Characterization of local hydride re-orientation in high burn-up PWR fuel rods induced by high pressure at high temperatures

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ABSTRACT

Hydride re-orientation in high burn-up PWR fuel rods was induced by high pressure at high temperatures. The high-burnup specimens were sectioned from PWR rods taken from a 15×15 assembly of the H.B. Robinson (HBR) Unit 2 reactor. Out-of cell benchmark tests performed on unirradiated hydrided Zircaloy-4 specimens were conducted to determine the appropriate temperature, pressure, cooling rate, and number of cooling cycles for the reorientation of the irradiated in-cell specimens. The in-cell hydride reorientation tests were performed using high-burnup fuel specimens under a hoop stress ≈145 MPa at 400°C. The specimens were heated to the target temperature of 400°C, held for 3 hours, cooled at 1°C/min to 170°C,and then heated at 1°C/min to the target temperature again for five cycles. Post test metallographic examinations showed that a significant amount of radial hydrides were induced in the HBR fuel rods. The length of radial hydride was up to 60 μm. For unirradiated materials, the ductility of the radial hydride treated specimens is significantly reduced as compared to the as-hydrided specimens having the same hydrogen concentration (≈300 wppm in this work). The mechanical testing on irradiated fueled samples with hydride reorientation experiments have been performed, and will be reported separately in the near future.

I. INTRODUCTION

Hydrogen embrittlement of zirconium alloys is a phenomenon of interest in the United States due to spent nuclear fuel (SNF) remaining in storage for longer time frames than initially envisioned. Normal operation of nuclear fuel in a reactor results in the formation of a waterside corrosion layer and the introduction of hydrogen into the zirconium cladding, which can cause cladding ductility and failure energy to decrease [1, 2]. The processes used during the drying of SNF as it is transferred from wet storage to dry storage can expose the SNF cladding to temperatures and pressure-induced tensile hoop stresses high enough that could allow radial hydrides to precipitate during subsequent cooling. These radial hydrides could provide an additional embrittlement mechanism as the cladding temperature decreases below the ductile-to-brittle transition temperature. To simulate this behavior, unirradiated Zircaloy-4 samples (hydrided by a gas charging method) and defueled irradiated cladding has been tested under high pressures at high temperatures to generate radial hydrides at several laboratories [3-6]. In this experiment, we expanded on the earlier work [7] by performing hydride reorientation tests using fueled high-burnup irradiated SNF rod segments. The post-test examinations were conducted to characterize the microstructure of hydride reoriented samples.

II. EXPERIMENTAL

The high-burnup specimens, sectioned from PWR rods taken from a 15×15 assembly of the H.B. Robinson (HBR) Unit 2 reactor, have been used for this work. This fuel had operated for seven cycles and reached a rod-average of 67 GWd/MTU. The nominal fuel pellet dimensions are 9.06 mm diameter x 9.93 mm height. The cladding is cold-worked/stress-relieved, 10.77 mm outer diameter (OD) × 9.25 mm inner diameter (ID), with a nominal tin content of 1.42%. A detailed description of the HB R fuel cladding dimesons and characterization. is given by the plant operator and fuel vender [8].

The hydride reorientation (HR) system consists of a high pressure piping system and test chamber within a programmable crucible furnace. The system was installed into hot cell. The HBR fuel samples for hydride reorientation tests were prepared in the irradiation fuel examination laboratory. The specimens were sectioned with a low speed saw to 152 mm long pieces, and the surface oxide layer and fuel were removed at approximately 12.5 mm from each end where end-caps were then welded. After fabrication, the specimen was assembled into a holder within the test chamber, pressurized with Ar gas such that it reached a maximum hoop stress of 145 MPa at T=400°C, and was then exposed to thermal cycling to increase the number of radial hydrides. The thermal cycles consisted of heating the specimen to a target temperature of 400°C, holding at this temperature for 3 hours, cooling at 1°C/min to 170°C, and then heating at 1°C/min to target hold temperature 400°C again for five cycles. The sample was furnace cooled from 170°C to room temperature for the last cycle. All in-cell tests had the same temperature profiles. Figure 1 shows the temperature history of the first in-cell hydride reorientation test HR1 with a HBR high-burnup sample.

