Ductile-Brittle Transition Temperature for High Burnup Zircaloy-4 and ZIRLO™ Cladding Alloys Exposed to Simulated Drying Storage Conditions

Nuclear Engineering Division

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EXECUTIVE SUMMARY

Structural analyses of high-burnup fuel storage and transport casks require cladding mechanical properties and failure limits to assess fuel behavior. Properties and limits currently used are based on as-irradiated cladding samples from fuel rods discharged from reactors and stored in pools. Such cladding has reduced ductility due to irradiation effects and the presence of circumferential hydrides. However, as-irradiated cladding is not brittle. Pre-storage drying-transfer operations and early stage storage subject cladding to higher temperatures and much higher pressure-induced tensile stresses than experienced in-reactor or during pool storage. If these are high enough, then radial hydrides could precipitate during pre-storage operations (slow cooling) and during dry-cask storage (very slow cooling). Radial hydrides represent an additional embrittlement mechanism that can reduce cladding failure limits during storage as the cladding temperature decreases below a ductile-to-brittle transition temperature (DBTT). As such, the effects of radial hydrides must be included in structural analyses when the cladding temperature is below or marginally above the DBTT.

A test procedure was developed to simulate the effects of elevated temperatures, pressures and stresses during transfer-drying operations and early storage, as well as cooling slowly under decreasing pressures and stresses during both drying and storage. Pressurized and sealed rodlets were heated to the target temperature for a prescribed hold time and slow cooled at a computer-controlled linear temperature rate. The procedure was applied to both non-irradiated/pre-hydrided (PH) and high-burnup Zircaloy-4 (Zry-4) and ZIRLO™ using the NRC-recommended 400°C maximum temperature limit and a 5°C/h cooling rate. Following drying-storage simulation, samples sectioned from rodlets were subjected to ring-compression tests (RCTs). The RCT was used as a ductility screening test and to simulate pinch-type loading that would occur during cask transport. RCT load-displacement results were used to determine the DBTT for each cladding alloy as a function of the drying-storage hoop stress at 400°C. A strong correlation was found between the extent of radial hydride formation across the cladding wall and the extent of wall cracking during RCT loading.

Prior to testing high-burnup cladding, a large number of tests were conducted with PH Zry-4 and ZIRLO™. It was observed that PH cladding with the same hydrogen content (350 to 650 wppm) was a poor surrogate for high-burnup cladding with respect to extent of radial-hydride precipitation and ductility reduction following simulated drying-storage. This was due to the uniform pre-test distribution of hydrides and the high-ductility metal matrix characteristic of PH cladding. However, test results for PH cladding were useful in defining a metric for the extent of radial hydride precipitation and in identifying the higher susceptibility of cladding with lower hydrogen contents (<300 wppm) to radial hydride precipitation.

Zry-4 and ZIRLO™ cladding from commercial reactor fuel rods irradiated to high-burnup were used as test samples. Hydrogen contents ranged from 520 to 620 wppm for high-burnup Zry-4 and 350 to 650 wppm for high-burnup ZIRLO™.

The extent of radial-hydride precipitation was characterized by the radial-hydride continuity factor (RHCF): the percentage of cladding wall within a 0.15-mm circumferential band containing continuous radial and circumferential hydrides along which an unstable crack may propagate. For high-burnup Zry-4, RHCF values were relatively low (9% and 16%) compared to values (30% and 65%) for high-burnup ZIRLO™ subjected to peak drying-storage hoop stress of 110 MPa and 140 MPa at 400°C.

RCT samples were sectioned from high-burnup rodlets following the drying-storage simulation. The following figure shows RCT offset strain vs. test temperature for 18 high-burnup rings, along with trend curves for offset strain as a function of test temperature. DBTT values for high-burnup Zry-4 were <20°C and 55°C, respectively, for 110-MPa and 140-MPa peak hoop stresses. The corresponding DBTT values for high-burnup ZIRLO™ were 125°C and 185°C. It was observed that susceptibility to radial-hydride formation and radial-hydride-induced embrittlement is highly dependent on cladding material, irradiation conditions, and pre-drying distribution of hydrides across the cladding wall, as well as peak drying-storage hoop stress.

RCT ductility data vs. test temperature for high-burnup Zry-4 and ZIRLO™ cladding materials following simulated drying-storage. Offset strains < 2% imply brittle failure.

Results in this report were used to generate the Journal of Nuclear Materials (JNM) article: M.C. Billone, T.A. Burtseva, R.E. Einziger, J. Nucl. Mater. 433 (2013) 431-448. The JNM article is the primary reference, and this report should be referenced only for supporting material.

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1. Introduction

Fuel rods with burnup levels >45 GWd/MTU are classified by the Nuclear Regulatory Commission (NRC) as high burnup. Relative to lower-burnup rods, high-burnup rods are characterized by increased: decay heat following reactor discharge, internal gas pressure, cladding corrosion layer thickness, and cladding hydrogen content. Under pool-storage conditions following reactor discharge, hydrogen in the spent fuel cladding will be in the form of precipitated hydride platelets, which are primarily oriented in the circumferential direction. Circumferential hydrides, in combination with hardening due to irradiation effects, will decrease cladding ductility in response to axial and hoop loads, but high-burnup cladding retains some ductility [1]. At elevated temperatures ($\leq 400^\circ\text{C}$) during drying-transfer operations and the early stage of dry-cask storage, some of the hydrogen goes into solution (e.g., about 200 wppm at 400°C [2,3]) and the pressure-induced cladding tensile hoop stress is high relative to in-reactor and pool-storage conditions. During cooling under tensile hoop stress, some of the dissolved hydrogen will precipitate in the radial direction across the cladding wall if the hoop stress is high enough. After cooling to about 200°C , most of the dissolved hydrogen re-precipitates as hydrides. Further cooling during storage may result in radial-hydride-induced embrittlement. The temperature at which embrittlement occurs is the ductile-to-brittle transition temperature (DBTT). Radial-hydride-induced embrittlement and the corresponding DBTT may have a significant effect on cladding mechanical properties used in structural analyses for storage and transport casks. In particular, hoop tensile properties and failure limits determined for as-irradiated cladding samples (e.g., Ref. 1 for Zircaloys) are not governing for cladding with a significant fraction of radial hydrides at temperatures below or marginally above the DBTT.

In Interim Staff Guidance – 11, Revision 3 (ISG-11 Rev. 3), the NRC recommends a peak cladding temperature of 400°C for drying-storage operations and storage of spent-nuclear-fuel (SNF) storage and transport casks containing high-burnup fuel [4]. One concern for high-burnup fuel cladding was the possible precipitation of radial hydrides, which could embrittle cladding in response to tensile hoop-stress loading. The limits established in ISG-11 Rev. 3 relied on data available prior to 2002, which were primarily for low-burnup and non-irradiated/pre-hydrated (PH) Zircaloy-4 (Zry-4) – a cold-worked/stress-relieved-annealed (CWSRA) alloy. Most of the data were based on tests with conditions with limited relevance (e.g., fast cooling rates $>25^\circ\text{C/h}$ and constant stress during cooling) to SNF drying and storage. At the time ISG-11 Rev. 3 was issued, it was recognized that data for high-burnup fuel cladding alloys were needed to determine the extent of radial-hydride formation and radial-hydride-induced embrittlement under conditions closer to those existing during drying-transfer operations and storage. These conditions include slow cooling rates ($\leq 5^\circ\text{C/h}$) and decreasing hoop stress during cooling.

The fast cooling rates (e.g., 25°C/h to 600°C/h reported in Refs. 5-9) used to test non-irradiated PH and irradiated cladding tend to result in shorter radial hydrides and lower DBTT values because the hydrogen-diffusion path length is limited. Also, non-irradiated PH cladding alloys can have lower DBTT values following cooling under tensile stress than would be relevant to the same cladding alloys irradiated to high burnup and subjected to cooling under drying and storage conditions. PH cladding differs from high-burnup cladding in two important areas: (a) it consists of brittle hydrides in a high-ductility metal matrix and (b) methods (e.g., gaseous charging with temperature cycling) commonly used to pre-hydrate cladding result in a relatively uniform distribution of hydrides across the cladding wall. However, high-burnup Zry-4 and ZIRLO™ with about 300 to 650 wppm hydrogen have highly non-uniform distributions of hydrides across the cladding wall: relatively dense hydride rim at the cladding outer surface and decreasing hydride density from below the rim to the cladding inner surface. In general, the average hydrogen content below the high-burnup cladding rim is <200 wppm independent of the total hydrogen content. During one-cycle drying at 400°C, high-burnup Zry-4 and ZIRLO™ would have ≈200-wppm hydrogen in solution and negligible circumferential hydrides below the rim. The presence of a high density of circumferential hydrides in 300 to 650 wppm PH cladding at 400°C biases the precipitation of additional circumferential hydrides and limits the number and length of radial hydrides that precipitate during cooling under stress.

Several metrics have been used to characterize the extent of radial-hydride formation. The radial hydride fraction (RHF, percent of hydrides with orientation within 40° or 45° of the radial direction) is the most commonly used metric [6]. The hydride continuity coefficient (HCC) has been used by Canadian researchers to characterize the extent of radial-hydride formation in Zr-2.5%Nb pressure tubes [10,11]. It is determined by projecting the radial hydrides within a 0.11-mm circumferential band onto the radius across the metal wall. The net length of these projected hydrides is normalized to the metal wall thickness to determine HCC. Neither RHF nor HCC is sufficient for predicting crack growth under RCT loading. For example, rings with short, discontinuous radial hydrides and rings with long, continuous hydrides might have RHF and HCC values of 100%, but the latter would be more prone to significant cracking during RCT loading. As a result, a new metric was developed, the radial-hydride-continuity factor (RHCF), to better correlate hydride orientation and morphology with crack initiation and growth.

This report describes a procedure for consistently measuring the extent of hydride reorientation and its effect on the DBTT. Data supporting the effects of irradiation conditions, cladding material, and stress level on the DBTT are reported and discussed.

2. Materials and Test Conditions

2.1 Materials

As-fabricated (AF) 15×15 Zry-4 (vintage and modern) and 17×17 ZIRLO™ were provided by AREVA and Westinghouse, respectively. These materials were used to generate baseline data on drying-storage conditions that result in radial-hydride precipitation in non-irradiated cladding and the effects of radial hydrides on cladding ductility. The vintage Zry-4 and ZIRLO™ cladding materials were selected by the vendors to have geometries, chemistries, and

thermo-mechanical treatments similar to what was used for the high-burnup fuel-rod cladding provided to Argonne. Table 1 lists the relevant properties of these three cladding materials.

