28], i.e., the basal planes tend to align with their poles inclined approximately $\pm 30^\circ$ away from the normal of the tube surface and oriented toward the transverse direction [29].

The stress-relieving and hydrogen gas charging operations were conducted simultaneously in the following manner. Prior to hydrogen charging, the native oxide layer was removed from the surface using an acid solution, and the specimen was then coated with a 200 nm thick nickel layer to forestall the reformation of oxides that would impede hydrogen ingress. The specimen was put into a quartz tube furnace to which a controlled amount of hydrogen-argon gas mixture (12.5% H_2 and 87.5% Ar) was subsequently introduced into the chamber (initially at a pressure of less than 13.3 μPa) containing the cold-worked (CW) specimen at 500 °C. The temperature of the chamber was maintained for 2 h to simultaneously induce stress relief, hydrogen absorption, and the homogenizing of the hydrogen distribution throughout the sample thickness. After completion of stress anneal and hydrogen charging, the specimens were slowly furnace-cooled to 25 °C. The hydrogen concentration of each sample was determined using hot vacuum extraction by Luvak Inc. The Zircaloy-4 specimens in this study were hydrogen charged to approximately 180 wt ppm. (max. 197 wt ppm and min 157 wt ppm). This hydrogen content was chosen so that on the one hand it was sufficiently low that the hydrides present at low temperature completely dissolved at high temperature and on the other hand high enough to cause hydride precipitation at a high enough temperature where the hydrogen was sufficiently mobile.

The tensile properties of the CSWR Zircaloy-4 sheet were obtained by performing tensile tests at temperatures in the range of 25–450 °C. The mechanical response of the Zircaloy-4 sheet after it plastically yields was modeled by power law hardening; the flow parameters were determined from uniaxial tension sample experiments and from studies performed previously using similar materials [18,26,28,30–34]. The strain-hardening exponent of the sheet used in this study ($n = d\ln\sigma/d\ln\varepsilon = 0.014$ at 25 °C, 0.023 at 300 °C, and 0.022 at 400 °C) is significantly smaller than that of standard cladding ($n \cong 0.06$ at 300 °C) [31]. In fact, the strain hardening behavior of this CWSR sheet material is similar to that of irradiated cladding [35].

Three types of test specimens were designed and used to determine the effect of stress state on hydride reorientation: (1) tapered uniaxial tension, (2) "plane-strain" tension [31], and (3) "near-equibiaxial" tension specimens, As shown in Fig. 1, these samples were used to determine the threshold stresses for the onset of "out-of-plane" (radial) hydride precipitation or hydride reorientation. To simulate the hydride reorientation threshold stress for spent nuclear fuel cladding, Zircaloy-4 sheet tensile specimens were machined with the load axis along their long-transverse direction which corresponds to the circumferential (or hoop) direction of nuclear fuel cladding tubes.

Finite element analyses were performed to determine the stress state distributions within the gauge sections of the double-edge notched ("plane-strain" and "near-equibiaxial" tension) samples. Because of the small thickness of the sheet material, two-dimensional finite element computations were performed under the assumptions of (i) plane stress through the specimen thickness, (ii) specimen plastic deformation (if it occurred) with flow behavior at ~400 °C (close to the onset of hydride precipitation), and (iii) isotropic mechanical properties obtained in MATPRO [35].

The finite element analysis of "plane-strain" tension sample in the elastic region indicates that the stress biaxiality ratio across the gauge width decreases from 0.57 near the mid-point between the two notches to 0 (uniaxial tension) near the notch. While most of the specimen deforms elastically, a small plastic zone may be created near the notch under the loading conditions obtained in this study.

In order to investigate the stress state effect for stress biaxiality ratios greater than 0.57, the double edge-notched plane-strain specimen was modified by the addition of two "stress state" holes as shown in Fig. 1c. Under load, the specimen creates a region of near-equibiaxial stress state ($\sigma_1/\sigma_2 \approx 0.83$). Fig. 2 shows the results of finite element calculation of the stress state in the near-equibiaxial tension sample, in terms of the principal stress distributions along both the gauge width and the gauge length when an average gauge section stress of 210 MPa is applied. The notches and the holes produce a range of multiaxial stress states that vary with location within a sample under load. Specifically, a near equibiaxial stress state of ≈0.83 is achieved at the center of the specimen sample; see Fig. 2c. A similar high degree of stress biaxiality is also computed along a line between the centers of the stress state holes as shown in Fig. 2d. In addition, Fig. 2b shows that there is an extensive region near the center of the gauge section in which the major principal stress is nearly constant while the minor principal stress is decreasing, resulting in a range of stress states that depend on specimen location.[2]

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[2] Note that for these plane-stress analyses, the minimum principal stress is always the through-thickness stress, which has a value of zero, and thus we adopt the notation for the in-plane stresses as the "major" and "minor" stresses; both are principal stresses and the major stress is also the maximum principal stress.