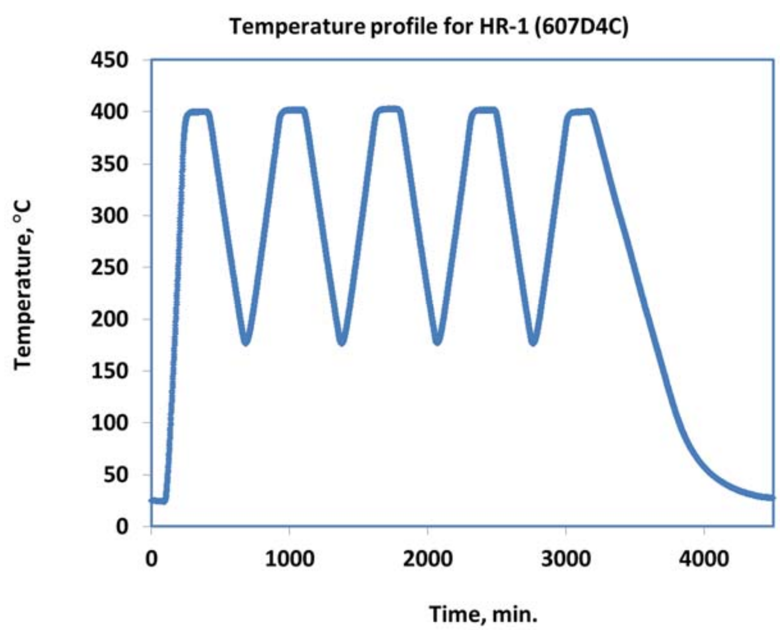
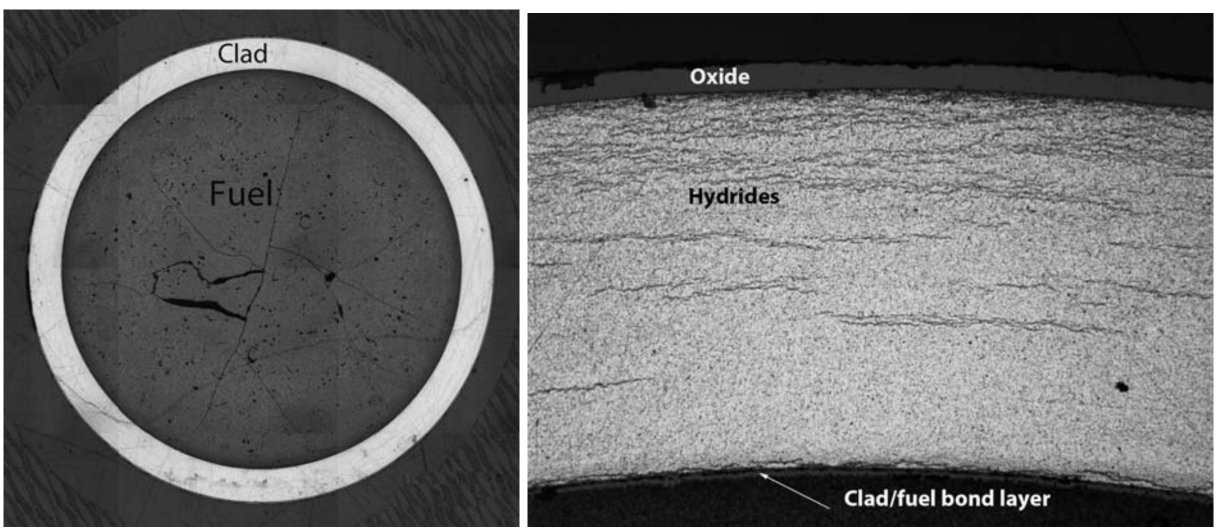


Figure 1. Sample temperature as a function of time for in-cell hydride reorientation test.