Table 1

Geometry and chemistry of as-fabricated cladding; HBR = H.B. Robinson and NA = North Anna

Cladding Material	Outer Diameter, mm	Wall Thickness, mm	Sn Content, wt%	Nb Content, wt%	O Content, wt%	Comment
Modern 15×15 Zry-4	10.91	0.67	1.29	---	0.124	---
Vintage 15×15 Zry-4	10.77	0.76	1.29	---	0.136	1.40 wt% Sn for cladding irradiated in HBR
17×17 ZIRLO™	9.50	0.57	0.99	0.98	0.120	Similar to cladding irradiated in NA

AF cladding samples were pre-hydrided by gaseous charging to hydrogen content values in the range of 100 to 1000 wppm. Segments were heated to 350-400°C in flowing or static H₂-He gas with H₂ contents of 4%, 30%, or 100%. For most runs, segments were heated to 400°C, and subjected to static 30% H₂ gas mixture and to five thermal cycles prior to annealing at 400°C for 24-36 h in high-purity Ar and cooling. Specimen preparation evolved to increase the rate of hydrogen pickup and to minimize hydrogen concentration gradients in the axial and circumferential directions: pickling of surfaces with ≈1% HF-containing acid mixture, wrapping samples in high-purity Al foil, and finally subjecting only the outer surface to pickling, Al foil, and hydrogen flow.

Two 15×15 Zry-4 segments were sectioned and defueled from fuel rod E02 irradiated to 67 GWd/MTU in the HBR Unit 2 Reactor. Operating conditions and post-irradiation characterization data for these fuel rods are given in Refs. 12-13. Each segment was about 76-mm long. Axial locations for the segments were approximately 0.4 m above the fuel mid-plane (605C6) and 0.7 m above the fuel mid-plane (605C2). Both segments were sectioned from regions away from grid-spacer locations and within the uniform burnup region of the rod. Extensive characterization of sibling rods irradiated in the same lead test assembly was performed for the NRC-sponsored loss-of-coolant accident (LOCA) research program conducted by Argonne [14]. Based on sibling-rod characterization, the expected corrosion layer thickness (δ_{ox}) and hydrogen content (C_H) were: (a) 75 μ m and 550 wppm for segment 605C6 and (b) 100 μ m and 750 wppm for segment 605C2. Figure 1 shows the hydride morphology, orientation, and radial distribution for sibling rod F07 at the fuel mid-plane (Fig. 1a) and at about 0.65 m above the fuel mid-plane (Fig. 1b) [14]. δ_{ox} increased from 71±5 μ m to 95±5 μ m and C_H increased from 550±100 wppm to 740±110 wppm with the 0.65-m increase in axial elevation. The “±” C_H values represent one-standard deviation in data for quarter-ring samples at the same elevation and for rings at multiple axial locations within a short span. Cladding hydrides were in

(a) Fuel mid-plane

(b) 0.65 m above fuel mid-plane

Fig. 1. Hydride distribution and morphology in Zry-4 cladding from high-burnup fuel rod F07 irradiated to 64 GWd/MTU in one of the H. B. Robinson reactors: (a) fuel mid-plane (550 wppm hydrogen); (b) 0.65 m above fuel mid-plane (740 wppm hydrogen). The corresponding δ_{ox} values were 71 and 95 μm [14].

the form of platelets oriented in the circumferential direction. Although most of the hydrogen was contained in the outer third of the Zry-4 wall, the hydride rim was not as localized as anticipated for high-burnup fuel cladding. This may have been due to the relatively low temperature gradient across the cladding wall during the burnup increase from 45 to 64 GWd/MTU. The fuel enrichment was only 2.9% U-235 and the rod was irradiated for 7 reactor cycles (about 2300 effective full-power days). The high number of reactor startups and shutdowns may have also contributed to the observed radial distribution of hydrides. δ_{ox} (74 ± 5 μm), C_H (550 ± 80 wppm), and hydride morphology for fuel rod F07 at 0.32 m above the fuel mid-plane were essentially the same as they were at the fuel mid-plane location (Fig. 1a) [14]. Thus, Figs. 1a and 1b should be representative of high-burnup Zry-4 segments 605C6 and 605C2, respectively.

Three 17×17 ZIRLO™ segments, each about 80-mm long, were sectioned from fuel rod AM2-L17 irradiated for 4 cycles in the North Anna reactors (first 3 cycles in Unit 1 and 4th cycle in Unit 2) to 70 GWd/MTU. Some information about fuel rod characterization following the 3rd irradiation cycle can be found in Refs. 15-16. The fuel enrichment was about 4% U-235, which suggests that the time-averaged fuel thermal power and cladding temperature gradient were higher for this rod than for the Zry-4 rod described above. Axial locations of the segments were from about 0.64 m (648C), 0.72 m (648D), and 1.20 m (648G) above the fuel mid-plane. These segments were located within the uniform burnup region of the fuel rod away from grid-spacer locations. Two other segments from neighboring axial locations of the same rod were well characterized for the NRC LOCA program [14,17]. Axial locations of these characterized segments were 0.44 m (648A) and 1.09 m (648F) above the fuel mid-plane. δ_{ox} increased from 26 ± 1 μm at 0.44 m to 42 ± 3 μm at 1.09 m above the fuel mid-plane. δ_{ox} values were measured at 8 (648A) and 16 (648F) circumferential orientations. Consistent with the increase in δ_{ox} , C_H increased from 320 ± 30 wppm to 620 ± 140 wppm.

Figure 2 shows the hydride morphology and distribution for high-burnup ZIRLO™ at 0.44 m above the fuel mid-plane of fuel rod AM2-L17 [17]. The hydride rim was well developed for 320-wppm hydrogen, and the average hydrogen content between the rim and the cladding inner surface appears to be relatively low (<150 wppm). Figure 2a shows the typical circumferential hydride orientation (in 7 of 8 micrographs). Figure 2b shows a local region with a mixture of circumferential and radial hydrides. The hydride morphology and distribution at a higher axial elevation (1.09 m above the fuel mid-plane) are shown in Fig. 3 for the same fuel rod [14]. The increase in average C_H (from 320 to 660 wppm) appears to be due to the increase in hydride-rim thickness. The hydride distribution and hydrogen content below the rim look very similar to what is observed at the lower axial location (Fig. 2). Figure 3a shows the circumferential location at which the hydride rim is thickest ($\approx 70\ \mu\text{m}$) while Fig. 3b shows the location at which the hydride rim is thinnest ($\approx 40\ \mu\text{m}$). Both images are from the same cross section at the 1.09-m elevation. Based on examination of images taken at 8 circumferential orientations, the hydride-rim thickness varied monotonically, rather than randomly, with circumferential orientation. This systematic variation suggests a circumferential variation in cladding temperature and perhaps heat flux during irradiation. These metallographic results are consistent with the measured variation in C_H levels (500 to 840 wppm) for 4 quarter-arc segments of a ring adjacent to the cross section imaged in Fig. 3. However, more localized hydrogen measurements would probably have led to larger variations than recorded for the quarter-arc segments.

As two of the ZIRLO™ segments (648C and 648D) were slightly higher in elevation than the 648A segment, these experimental segments should have >320 wppm C_H . Segment 648G was slightly higher in elevation than 648F. As such it is expected to have >620 wppm C_H .

Both high-burnup ZIRLO™ and Zry-4 cladding exhibited the same trends with regard to increased corrosion-layer thickness and hydrogen content with increased axial elevation. It is interesting to note that cladding with less than about 350 wppm hydrogen has a relatively small circumferential variation (about 10%) in C_H , while cladding with >500 wppm hydrogen has a very significant circumferential variation in C_H due to the variation in hydride rim thickness.

The characterization results presented in this section are for cladding from fuel rods discharged from the reactor and stored in the spent-fuel pool. They represent the initial conditions of high-burnup fuel cladding samples used in this test program prior to subjecting the cladding to simulated drying-storage temperature and pressure histories. Characterization results for high-burnup cladding following simulated drying-storage are presented in Section 4 for Zry-4 (605C6 and 605C2) and ZIRLO™ (648C, 648D, and 648G) segments.

(a) Circumferential hydrides

(b) Circumferential and radial hydrides

Fig. 2. Hydride distribution, orientation, and morphology at 0.44 m above the fuel mid-plane for ZIRLOTM cladding from fuel rod irradiated to 70 GWd/MTU: (a) circumferential location with typical (7 of 8 images) circumferential hydrides and (b) circumferential location with region of circumferential and radial hydrides. δ_{ox} was 26 μm , and C_H was 320 wppm for an adjacent ring [17].

(a) Thick hydride rim at about 1.1 m above the fuel mid-plane

(b) Thin hydride rim at about 1.1 m above the fuel mid-plane

Fig. 3. Hydride distribution, orientation, and morphology at about 1.1 m above the fuel mid-plane for ZIRLOTM cladding from a fuel rod irradiated to 70 GWd/MTU: (a) circumferential region with thickest ($\approx 70 \mu\text{m}$) hydride rim and (b) circumferential location with thinnest ($\approx 40 \mu\text{m}$) hydride rim. δ_{ox} was $42 \mu\text{m}$, and C_{H} was 660 wppm for an adjacent cladding ring [14].

2.2 Sample Preparation and Test Conditions for Drying-Storage Simulation

High-burnup and PH segments were used to fabricate sealed and pressurized (with argon) rodlets. Bottom and top end fixtures fabricated from Zircadyne-702 were welded circumferentially to each cladding segment using an Astro Arc welder in a controlled argon environment. The cladding-segment/end-fixture remained stationary, and the weld head rotated around the joint to be welded. A zirconia pellet (25-mm-long) was inserted into the rodlet to reduce the stored energy. The heat-affected zone was relatively narrow for the weld head design and the welding parameters used. The top end fixture contained a small hole to allow for pressurization of the rodlet. RT pressures were selected to achieve target values of pressure and cladding hoop stress at 400°C. Following circumferential welding and pressurization in a specially designed sealed chamber with a sapphire lens for the laser beam, the small hole was sealed by laser welding. Leak testing was performed within the pressurization chamber. For high-burnup cladding, the corrosion and fuel-cladding bond layers were removed by mechanical means from about 15 mm from each segment end to ensure a high-quality metal-to-metal weld of the end fixtures. During this process, some of the cladding metal was also removed between these layers. Figure 4 shows a non-irradiated PH 17×17 cladding segment before (upper photo) and after (lower photo) fabrication of the pressurized rodlet.