III. RESULTS

Pre-test Characterization

Before the HR tests, characterization was performed by metallography to determine the fuel, fuel-cladding bond, corrosion layer, and hydride morphology. Figure 2a shows a low magnification image of the cross-section of the high-burnup fuel used for reorientation tests. The fuel morphology in Figure 2a reveals the typical start-up and shut-down cracks. Figure 2b is a high magnification image within one circumferential sector of the fuel and clad shown in Figure 2a. The fuel-cladding bond appears to be well developed. The bond thickness is ≈10 μm. According to Une et al. [9], the bond layer is primarily ZrO2 with some UO2 in solid solution. The corrosion layer (labeled as Oxide in Figure 2b) thickness on sample OD surface was measured to be 55 μm. Within the layer, occasional radial cracks can be seen, but oxide spallation is not prevalent. Circumferential hydrides (dark lines in Figure 2b) were observed. The density of hydrides near the OD of the cladding is high and forms a hydride rim. Total hydrogen content in the cladding was estimated to be ≈400 ppm, based on the corrosion layer thickness.

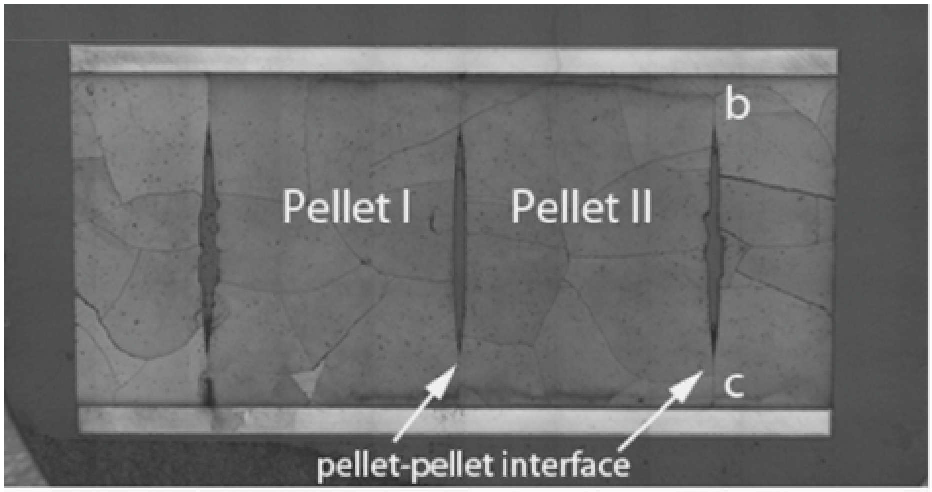


(a) (b)

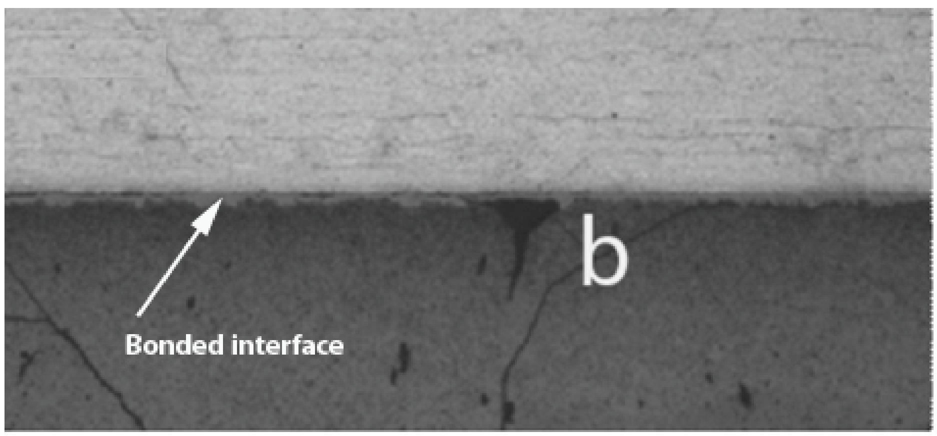
Figure 2. (a) High-burnup HBR fuel morphology for the cross section of fuel rod segment used for hydride reorientation tests. (b) High magnification image within one circumferential sector of the clad shown in Figure 2a, showing the circumferential hydrides across the wall thickness.