Fig. 4. Pre-fabrication cladding segment (upper) and fabricated rodlet (lower) for drying-storage simulation. The cladding was non-irradiated, PH 17×17 ZIRLO™ cladding.

Parameters for five tests with high-burnup fuel cladding are summarized in Table 2. The tests are listed in the order in which they were conducted. Prior to rodlet pressurization, the outer diameter (D_o or OD) of each corroded segment was measured at two orientations (90° apart) and at three axial locations. These six values were averaged to give the D_o values listed in Table 2. δ_{ox} was estimated from sibling rod data (Zry-4) or from interpolation-extrapolation of

data from the same rod at different axial locations (ZIRLO™). The same approach was used to estimate metal wall thickness (h_m). The metal outer diameter (D_{mo}) was calculated from $D_o - 2\delta_{ox}$, and the metal inner diameter (D_{mi}) was calculated from $D_{mo} - 2 h_m$. The ratio R_{mi}/h_m , where R_{mi} is the metal inner radius, was used in Eq. 1 to calculate the average hoop stress (σ_θ) from the pressure difference ($\Delta P = P_i - P_o$) across the cladding wall.

$$\sigma_\theta = (R_{mi}/h_m) \Delta P \quad (1)$$

Table 2

Summary of test conditions for simulated drying-storage of high-burnup rodlets.

Material	Rodlet ID	D_o , mm	Measured RT P_i , MPa (gauge)	Calculated 400° ΔP , MPa	Target σ_θ , MPa	Hold Time at 400°C, h
17×17 ZIRLO™	648G	9.52	8.00	17.9	140	1
	648D	9.51	6.50	14.6	110	1
	648C	9.50	6.50	14.6	110	24
15×15 Zry-4	605C6	10.77	7.65	17.0	110	8
	605C2	10.78	9.70	21.6	140	3

After fabrication, the rodlet was assembled into a holder within the furnace for heating to 400°C, held at 400°C for a pre-set time, cooled at 5°C/h to 200°C, and cooled at a faster rate from 200°C to RT. Figure 5 shows the temperature history for a drying-storage simulation experiment conducted with a 1-h hold time and 1-cycle cooling. The temperature plotted is the average of two thermocouple (top and bottom locations) readings. Some tests were conducted with slow and/or fast temperature cycling (from 400°C to 300°C to 400°C) prior to cooling to 200°C at 5°C/h. In these experiments, simulated drying-storage is referred to as radial hydride treatment (RHT).

Finite element analysis (FEA) was performed to predict the thermal, elastic, and creep response of pressurized 17×17 and 15×15 cladding rodlets subjected to the reference simulated drying-storage temperature history shown in Fig. 5. For 17×17 cladding with RT (27°C) internal pressures of 11.58, 10.45, 9.10, and 8.30 MPa, the FEA-predicted internal pressures at the beginning of the 400°C hold time were close to values predicted by the ideal gas law at constant volume: -2.45%, -1.74%, -0.40%, and -0.27%, respectively. For non-irradiated cladding subjected to 1-cycle heating cooling, outer surface creep strains of 0.2% (modern Zry-4 with 120-MPa hoop stress at 400°C) to 1.4% (ZIRLO™ with 190-MPa hoop stress at 400°C) were determined from pre- and post-test laser profilometry measurements. Such low creep strains had very little effect on the average hoop stress. For high-burnup cladding, creep deformation was estimated to be negligible for stresses and hold times listed in Table 2 [7].

Fig. 5. Cladding-rodlet temperature history simulating 1-cycle heating-cooling with a 1-h hold time at 400°C and a cooling rate of 5°C/h to 200°C.

2.3 Ring Compression Test Apparatus and Conditions

RCTs were conducted in two mechanical test systems: a table-top Instron 5566 screw-type machine (limited to about 8 mm/s displacement rate) for non-irradiated samples and an Instron 8511 servohydraulic mechanical test system enclosed in a glove box for irradiated samples. Figure 6a shows a schematic of the RCT sample, loading, and displacement. The tests were run in the displacement-controlled mode. The crosshead displacement rate was pre-set at 5 mm/s and the displacement (δ) was measured as a function of time. The response load (P) was measured by a load cell located in the static support component (6 o'clock position). Figure 6b shows a cladding ring resting on the support plate of the Instron 8511 with a small space between the loading plate and the top (12 o'clock) of the ring outer diameter. A clamshell radiant-heating furnace, which was opened for sample viewing purposes, provided the heat for elevated temperature tests with high-burnup cladding. Also, the three thermocouples (TCs), which contact the ring at the 3, 6, and 9 o'clock positions for elevated-temperature tests, were moved to the viewer's right for better sample imaging. For RCTs with non-irradiated cladding, oven-heating and a single TC at the 6 o'clock position were used. For both heating systems, the TC at the 6 o'clock position was used to control the furnace or oven power, as well as to secure the sample by means of TC spring loading.

Before each high-burnup cladding test campaign, benchmark tests were conducted with as-fabricated cladding rings to ensure that the machine was operating properly and that load-displacement results were within expected values. The measured elastic loading stiffness (K_m) was compared to the calculated stiffness (K_c). K_m is expected to be $< K_c$ because of the

(a) Schematic of ring compression test

(b) Photograph of Instron 8511 with ring sample

Fig. 6. Schematic (a) and photograph (b) of the ring compression test. The top plate (above the ring) is attached to the movable loading rod. The bottom plate (below the ring) is attached to the static support rod, which contains a load cell to measure P . Tests are conducted in a displacement (δ) controlled mode.

influence of machine compliance. Following total displacement to ≤ 2 mm, the expected deviation between the offset displacement (δ_p) and the directly measured permanent displacement (d_p , difference between pre-test diameter and post-test diameter in the loading direction) was ≤ 0.2 mm, with $\delta_p > d_p$ because of the inherent error in using the traditional offset-displacement methodology for RCTs. The difference is due to the decrease in ring stiffness with increasing displacement as the ring cross-sectional shape progresses from circular to oval to oval with some flattening under the loading plate and above the support plate. As this occurs, the load progresses from a concentrated line load to a distributed load at the contact points. These geometric changes, along with residual stresses during unloading due to localized plastic deformation, lead to an unloading slope that is less than the loading slope that is usually used to determine offset displacement. In the current work, the ring is treated as a spring. The permanent displacement, which is determined after removal of the load, is considered to be the more accurate measure of the plastic deformation of the spring. “Strain” is determined from the RCT results by normalizing offset and permanent displacements to the pre-test outer diameter of the ring sample. Although hoop strains within the material increase as displacement increases, the strain of the spring does not represent strains within the material.

Figure 7 shows benchmark load-displacement results for an AF 17×17 cladding ring with 9.48-mm D_o , 0.63-mm h_m , and 7.7-mm length (L). Test conditions were the same as those used for the high-burnup cladding at RT: 5-mm/s loading displacement rate, 1.7-mm total loading displacement, and 5-mm/s unloading displacement rate. K_m was 23% lower than K_c , which is consistent with previous benchmark test results. Because L/h_m is >10 , wide-beam theory (i.e., plane-strain constraint in the axial direction) was used to determine the K_c value given in Fig. 7. Under these conditions, an axial stress (σ_z), which is about $0.36 \sigma_\theta$ at RT in the elastic

Fig. 7. Results of RCT benchmark using the Instron 8511 to compress an as-fabricated 17×17 M5[®] cladding ring at RT and 5-mm/s loading and unloading displacement rates.

deformation regime, is also induced. The traditional offset-displacement methodology calls for the use of the loading slope to “unload” the sample after the loading phase in the determination of the offset displacement. As indicated in Fig. 7, δ_p and d_p were 1.19 mm and 1.05 mm, respectively. The difference, $\delta_p - d_p = 0.14$ mm, was within the expected value based on a large Argonne database for as-fabricated cladding. Normalizing the displacements to D_o gave 12.6% offset strain and 11.1% permanent strain. The strain difference is 1.5%. Although permanent strain gives a more accurate measure of plastic strain for rings that do not fail, it cannot be determined accurately for rings that crack prior to 1.7-mm total displacement. Based on assessments of error measurements and data trends, cladding with either $\geq 1\%$ permanent strain or $\geq 2\%$ offset strain prior to the first crack extending through $>50\%$ of the wall is classified as ductile. Cladding that cracks through $>50\%$ of the wall prior to achieving 2% offset strain is classified as brittle in the current work.

3. Results for Non-Irradiated PH Cladding after Simulated Drying and Storage

Although it was found that PH cladding with a uniform distribution of hydrides across the wall was not a good surrogate for high-burnup cladding, the experience gained from conducting tests with PH cladding proved to be useful in determining a metric to define the extent of radial hydride formation, in visualizing the discontinuity of cracks along the axial direction (indicative of discontinuity of radial-hydride platelets), and in determining the adverse effects of low hydrogen content on radial-hydride precipitation and radial-hydride-induced embrittlement.

Figure 8 shows the uniform distribution of circumferential hydrides in ZIRLO™ charged with low (220 wppm in Fig. 8a) and high (530 wppm in Fig. 8b) C_H values. The uniform distribution of hydrides across the cladding wall is significantly different from the radial distribution of hydrides in high-burnup cladding (see Figs. 1-3). At 400°C, most of the hydrogen (≈ 200 wppm) in Fig. 8a would dissolve while $<40\%$ of the hydrides in Fig. 8b would dissolve. If radial hydride platelets precipitated during cooling for high C_H samples, they would tend to be short and to be located between densely packed circumferential hydride platelets.

3.1 Radial hydride continuity factor

Figure 9a shows the hydride morphology following RHT at 190 MPa and one of three long cracks that formed during 1.6-mm displacement for a ring containing 630 ± 110 wppm C_H . The first significant load drop (corresponding to $>50\%$ wall cracking) occurred at 1% offset strain, which is indicative of brittle behavior. The crack imaged in Fig. 9a initiated at the cladding outer surface, propagated along radial hydrides at three locations, and propagated along circumferential hydrides at two intermediate locations. As the matrix metal is very ductile at 150°C, it is assumed that the crack path is also the path of radial-circumferential hydrides. Circumferential hydrides connecting the radial hydrides were within a 150- μ m band. The RHCF is assessed to be 100% within this 150- μ m band. Figure 9b shows a section of a ring with 180 ± 20 wppm C_H subjected to RHT at 190 MPa. This ring was displaced only 0.6 mm to better image crack patterns prior to post-crack deformation. Very tight cracks, which initiated at the cladding inner surface, were observed at 12 and 6 (Fig. 9b) o'clock after $<1\%$ offset strain. The 12 o'clock crack extended through about 90% of the wall, and the 6 o'clock crack extended through 100% of the wall. The RHCF is clearly 100% for this image.