Post-test characterization

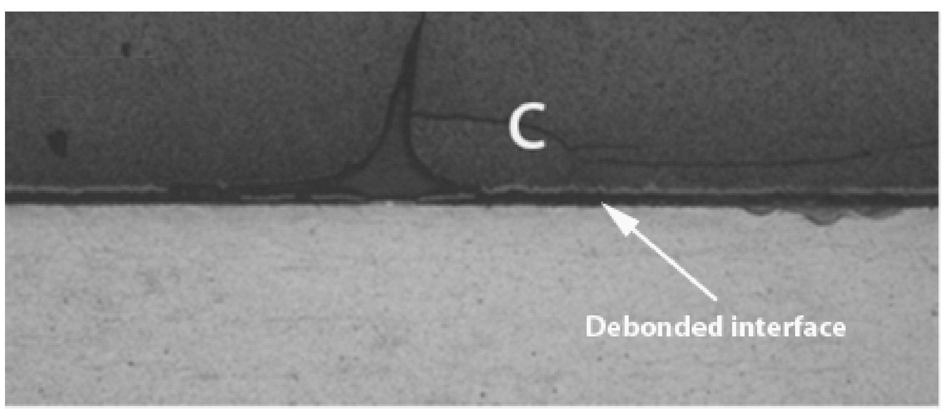
After HR tests, metallographic examinations were performed in both cross sectional and longitudinal directions. Figure 3 shows images of a post-HR test sample in the longitudinal direction. Gaps between the fuel pellets were clearly seen in Figure 3a. High magnification images show that there are two kinds of fuel-clad interfaces: a bonded fuel-clad interface (see Figure 3b) and a de-bonded interface (see Figure 3c). The gap between the clad and fuel is about 15-20 μm for the de-bonded interface. The impact of the gap to the mechanical property needs to be investigated further.



(a)



(b)



(c)

Figure 3. (a) Low magnification image showing the pellet's pellet-pellet interface of a post-HR test sample in longitudinal direction. (b) High magnification image of Area b in Figure 3a showing the bonded clad-fuel interface. (c) High magnification image of Area c in Figure 3a showing the de-bonded clad-fuel interface (gap between fuel and clad is about 15-20 μm).

Figure 4 shows images of a post-HR test sample in the cross sectional direction. It reveals that our hydride reorientation procedure resulted in large amounts of radial hydrides with a uniform distribution of circumferential hydrides across the Zircaloy-4 wall. From the image in Figure 4, it is clear that the length of radial hydrides near the inner surface is longer than those near the outer surface. Ring compresion tests indicate that the ductility of defueled high-burnup or pre-hydrided Zr-alloy cladding can be dramatically reduced after the radial hydrides are introduced by the hydride reorientation treatment [4,5]. Our tests were performed with high-burnup fuel samples. The impact of radial hydrides on the mechanical properties of the specimens needs to be investigated further.