(a) 220 wppm C_H

(b) 530 wppm C_H

Fig. 8. Uniform distribution of hydrides in AF ZIRLO™ cladding charged with: (a) 220 wppm C_H and (b) 530 wppm C_H .

(a) PH and RHT ZIRLO™ with 630 ± 110 wppm C_H

(b) PH and RHT ZIRLO™ with 180 ± 20 wppm C_H

Fig. 9. Through-wall crack paths for PH and RHT ZIRLO™ with (a) 630 ± 110 wppm C_H and (b) 180 ± 20 wppm C_H . RHCF is 100% for both images.

3.2 Discontinuity of radial hydride platelets in the axial direction

Macroscopic images of rings following RCT displayed jagged and discontinuous cracks along the axial direction. Figure 10 shows the crack morphology along the axial direction of two rings (H and J) from the same rodlet as ring D, from which the metallographic image shown in Fig. 9a was taken. C_H (620 ± 110 wppm) for rings H and J was essentially the same as for ring D. The cracks at the top of the rings (12 o'clock position) initiated at the cladding inner surface. Based on Fig. 10, it is clear that the cracks are jagged and somewhat discontinuous in the axial direction. The discontinuity along the outer surface (not imaged) of these two rings is more pronounced than along the inner surface. The results suggest discontinuity of radial hydride platelets along the axial direction. The lack of axial continuity can also be seen in metallographic images from cross sections that were about 0.5-mm apart along the axis of the ring.

Fig. 10. Post-RCT appearance of two cladding rings (H and J) with about the same hydrogen content and RHCF as shown in Fig. 9a for ring D of PH ZIRLO™ rodlet 634B2D4. The top (12 o'clock) cracks initiated at the cladding inner surface and propagated through the wall and along the axis in jagged pathways.

3.3 Effects of hydrogen concentration of radial-hydride formation and embrittlement

The results of scoping RCTs for PH and RHT non-irradiated cladding samples were initially evaluated on a pass-fail basis in the sense that the success criterion was survival of 1.7-mm RCT displacement with cracks that penetrated into $\leq 50\%$ of the wall thickness. Although this criterion is severe, the results were useful in determining the effects of hydrogen content on radial-hydride-induced cracking. Figure 11 shows the results of the pass-fail RCTs conducted at 150°C. The open symbols indicate cladding rings that survived 1.7-mm displacement (at 5 mm/s) with either no cracking or with $\leq 50\%$ wall cracks (one ring). The results are particularly interesting because they show the “ductile-to-brittle” transition C_H values at fixed 400°C RHT hoop stresses. “Pass-fail” embrittlement was observed as C_H dropped below a critical level: 275 ± 25 wppm for 150-MPa hoop stress at 400°C, 200 ± 20 wppm for 135-MPa hoop stress at 400°C, and <100 wppm for 120-MPa hoop stress at 400°C. These results appear to be counter-intuitive in that embrittlement occurred at lower C_H values. However, the results are consistent with metallographic examinations, which show that longer radial hydrides precipitated at lower C_H levels.

The results of RCTs conducted at RT were quite different in that all but 2 of the rings developed multiple cracks extending through $>50\%$ of the wall thickness. These results are shown in Fig. 12. The ring subjected to the lowest RHT stress (120 MPa) and with relatively low C_H (114 ± 10 wppm) survived the RT displacement of 1.7 mm with no cracking.

Fig. 11. RCT data at 150°C for PH Zry-4 and ZIRLO™ following RHT. Open symbols represent rings that survived 1.7-mm displacement with $\leq 50\%$ wall cracks. Solid symbols represent samples with cracks $>50\%$ of the wall thickness.

Fig. 12. RCT data at RT for PH Zry-4 and ZIRLO™ following RHT. Open symbols represent rings that survived 1.7-mm displacement with $\leq 50\%$ wall cracks. Solid symbols represent samples with cracks $> 50\%$ of the wall thickness.

Figure 13 is useful for visualizing the effects of C_H on extent of radial hydride precipitation during RHT. The results are for a PH ZIRLO™ rodlet with an axial variation in C_H , which was subjected to the temperature history shown in Fig. 5 at 135-MPa peak hoop stress. Figure 13a shows the hydride orientation and morphology for a ring with 190-wppm C_H ; long radial hydrides precipitated and formed brittle pathways across the cladding wall. For this C_H level, all the hydrogen would be in solution at 400°C and available for precipitation as radial and/or circumferential hydride platelets. The second ring had 250-wppm C_H . The hydride morphology for this case is shown in Fig. 13b. Short radial hydride platelets are observed between circumferential hydrides. For this C_H level, ≈ 50 wppm hydrogen would be present as circumferential hydride platelets at 400°C. The presence of these circumferential hydride platelets at the initiation of cooling appears to have caused an increased precipitation of circumferential hydride platelets (relative to what is shown in Fig. 13a). Also, growth of the circumferential hydrides present at 400°C may have initiated at a higher temperature during cooling than the temperature for precipitation of new hydride platelets. The presence of circumferential hydride platelets at the temperature ($\approx 350^\circ\text{C}$) for precipitation of new hydrides may have limited the lengths of radial hydride platelets and the corresponding RHCF.

(a) Area of ring with 190 wppm C_H

(b) Area of ring with 250 wppm C_H

Fig. 13. Hydrogen content and hydride morphology for a non-irradiated PH ZIRLO™ rodlet subjected to RHT at 135-MPa peak hoop stress: (a) area of ring with 190 wppm C_H and (b) area of ring with 250 wppm C_H .

4. Results for High-Burnup Cladding following Radial Hydride Treatment

Table 3 gives the calculated pressure difference (ΔP) across the cladding wall and the hoop stress at 400°C. The hoop stress was determined from Eq. 1 using measured values for h_m and R_{mi} . Metallographic examinations were performed to determine these parameters. Detailed results from the examinations are given in subsequent sections. However, for convenience, it is useful to present a summary of characterization results, so that actual 400°C hoop stresses can be identified. Within the uncertainty of the calculation, the actual and target hoop stresses are 110 and 140 MPa. The hoop stress chosen for rodlet 648G was based on data for PH (350-650 wppm) and RHT (150 MPa at 400°C) ZIRLO™ rodlets, which exhibited high ductility at 150°C.

Table 3

Summary of measured dimensions, ΔP across cladding wall at 400°C, and σ_θ at 400°C prior to cooling at 5°C/h under conditions of decreasing pressure and stress.

Parameter	High-Burnup ZIRLO™			High-Burnup Zry-4	
	648G	648D	648C	605C6	605C2
D_o , mm	9.52±0.02	9.51±0.01	9.50±0.00	10.77±0.01	10.78±0.01
δ_{ox} , μm	52±6	33±3	26±2	82±2	102±4
D_{mo} , mm	9.42±0.03	9.44±0.02	9.45±0.00	10.61±0.01	10.58±0.02
h_m , mm	0.54±0.00	0.56±0.00	0.56±0.00	0.71±0.01	0.70±0.01
D_{mi} , mm	8.34±0.03	8.33±0.02	8.34±0.01	9.19±0.04	9.18±0.04
R_{mi}/h_m	7.74±0.06	7.52±0.03	7.51±0.06	6.50±0.14	6.58±0.13
ΔP , MPa	17.9	14.6	14.6	17.00	21.60
σ_θ , MPa	139±1	110±0	110±1	110±2	142±3

Equation 1 gives the wall-average hoop stress. Elastic stress values at the ID and OD are about 10 MPa higher and lower, respectively. The ID and OD stresses approach the average value during the hold time if a small amount of thermal creep (0.2 to 1%) occurs. These stresses decrease linearly with pressure during the cooling period.

4.1 Post-RHT Characterization Results for High-Burnup Cladding

4.1.1 High-Burnup ZIRLO™ Rodlet 648G

Rodlet 648G was subjected to a cladding hoop stress of 140 MPa at 400°C, held for one hour at 400°C, cooled slowly at 5°C/h to 200°C, and cooled more rapidly from 200°C to RT. Following this RHT, samples were sectioned for C_H determination (6 rings), metallographic examination (2 rings) and ring compression testing (3 rings). Figure 14 shows the post-RHT sectioning diagram.

Fig. 14. Sectioning diagram for high-burnup ZIRLO™ rodlet 648G following RHT at 400°C peak temperature and 140-MPa peak hoop stress.

Table 4 summarizes sample masses and C_H data for rings 2, 4, 6, 9, and 11, as well post-RCT ring 10. At each of these locations, cladding rings were sectioned into four quarter-arc samples to allow for sample insertion into the LECO hydrogen-analyzer drop chute. As can be seen from Fig. 14 and Table 4, the circumferential variation in C_H was much higher than the axial variation. The mass-averaged C_H was 650 ± 190 wppm. This value is consistent with the 620 ± 140 wppm C_H value measured for segment 648F (see Fig. 3), whose mid-span axial location was 130 mm below segment 648G.

Table 4

C_H data for high-burnup ZIRLO™ rodlet 648G following RHT with a one-hour hold time at 400°C and a cooling rate of 5°C/h.

Ring	Mass, g	C_H , wppm	# of Data Points
2	0.178	652 ± 163	4
4	0.198	689 ± 262	4
6	0.189	720 ± 310	4
9	0.171	639 ± 79	4
10	0.805	639 ± 231	4
11	0.168	625 ± 115	4
Total	1.71	---	24
Average	---	650 ± 190	---

Metallographic examination was performed at eight circumferential locations for rings 7 and 8 at the indicated axial locations (see circles in Fig. 14). Average values were 52 ± 6 μm for δ_{ox} and 539 ± 4 μm for h_m . Magnifications used to determine these parameters were 200X and 100X, respectively. Eight 100X images include about 25% of the cross-sectional area. Results for each ring are summarized in Table 5.

Table 5

δ_{ox} and h_m data for high-burnup ZIRLO™ rodlet 648G following RHT with a one-hour hold time at 400°C and a cooling rate of 5°C/h.

Ring	δ_{ox} , μm	h_m , μm	# of Data Points
7	52±7	539±5	8
8	52±4	539±4	8
Total	---	---	16
Average	52±6	539±4	---

Detailed metallographic examination was performed for polished-and-etched cladding surfaces to determine hydride morphology, orientation, and RHCF. Figures 15 and 16 show the hydride morphology and orientation for high-burnup ZIRLO™ rodlet 648G rings 7 and 8, respectively, at circumferential locations of maximum and minimum values of RHCF: 80% and 38% in Fig. 15, and 80% and 36% in Fig. 16. The set of eight images for rings 7 and 8 are shown in Appendix A. The lines L_R and h_m used to determine the RHCF are also shown in Fig. 15. For ring 7 and 8 locations, the RHCF values were determined to be 70±17% and 61±16%, respectively. Averaging the data at the 16 circumferential locations gave 65±17% RHCF for rodlet 648G. These values take into account the effects of etching on hydride thickness, length, and continuity. Cross sections that are “over-etched” result in images that exaggerate the thickness of hydrides, while “under-etched” cross sections give the appearance of thinner hydrides with more regions of apparent discontinuity. To account for the effects of etching, radial-circumferential hydrides with ≤5- μm -gap regions are treated as continuous in determining the RHCF. These gaps are more apparent for the images taken at magnifications >100X. Another way of interpreting the use of the 5- μm gap is to assume that a crack growing in the radial direction can traverse such a gap consisting of low-ductility matrix metal.

4.1.2 High-Burnup ZIRLO™ Rodlet 648D

Rodlet 648D was subjected to a cladding hoop stress of 110 MPa at 400°C, held for one hour at 400°C, cooled slowly at 5°C/h to 200°C, and cooled more rapidly from 200°C to RT. Following this RHT, samples were sectioned for C_H determination (5 rings), metallographic examination (1 ring) and ring compression testing (4 rings). Figure 17 shows the post-RHT sectioning diagram for this rodlet.

Table 6 summarizes sample masses and C_H data for rings 2, 4, 6, 9, and 11. The mass-averaged C_H was 425±63 wppm. This value is consistent with the 310±30 wppm C_H value measured for segment 648A (see Fig. 2) and the 370±30 wppm measured for 648B [17], whose mid-span axial location was 160 mm below that of segment 648D.

Metallographic examination was performed at eight circumferential locations for ring 7 at the indicated axial location (see circle in Fig. 17). δ_{ox} and h_m were 33±3 μm and 555±2 μm (about 0.56 mm), respectively.

(a) Maximum RHCF (80%)

(b) Minimum RHCF (38%)

Fig. 15. Metallographic images from high-burnup ZIRLO™ rodlet 648G ring 7 following RHT at 400°C peak temperature and 140-MPa peak hoop stress prior to cooling at 5°C/h: (a) region with maximum RHCF (80%) and (b) region with minimum RHCF (38%). C_H was 720 ± 310 wppm for adjacent ring 6.

(a) Maximum RHCF (80%)

(b) Minimum RHCF (36%)

Fig. 16. Hydride morphology for rodlet 648G ring 8 showing regions of: (a) maximum RHCF (80%) and (b) minimum RHCF (36%). Images were taken following 1-cycle-cooling RHT at 5°C/h with 140-MPa hoop stress at 400°C. C_H was 630 ± 90 wppm for adjacent ring 9.

Fig. 17. Sectioning diagram for high-burnup ZIRLO™ rodlet 648D following RHT at 400°C peak temperature and 110-MPa peak hoop stress.

Table 6

C_H data for high-burnup ZIRLO™ rodlet 648D following RHT with a one-hour hold time at 400°C and a cooling rate of 5°C/h.

Ring	Mass, g	C _H , wppm	# of Data Points
2	0.171	455±67	4
4	0.102	425±108	4
6	0.219	426±77	4
9	0.144	396±18	4
11	0.119	412±42	4
Total	0.76	---	20
Average	---	425±63	---

Detailed metallographic examination was performed for polished-and-etched cladding surfaces to determine hydride morphology, orientation, and the RHCF. Figure 18 shows the hydride morphology and orientation for high-burnup ZIRLO™ rodlet 648D ring 7 at circumferential locations of maximum and minimum values of RHCF: 48% in Fig. 18a and 18% in Fig. 18b. The set of eight images for rodlet 648D ring 7 is shown in Appendix B. Based on data at eight circumferential locations, the average RHCF was 27±10% for rodlet 648D.

(a) Maximum RHCF (48%)

(b) Minimum RHCF (18%)

Fig. 18. Hydride morphology for 648D7 showing regions of: (a) maximum RHCF (48%) and (b) minimum RHCF (18%). Images were taken following 1-cycle-cooling RHT at 5°C/h with 110-MPa hoop stress at 400°C. C_H was 425 ± 77 wppm for adjacent ring 6.

4.1.3 High-Burnup ZIRLO™ Rodlet 648C

Prior to conducting RHT for rodlets 648G and 648D, the expectation was that about 200 wppm of hydrogen would go into solution beneath the dense hydride rim during the hold time at 400°C. However, for some of the images in Appendices A and B, it appears that much less than 200 wppm of hydrogen precipitated in these regions. Based on calculations performed for diffusion across the cladding wall, it would take less than 30 minutes for the cladding below the hydride rim to reach 93% of the equilibrium concentration at 400°C even for extreme initial conditions, which assume that all the hydrogen is in the hydride rim and the cladding wall thickness is 0.57 mm. The characteristic diffusion time (τ), which corresponds to the time it takes to reach 93% of steady state, is defined as $\tau = (h_m)^2/D$, where D is the hydrogen diffusivity. The calculation was performed for the lesser of the D values at 400°C given by Kammenzind et al. [$2.09 \times 10^{-6} \text{ cm}^2/\text{s}$ in Ref. 3] and Kearns [$4.12 \times 10^{-6} \text{ cm}^2/\text{s}$ in Ref. 18]. Using the lower value of D gives $\tau = 1555 \text{ s}$.

To allow for additional time for hydrogen diffusion, rodlet 648C was held for 24 hours at 400°C. Also, in order to maintain orientation, a horizontal line was drawn on the rodlet. With the exception of the longer hold time, rodlet 648C was subjected to the same RHT as rodlet 648C, including the 110-MPa hoop stress at 400°C. Following RHT, samples were sectioned for C_H determination (5 rings), metallographic examination (1 ring) and ring compression testing (4 rings). Figure 19 shows the post-RHT sectioning diagram for this rodlet.

Fig. 19. Sectioning diagram for high-burnup ZIRLO™ rodlet 648C following RHT at 400°C peak temperature and 110-MPa peak hoop stress. Hold time at 400°C was 24 hours.

Table 7 summarizes sample masses and C_H data for rings 2, 4, 6, 9, and 11. The mass-averaged C_H was $350 \pm 80 \text{ wppm}$. This value is consistent with the $370 \pm 30 \text{ wppm}$ measured for 648B [17], whose mid-span axial location was 80 mm below that of segment 648C. Even with the 24-hour hold time at 400°C, the hydrogen loss to the end fixtures during RHT appears to be insignificant for the middle 45 mm of rodlet 648C.

Table 7

C_H data for high-burnup ZIRLO™ rodlet 648C following RHT with a 24-hour hold time at 400°C and a cooling rate of 5°C/h.

Ring	Mass, g	C_H , wppm	# of Data Points
2	0.177	329±66	4
4	0.172	346±61	4
6	0.207	325±72	4
9	0.180	367±103	4
11	0.149	384±113	4
Total	0.89	---	20
Average	---	350±80	---

Metallographic examination was performed at eight circumferential locations for ring 7 at the indicated axial location (see circle in Fig. 19). δ_{ox} and h_m were $26 \pm 2 \mu m$ and $555 \pm 4 \mu m$ (about 0.56 mm), respectively.

Detailed metallographic examination was performed for polished-and-etched cladding surfaces to determine hydride morphology, orientation, and RHCF. Figure 20 shows the hydride morphology and orientation for high-burnup ZIRLO™ rodlet 648C ring 7 at circumferential locations of maximum and minimum RHCF values: 54% in Fig. 20a and 26% in Fig. 20b. Based on the data at eight circumferential locations, the average RHCF was $35 \pm 12\%$ for rodlet 648C.

Prior to cooling from 400°C, the cladding metal below the hydride rim should have had about 200 wppm hydrogen in solution uniformly distributed around the cross section. Based on the results shown in Appendix C, it is clear that some hydrogen diffused in the circumferential direction prior to precipitation. This can be seen from the images in Fig. 21 from regions adjacent to each other. Figure 21a shows a region with essentially no visible hydrides within the inner third of the cladding wall. This region contained only one short radial hydride and a couple of short circumferential hydrides. Although the RHCF was 31% for this region, the indicated radial hydride is located in a relatively benign location where hoop bending stresses due to RCT loading would be low. The visible hydrides in this inner-third of the cladding wall correspond to an average hydrogen content considerably less than 100 wppm. The adjacent region shown in Fig. 21b contained a relatively high concentration of hydrides, especially radial hydrides, within the inner half of the cladding wall. The RHCF for this region was 48% (based on Fig. 21b), and the location of the radial hydrides with respect to the inner cladding wall is such that they would be exposed to the maximum hoop bending stresses under the RCT loading. It appears that hydrogen diffused distances on the order of 1 mm in the circumferential direction prior to precipitation as hydrides during slow cooling. It also appears that sites with radial hydrides attracted dissolved hydrogen during the precipitation process.

Figure 22 shows a 200X image of a sub-region of Fig. 21b. The total hydride length in Fig. 22 is $\approx 2100 \mu m$. The total cross-sectional area of metal and hydrides is $10.7 \times 10^4 \mu m^2$. The apparent width of radial hydrides is in the range of 2 to 10 μm . However, it is well known that optical-microscopy images exaggerate the width of hydrides. The apparent width also increases

with the degree of etching (i.e., HF content and exposure time) prior to imaging. Using Kearns's value for average hydride width ($2.5\text{ }\mu\text{m}$ in Ref. 19), the area of hydrides is $5300\text{ }\mu\text{m}^2$ or 5% of the total area in Fig. 22. According to Kearns [19], the mass fraction of H to Zr in the hydrides is 1.51×10^4 wppm. Thus, the hydrogen concentration in Fig. 22 is ≈ 740 wppm. The primary uncertainties in this calculation are the thickness of individual hydrides and the continuity of hydrides in the thickness direction. Based on other images, thin hydride platelets precipitated close to each other and appeared to form a continuous hydride. Regardless of these uncertainties, it seems clear that the C_H within the inner third of the cladding is much less than 200 wppm at one circumferential location (Fig. 21a) and much greater than 200 wppm at an adjacent circumferential location (Fig. 21b). This observation supports the hypothesis that hydrogen diffused circumferentially from the region shown in Fig. 21a to the region shown in Fig. 21b.