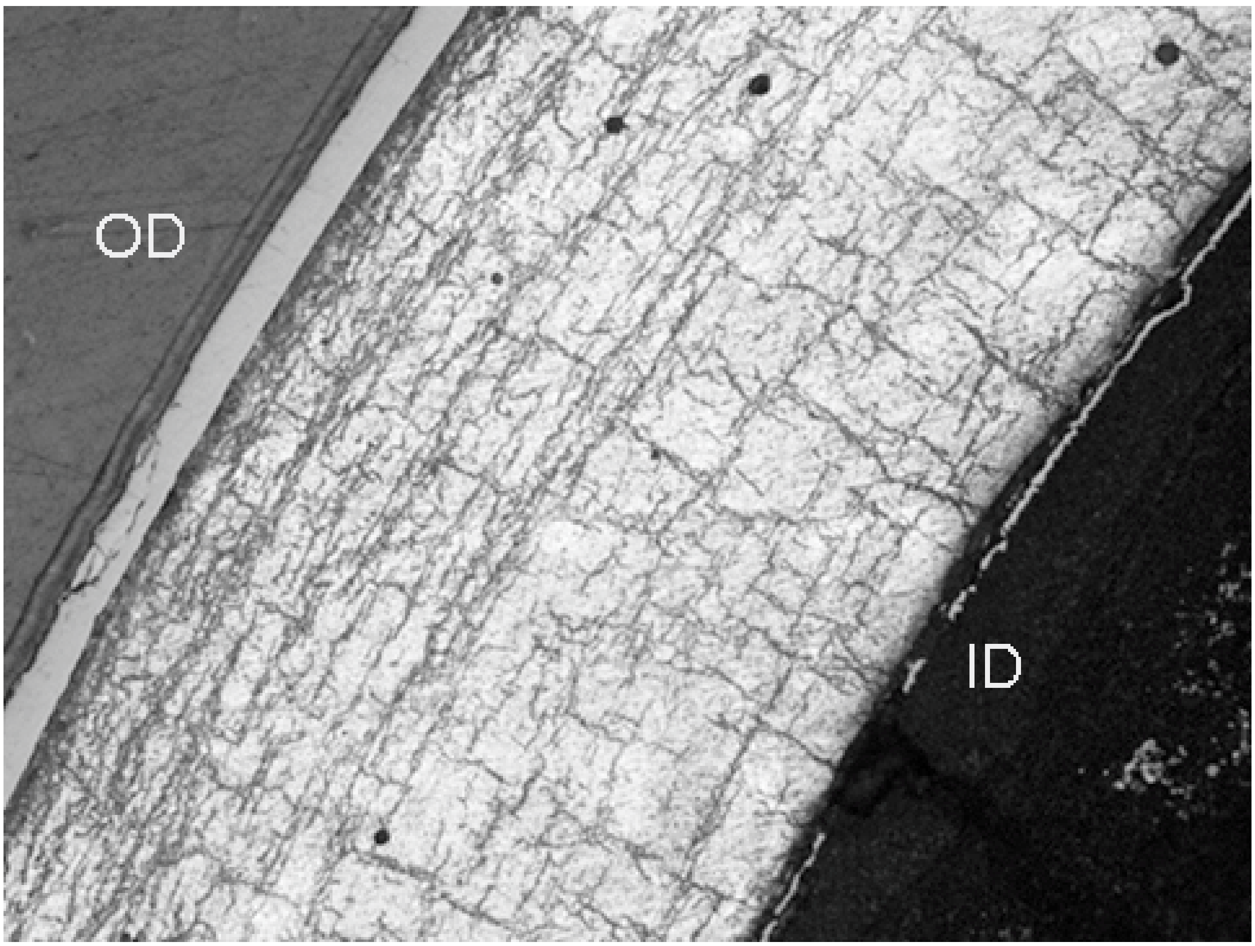


Figure 4. Micrographs showing radial hydrides after the HR test at 400°C for the maximum pressure P=144 MPa.

IV. SUMMARY

(1) Hydride reorientation in the high-burnup PWR fuel rods was successfully induced by high pressure at high temperatures under the hoop stress ≈145 MPa at 400°C. The specimens were heated to target temperature, held for 3 hours, cooled at 1°C/min to 170°C, and then heated at 1°C/min to target temperature 400°C again for five thermal cycles.

(2) Pre-hydride reorientation test characterization of the high-burnup sample shows a large amount of circumferential hydrides in cladding. The density of hydrides near the OD is high, and formed a hydride rim. Hydrogen content in the cladding was calculated to be ≈400 ppm, based on the corrosion layer thickness. The fuel-cladding bond was observed and its thickness is ≈10 μm.

(3) Metallographic examinations of post-hydride reorientation tests in the cross sectional direction showed that a significant amount of radial hydrides were induced in the HBR fuel rods. The length of radial hydrides near the inner surface is longer than those near the outer surface.

(4) Metallographic examinations of post-hydride reorientation tests in the longitudinal direction showed two kinds of clad/bond interfaces: the bonded fuel-clad interface and the de-bonded interface. The gap between the clad and fuel is about 15-20 μm for the de-bonded interface. The impact of gap to the mechanical properties needs to be investigated further.

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