The eight images shown in Appendix C cover only 24% of the cladding cross section. Given the circumferential variation in RHCF, a second set of images was taken at 33 orientations in order to cover the whole cross section of ring 7. The corresponding RHCF was $33 \pm 14\%$. This RHCF derived from 33 circumferential locations is remarkably close to the RHCF derived from images at only 8 circumferential locations. Averaging the original 8 values with these 33 RHCF values gives an RHCF of $33 \pm 13\%$. The effective lengths of 54 radial hydrides were measured within the 33 images. Figure 23 shows the results of this detailed analysis. The reference point at 0 mm corresponds to the 12 o'clock position. Local values for the effective radial-hydride length varied from $45\text{ }\mu\text{m}$ to $325\text{ }\mu\text{m}$; the average value was $158 \pm 79\text{ }\mu\text{m}$, which corresponds to $28 \pm 14\%$ RHCF. This value for 54 data points is understandably less than the $33 \pm 13\%$ RHCF for 33 data points based on maximum RHCF per image. The average spacing between radial hydrides was $0.45 \pm 0.44\text{ mm}$.

(a) Maximum RHCF (54%)

(b) Minimum RHCF (26%)

Fig. 20. Hydride morphology for 648C7 showing regions of: (a) maximum RHCF (54%) and (b) minimum RHCF (26%). Images were taken following 1-cycle-cooling RHT at 5°C/h with 110-MPa hoop stress at 400°C. C_H was 325 ± 72 wppm for adjacent ring 6.

(a) Low hydride concentration within inner third of cladding wall

(b) High hydride concentration within inner half of cladding wall

Fig. 21. Metallographic images of adjacent regions of the cross section for ring 7 of high-burnup ZIRLO™ rodlet 648C: (a) low hydride concentration within inner third of cladding wall and (b) high hydride concentration within inner half of cladding wall.

Fig. 22. Enlarged image of a region from Fig. 21b showing a relatively high density of radial and circumferential hydrides within the inner half of the cladding wall for ring 7 from high-burnup ZIRLO™ rodlet 648C.

Fig. 23. Effective radial hydride length as a function of circumferential location along the cladding inner surface of high-burnup ZIRLO™ rodlet 648C ring 7. h_m was 555 μm.

4.1.4 High-Burnup Zry-4 Rodlet 605C6

Rodlet 605C6 was subjected to a cladding hoop stress of 110 MPa at 400°C, held for eight hours at 400°C, cooled slowly at 5°C/h to 200°C, and cooled more rapidly from 200°C to RT. Following this RHT, samples were sectioned for C_H determination (4 rings), metallographic examination (1 ring) and ring compression testing (4 rings). Figure 24 shows the post-RHT sectioning diagram. The black line represents the 12 o'clock position for metallographic examination and ring compression testing. The red line gives the orientation that allows the distinction to be made between the 3 o'clock and 9 o'clock orientations.

Fig. 24. Sectioning diagram for high-burnup Zry-4 rodlet 605C6 following RHT at 400°C peak temperature and 110-MPa peak hoop stress.

Table 8 summarizes sample masses and C_H data for rings B, D, F, and I. At each of these locations, cladding rings were sectioned into four quarter-arc samples to allow for insertion of the sample into the LECO hydrogen-analyzer drop chute. As can be seen from Fig. 24 and Table 8, the circumferential variation in C_H was higher than the axial variation. The mass-averaged C_H was 520 ± 80 wppm. This value is consistent with the 545 ± 80 wppm C_H value measured for sibling rod F07 at 320 mm above the fuel mid-plane.

Several metallographic examinations were performed for ring G at the indicated axial locations (see circle in Fig. 24): as-polished condition to determine δ_{ox} and h_m (7 images); polished-and-etched condition to determine hydride morphology (6 images); re-polished/re-etched condition to better image hydrides (9 images); and re-ground (0.5 mm)/re-polished/re-etched to determine variation of radial hydrides over a short axial distance (6 images). δ_{ox} and h_m were 82 ± 2 μm and 710 ± 15 μm , respectively.

Table 8

C_H data for high-burnup Zry-4 rodlet 605C6 following RHT with an eight-hour hold time at 400°C and a cooling rate of 5°C/h.

Ring	Mass, g	C_H , wppm	# of Data Points
B	0.181	533±106	4
D	0.146	551±96	4
F	0.261	512±86	4
I	0.296	498±92	4
Total	1.71	---	16
Average	---	520±90	---

The metallographic images for characterizing radial hydrides are shown in Appendix D. These eleven images are from the re-polished/re-etched surface of ring G. Figure 25 shows the circumferential orientations with the maximum (Fig. 25a) and minimum (Fig. 25b) values of RHCF: 21% and 3%, respectively. These images are from the 12 o'clock and 11 o'clock orientations, respectively. The radial hydrides at the 12 o'clock position are relatively short hydrides connected by circumferential hydrides. As indicated by the arrow in Fig. 25b, the location of these radial hydrides is such that they might contribute to crack propagation from the outer surface inward if they were located at the 3 or 9 o'clock orientations in the RCT. However, they would not contribute to crack initiation, especially at the 12 and 6 o'clock orientations where the maximum tensile stress is located at the inner surface.

Unlike the radial hydrides observed for high-burnup ZIRLO™ cladding subjected to the same RHT conditions, the radial hydrides for high-burnup Zry-4 are short and more discontinuous. Figures 26a and 26b show 100X and 500X images, respectively, for a radial hydride at the 2:30 o'clock orientation. The hydride indicated by the arrow in Fig. 26a appears to be continuous over a projected length (onto the radius) of more than 130 μm . Based on Fig. 26b, there are regions of discontinuity along the radial-circumferential hydrides. Allowing for up to 5- μm discontinuity, the projected radial hydride length is only 100 μm .

The RHCF for high-burnup Zry-4 rodlet 605C6 was determined to be 9±5% based on the images shown in Appendix D, as well as other images at magnifications of 200X and 500X.

(a) Maximum RHCF (21%)

(b) Minimum RHCF (3%)

Fig. 25. Hydride morphology for high-burnup Zry-4 rodlet 605C6 ring G showing regions of: (a) maximum RHCF (21%) and (b) minimum RHCF (3%). Images were taken following 1-cycle-cooling RHT at 5°C/h with 110-MPa hoop stress at 400°C. C_H was 512 ± 86 wppm for adjacent ring F.

(a) 100X image

(b) 500X image

Fig. 26. Hydride morphology for high-burnup Zry-4 rodlet 605C6 ring G at the 2:30 o'clock position (a) 100X image and (b) 500X image. Images were taken following 1-cycle-cooling RHT at 5°C/h with 110-MPa hoop stress at 400°C. RHCF = 13%.

4.1.5 High-Burnup Zry-4 Rodlet 605C2

Rodlet 605C2 was subjected to a cladding hoop stress of 140 MPa at 400°C, held for three hours at 400°C, cooled slowly at 5°C/h to 200°C, and cooled more rapidly from 200°C to RT. Following this RHT, samples were sectioned for C_H determination (3 rings), metallographic examination (1 ring) and ring compression testing (3 rings). Figure 27 shows the post-RHT sectioning diagram. The black line determines the 12 o'clock position for metallographic examination and ring compression testing. The red line gives the orientation that allows the distinction to be made between the 3 and 9 o'clock orientations.

Fig. 27. Sectioning diagram for high-burnup Zry-4 rodlet 605C2 following RHT at 400°C peak temperature and 140-MPa peak hoop stress.

Table 9 summarizes sample masses and C_H for rings B, D, and H. At each of these locations, cladding rings were sectioned into four quarter-arc samples to allow for sample insertion into the LECO hydrogen-analyzer drop chute. As can be seen from Fig. 27 and Table 9, the circumferential variation in C_H is higher than the axial variation. The mass-averaged C_H was 615±82 wppm. This value is lower than the 740±110 wppm C_H measured for sibling rod F07 at 650 mm above the fuel mid-plane. However, the difference may be due to rod-to-rod variation at comparable axial locations rather than hydrogen loss to the end fixtures. Also, C_H for rod F07 was measured for only one cladding ring with a mass of about 0.2 g. This would not capture axial variations in hydrogen content.

Metallographic examinations were performed at 12 orientations for ring E at the indicated axial location (see circle in Fig. 27). δ_{ox} and h_m were 102±4 µm and 698±12 µm, respectively.

Table 9

C_H data for high-burnup Zry-4 rodlet 605C2 following RHT with a three-hour hold time at 400°C and a cooling rate of 5°C/h.

Ring	Mass, g	C_H , wppm	# of Data Points
B	0.298	659±56	4
D	0.325	586±78	4
H	0.195	595±10	4
Total	0.82	---	12
Average	---	615±82	---

Metallographic images for characterizing radial hydrides are shown in Appendix E. These twelve images are from a ground, polished, and etched surface of ring E. Figure 28 shows the circumferential orientations with the maximum (Fig. 28a) and minimum (Fig. 28b) values of RHCF: 24% and 11%, respectively. These images are for the 2 and 12 o'clock orientations. Figure 29 shows higher-magnification images of localized regions at these two orientations. At the 2 o'clock orientation, a higher number density of longer radial hydrides is visible within the inner third of the cladding wall. These hydrides could participate in crack initiation and propagation. However, the longest radial-circumferential hydride path across the wall is located near the cladding mid-wall (see arrows in Figs. 28a and 29a). This relatively long hydride would contribute only to crack propagation. The radial hydrides at the 12 o'clock position are relatively short and connected by circumferential hydrides. As indicated by the arrows in Figs. 28b and 29b, the location of these radial hydrides is such that they might contribute to crack propagation from the inner surface outward if they were located at the 12 or 6 o'clock orientations in the RCT. However, they would not contribute to crack initiation at these orientations because they do not extend to the inner cladding surface.

In comparing high-burnup Zry-4 rodlets 605C6 and 605C2, the increase in RHT hoop stress from 110 MPa to 140 MPa did result in an increase in number density and length of radial hydrides close to the cladding inner surface. These are potentially more degrading than hydrides located near the cladding mid-wall. The RHCF for high-burnup Zry-4 rodlet 605C2 was determined to be 16±4% based on the images shown in Appendix E, as well as other images at 200X and 500X magnifications.

4.1.6 Summary of Characterization Results for High-Burnup Cladding after RHT

Table 10 summarizes the RHT and post-RHT characterization results from micrometer, C_H , and metallographic-examination measurements.

(a) Maximum RHCF (24%)

(b) Minimum RHCF (11%)

Fig. 28. Hydride morphology for high-burnup Zry-4 rodlet 605C2 ring E showing regions of: (a) maximum RHCF (24%) and (b) minimum RHCF (11%). Images were taken following 1-cycle-cooling RHT at 5°C/h with 140-MPa hoop stress at 400°C for 3-hours. C_H was 586 ± 78 wppm for adjacent ring D.

(a) Maximum RHCF (24%)

(b) Minimum RHCF (11%)

Fig. 29. Higher magnification images for the two orientations shown in Fig. 28 for high-burnup Zry-4 rodlet 605C2 ring E: (a) maximum RHCF (24%) and (b) minimum RHCF (11%).

Table 10

RHT and post-RHT characterization results for 70-GWd/MTU ZIRLO™ and for 67-GWd/MTU Zry-4.

Material	Rodlet ID	400°C Hold Time, h	σ_θ at 400°C, MPa	Metal D_{mo} , mm	Metal h_m , mm	δ_{ox} , μm	C_H , wppm	RHCF, %
ZIRLO™	648G	1	140	9.42	0.54	52±7	650±190	65±17
	648D	1	110	9.44	0.56	33±3	425±63	27±10
	648C	24	110	9.45	0.56	26±2	350±80	33±13
Zry-4	605C6	8	110	10.61	0.71	82±2	520±90	9±5
	605C2	3	140	10.58	0.70	102±4	615±82	16±4

4.2 Post-RHT RCT Results for High-Burnup Cladding

The servohydraulic Instron 8511 was used to perform RCTs with high-burnup cladding samples. Although it is capable of achieving displacement rates $\gg 5$ mm/s, the reference displacement rate of 5 mm/s was used to allow direct comparison with results for PH samples (see Section 3). The pre-set 2-mm maximum displacement was maintained, along with the same lag time before the sample was unloaded by raising the load train. However, the Instron 8511 load train has a very short displacement lag such that the recorded sample displacement was approximately 2 mm for the pre-set 2-mm displacement. During the course of testing, two changes were made to the test protocol: (a) the pre-set crosshead displacement was reduced to 1.7 mm and (b) the unloading displacement rate was programmed to be 5 mm/s to eliminate the time lag for sample unloading. Table 11 shows the test conditions for the 18 RCTs conducted with high-burnup cladding.

As indicated in Fig. 7, the ring unloading slope (K_U) following plastic deformation is less than the loading slope (K_L). The use of K_L to mathematically unload the sample prior to significant wall cracking leads to an inherent error in δ_p that increases with plastic displacement. For rings that do not crack during the RCT, d_p determined from pre- and post-test diameter measurements may be used instead of the offset displacement. Also, a correlation may be developed from uncracked rings to estimate the unloading slope. This is important as most rings sectioned from RHT rodlets will crack prior to unloading. The post-test diameter measurement and the measured unloading slope are not meaningful for such rings. High-burnup ZIRLO™ ring 648C8, which exhibited no indication of cracks, had a traditional offset strain (δ_p/D_{mo}) of 12.2% and a permanent strain (d_p/D_{mo}) of 9.4%. The approximate unloading slope (0.64 kN/mm) was about 25% lower than the loading slope (0.85 kN/mm). Equation 2 was developed from these data to approximate the unloading slope for the other 10 high-burnup ZIRLO™ rings. It incorporates the observations from numerous benchmark tests that the change in unloading slope is negligible for $\leq 2\%$ traditional offset strain.

Table 11

Test conditions for high-burnup RCTs. $\delta_{\max}$ is the pre-set maximum displacement.

Material	ID	RHT σ_{θ} at 400°C, MPa	RCT T, °C	$\delta_{\max}$, mm	5 mm/s Unloading Rate
ZIRLO™	648G	140			
	5		150	2.0	No
	10		150	2.0	No
	3		195	1.7	Yes
ZIRLO™	648D	110			
	3		30	1.7	Yes
	5		150	2.0	No
	8		150	2.0	No
ZIRLO™	648C	110			
	5		30	1.7	Yes
	3		90	1.7	Yes
	10		120	1.7	Yes
Zry-4	605C6	110			
	E		24	1.7	Yes
	C		90	1.7	Yes
	J		120	1.7	Yes
Zry-4	605C2	140			
	G		30	1.7	Yes
	F		90	1.7	Yes
	C		150	1.7	Yes

$$K_U = [1 - 0.025 (\delta_p/D_{mo} - 2\%)] K_L \quad (2)$$

Using K_U to unload ring 648C8, the corrected offset strain is 10.2%, which is closer to the permanent strain (9.4%) than the traditional offset strain (12.2%) determined from K_L .

A similar approach was used to determine the relationship between unloading and loading slopes for high-burnup Zry-4 ring 605C6H, which exhibited no signs of cracking following the RCT. The loading and unloading slopes were 1.51 kN/mm and 1.10 kN/mm, respectively. The traditional offset and permanent strains were 11.1% and 8.4%, respectively. These data were used to derive the decrease in K_U as a function of traditional offset strain:

$$K_U = [1 - 0.030 (\delta_p/D_{mo} - 2\%)] K_L \quad (3)$$

Use of Eq. 3 gave a corrected offset strain of 9.2%, which is closer to the measured permanent strain of 8.4% than the 11.1% traditional offset strain. Equation 3 was used to correct the offset strain for the other six high-burnup Zry-4 RCT rings.

The embrittlement criterion evolved during the course of testing PH and high-burnup cladding samples. The initial criterion for pre-hydrided cladding was pass-fail: cladding samples that survived 1.7-mm displacement with $\leq 50\%$ cracks through the wall were classified as ductile, while samples with $>50\%$ wall cracks were classified as brittle. This criterion was relatively simple to apply to samples tested with the Instron 5566 machine. Although load-displacement curves were recorded, post-RCT visual inspection at low magnification was used to assess the extent of wall cracking. However, this method was too conservative as it did not distinguish between samples that developed $>50\%$ wall cracks during elastic deformation (clearly brittle) from samples that developed $>50\%$ wall cracks after 5 to 10% offset strain (clearly ductile). In order to determine the DBTT, a more realistic embrittlement criterion was needed. It was also quite clear that careful analysis of each load-displacement curve was needed, along with post-test visual and optical-microscopy imaging, to determine the offset strain prior to $>50\%$ wall cracking.

It is challenging to determine offset displacement and strain prior to $>50\%$ wall cracking for 5 mm/s displacement rate tests because: (a) multiple cracks may be associated with a single load drop; (b) multiple load drops may be observed during the 1.7-mm displacement test; (c) cracks that initiated early in the test may grow during the test prior to post-test observation; and (d) even for a single wall crack, the crack depth may vary along the axis of the ring. There is no unique relationship between the observed load drops and the location and extent of cracks that propagate partially through the wall. Two approaches were used to determine the extent of wall cracking: (a) for rings that cracked during the linearized loading phase, advanced strength of materials, FEA calculations, and observations from slow-displacement-rate tests were used to correlate the decrease in loading slope with extent of wall cracking and (b) for rings that cracked in the elastic-plastic deformation regime and exhibited only one crack, the percent load drop was correlated with crack depth based on post-test examinations.

Experimental and analytical results for tests conducted at 2 mm/min can be used to better correlate the decrease in loading slope with the extent of wall cracking. Figure 30 shows the load-displacement response for a high-burnup Zry-4 cladding ring sectioned from an Argonne creep sample that was cooled at 60°C/h at high, constant hoop stress following about 100 days of creep at 400°C [7]. The RCT was conducted at RT and was stopped after the second load drop. Post-test observation at low magnification indicated two cracks: (a) one at the 6 o'clock orientation that extended from the inner surface through about 60% of the wall and (b) one at the 9 o'clock position that extended from the outer-surface through $>90\%$ of the wall. FEA calculations were performed to determine that the 6 o'clock crack developed first followed by 9 o'clock crack. The decrease in reloading slope (i.e., stiffness) was 50% following the load drop and observed for the 60% wall crack. This led to the criterion that $>50\%$ reduction in loading slope implies a crack extending through $>50\%$ of the wall. The load drops of 33% and 41% are interesting, but they occur so fast that they tend to not be observed during the loading phase for the 5-mm/s displacement rate tests.

Fig. 30. Load-displacement curve for a Zry-4 ring from a high-burnup HBR rod following about 100 days of creep at 400°C and 190-MPa hoop stress and cooling at 60°C/h to 200°C under constant stress. The RCT was conducted at RT and 2-mm/min displacement rate.

Based on information presented in the following sections, it was determined empirically that a load drop of about 25% in the elastic-plastic deformation regime corresponded to a single crack through about 50% of the wall. Thus, offset strain in this deformation regime was determined from the point on the curve just prior to >25% load drop.

4.2.1 RCT Results for High-Burnup ZIRLO™ from Rodlet 648G

Following RHT at 400°C and 140-MPa hoop stress, three rings were sectioned from high-burnup ZIRLO™ rodlet 648G (see Fig. 14). Rings 5 and 10 were tested at 5 mm/s displacement rate and 150°C to 2-mm crosshead displacement with about a 20-s hold time prior to unloading. Load-displacement curves for these two rings are shown in Figs. 31 and 32, respectively. Based on Fig. 31, cracking initiated at a relatively low load during the elastic loading phase. The load drop was 21% prior to reloading at a slope that was 64% less than the initial loading slope. This was followed by a steep load drop of 61%. The offset displacement and strain were determined to be zero for this ring based on the >50% decrease in reloading slope. Post-test visual examination indicated extensive cracking ($\leq 90\%$) at the 12 and 6 o'clock positions. Post-RHT cracks for this ring are shown in Fig. 33.

For ring 10, a large load drop of about 80% occurred after only 1.4% offset strain. Therefore, 1.4% was used as the offset strain for this ring. Based on post-RCT visual examinations, ring 10 exhibited through-wall cracks at the 12 and 6 o'clock orientations, as well as a partial-wall crack at the 3 o'clock orientation.

For ring 3, the control TC was set at 195°C. The RCT was initiated when the side wall TCs at the 3 and 9 o'clock positions were 190°C and 200°C giving a ring temperature of $195 \pm 5^\circ\text{C}$. The load-displacement curve is shown in Fig. 34. Load drops of 11%, 15% were observed prior to the smooth load increase to 291 N followed by the gradual load drop to 273 N. The post-RCT visual exam indicated a through-wall crack (100%) at the 6 o'clock orientation as observed from both ends of the ring. Cracks were also observed on one end at the 1 and 12 o'clock orientations, but not at the other end. These cracks were $45 \pm 45\%$ and $40 \pm 40\%$ of the wall. Based on the load-displacement curve, it is difficult to assess the offset strain prior to 50% wall cracking. While the observed post-test cracks were extensive, load drops were not. The conservative approach taken for this case was to assume that the 11% and 15% load drops were for a single crack with a combined load drop of 26%. Just prior to the second load drop of 15%, the corrected offset strain was 4.5%. Additional stable crack growth may have occurred during the load drop from 291 N to 273 N. If it were assumed that cracks were <50% of the wall up to this maximum load, then the offset strain would be about 9%.

The DBTT for rodlet 648G was clearly between 150°C (brittle) and 195°C (ductile). Using 4.5% as the offset strain at 195°C, the DBTT would be about 185°C.

Fig. 31. Load-displacement curve for ring 5 from high-burnup ZIRLO™ rodlet 648G following 5°C/h cooling from 400°C and 140-MPa hoop stress. RCT was conducted at 150°C. Offset strain was zero for this ring with 65±17% RHCF and 650±190 wppm C_H.

Fig. 32. Load-displacement curve for high-burnup ZIRLO™ rodlet 648G ring 10 following 5°C/h cooling from 400°C and 140-MPa hoop stress. RCT was conducted at 150°C. Offset strain was 1.4% for this ring with 65±17% RHCF and 650±190 wppm C_H.

(a) Side view

(b) Tilted side view

Fig. 33. Photographs of ring 648G5 following 140-MPa RHT and ring compression at 150°C and 5 mm/s to 2-mm displacement: (a) side view and (b) tilted side view.

Fig. 34. Load-displacement curve for ring 3 from high-burnup ZIRLO™ rodlet 648G following 5°C/h cooling from 400°C and 140-MPa hoop stress. RCT was conducted at 195°C. Offset strain was 4.5% for this ring with 65±17% RHCF and 650±190 wppm C_H .

4.2.2 RCT Results for High-Burnup ZIRLO™ from Rodlet 648D

Following RHT at 400°C and 110-MPa hoop stress, four RCT rings were sectioned from high-burnup ZIRLO™ rodlet 648D (see Fig. 17). Rings 5, 8, and 10 were tested at 5 mm/s displacement rate and 150°C to 2-mm crosshead displacement with about a 20-s hold time prior to unloading. Ring 3 was tested at 30°C to 1.7-mm crosshead displacement and controlled unloading at 5 mm/s. Load displacement curves for these four RCTs are shown in Figs. 35-38. For ring 5 (Fig. 35), the load-displacement curve had no sharp load drops indicative of unstable crack growth. However, post-RCT inspection indicated >50% wall crack at the 6 o'clock orientation and partial wall cracks at the 3 and 9 o'clock orientations. Some of this crack initiation and growth may have occurred in a stable mode during the gradual load drop (511 N to 471 N) or even during the 20-s hold at 2 mm prior to unloading. To be consistent with offset strain measurements for tests conducted to 1.7-mm total displacement, the 9.5% offset strain was determined based on a total displacement of 1.7 mm. For ring D8 (Fig. 36), a 28% steep load drop occurred before 1.7-mm total displacement. The offset strain at the peak load prior to the load drop was 8.6%. Post-RCT visual observation indicated a single crack that was 60±30% of the wall thickness at the 3 o'clock orientation: 90% of the wall at one end and 30% of the wall at the other end. Similar results were obtained for ring 10 (Fig. 37). However, the 29% load drop occurred just beyond 1.7-mm ring displacement. The offset strain of 9.0% was determined for 1.7-mm total displacement. The results for these three RCTs conducted at 150°C were consistent: about 9.0±0.5% offset strain.

RCT results for ring 3 tested at 30°C (Fig. 38) displayed a 22% load drop followed by a decrease of >50% in the linear slope and a 54% load drop. Beyond 0.6% offset strain, no plastic strain was observed until after the 54% load drop. Thus, the offset strain was taken to be 0.6% prior to >50% wall cracking. Post-RCT visual examination of the ring revealed long cracks (≤95%) at all four critical locations: 12, 3, 6, and 9 o'clock orientations.

The DBTT could not be determined from rodlet 648D RCT data. However, 648D results are combined in the next section with rodlet 648C results to determine the DBTT for high-burnup ZIRLO™ cladding following RHT cooling from 400°C and 110-MPa hoop stress.

Fig. 35. Load-displacement curve for ring 5 from high-burnup ZIRLO™ rodlet 648D following 5°C/h cooling from 400°C and 110-MPa hoop stress. The RCT was conducted at 150°C. Offset strain was 9.5% for this ring with 27±10% RHCF and 425±63 wppm C_H .

Fig. 36. Load-displacement curve for ring 8 from high-burnup ZIRLO™ rodlet 648D following 5°C/h cooling from 400°C and 110-MPa hoop stress. The RCT was conducted at 150°C. Offset strain was 8.6% for this ring with 27±10% RHCF and 425±63 wppm C_H .

Fig. 37. Load-displacement curve for ring 10 from high-burnup ZIRLO™ rodlet 648D following 5°C/h cooling from 400°C and 110-MPa hoop stress. RCT was conducted at 150°C. Offset strain was 9.0% for this ring with 27±10% RHCF and 425±63 wppm C_H.

Fig. 38. Load-displacement curve for ring 3 from high-burnup ZIRLO™ rodlet 648D following 5°C/h cooling from 400°C and 110-MPa hoop stress. The RCT was conducted at 30°C. Offset strain was 0.6% for this ring with 27±10% RHCF and 425±63 wppm C_H.

4.2.3 RCT Results for High-Burnup ZIRLO™ from Rodlet 648C

Following RHT at 400°C and 110-MPa hoop stress, four RCT rings were sectioned from high-burnup ZIRLO™ rodlet 648C (see Fig. 19). Rings were tested at 5-mm/s loading displacement rate to 1.7-mm crosshead displacement and at 5-mm/s unloading displacement rate. Test temperatures were: 150°C for ring 8, 120°C for ring 10, 90°C for 3, and 30°C for ring 5. The test sequence was 150°C (highly ductile), 30°C (brittle), 90°C (brittle), and 120°C (brittle). Load displacement curves for these four RCTs are shown in Figs. 39-42.

Ring C8, which was tested at 150°C, exhibited a very small load drop at the early stage of plastic deformation (see Fig. 39). Post-test examination of the ring indicated no cracking. The small load drop may have been associated with minor corrosion/hydride-rim layer cracking on the cladding outer surface, which would not be visible due to corrosion layer cracks formed during RHT. At 1.7-mm total displacement, the traditional (using the loading slope K_L to unload the ring) offset strain was 12.2%, the corrected (using the reduced slope K_U to unload the ring) offset strain was 10.2%, and the permanent strain was 9.4%. The difference between the corrected offset strain and the measured permanent strain was <1%. These data were used to develop the correlation (Eq. 2) for unloading slope as a function of loading slope and traditional offset strain.

Ring 10, which was tested at 120°C, exhibited a 33% abrupt load drop after 1.7% offset strain and a 37% abrupt load drop at about 1.6-mm displacement (see Fig. 40). Post-RCT visual examination indicated a major crack at the 6 o'clock orientation ($70\pm 20\%$ – 90% crack at one end and 50% crack at the other end) and minor cracks at the 3 ($25\pm 25\%$) and 7 ($40\pm 20\%$) o'clock orientations. Although the load-displacement curve does not indicate the sequence of crack initiation and propagation, post-RCT examination results are consistent with the formation of a >50% wall crack after 1.7% offset strain. This offset strain is marginally below the 2% ductile-to-brittle transition strain.

Ring 3, which was tested at 90°C, exhibited abrupt load drops of about 50% after 0.3% offset strain and after about 2.35-mm total displacement (see Fig. 41). Post-RCT visual examination indicated major cracks ($\leq 95\%$) at the four critical locations: 12, 3, 6, and 9 o'clock orientations. Based on these results, it appears that each abrupt load drop represented the simultaneous formation of two cracks.

Ring 5, which was tested at 30°C, appeared to crack during the linearized loading ramp (see Fig. 42). Although the load drop was very small, the decrease in reloading stiffness (about 70%) after 0.6% offset strain was greater than 50%, which is indicative of a crack >50% of the wall thickness. Subsequent abrupt load drops were 44% and 46%. Post-RCT visual examination indicated major cracks ($\approx 100\%$) at the 12 and 6 o'clock orientations and a partial wall crack at the 9 o'clock orientation. The offset strain was assessed to be $0.3\pm 0.3\%$.

The 648C RCT results can be combined with the 648D RCT results because the peak RHT stress was the same for both rodlets and the RHCF values ($27\pm 10\%$ and $33\pm 13\%$) overlapped. For the combined datasets, RHCF = $30\pm 12\%$ and DBTT $\approx 125^\circ\text{C}$.

Fig. 39. Load-displacement curve for ring 8 from high-burnup ZIRLO™ rodlet 648C following 5°C/h cooling from 400°C and 110-MPa hoop stress. RCT was conducted at 150°C. Offset strain was 10.2% for this ring with 33±13% RHCF and 350±80 wppm C_H.

Fig. 40. Load-displacement curve for ring 10 from high-burnup ZIRLO™ rodlet 648C following cooling 5°C/h from 400°C and 110-MPa hoop stress. RCT was conducted at 120°C. Offset strain was 1.7% for ring with 33±13% RHCF and 350±80 wppm C_H.

Fig. 41. Load-displacement curve for ring 3 from high-burnup ZIRLO™ rodlet 648C following 5°C/h cooling from 400°C and 110-MPa hoop stress. RCT was conducted at 90°C. Offset strain was 0.4% for this ring with 33±13% RHCF and 350±80 wppm C_H.

Fig. 42. Load-displacement curve for ring 5 from high-burnup ZIRLO™ rodlet 648C following 5°C/h cooling from 400°C and 110-MPa hoop stress. RCT was conducted at 30°C. Offset strain was 0.3% for this ring with 33±13% RHCF and 350±80 wppm C_H.

4.2.4 RCT Results for High-Burnup Zry-4 from Rodlet 605C6

Following RHT at 400°C and 110-MPa hoop stress, four RCT rings were sectioned from high-burnup Zry-4 rodlet 605C6 (see Fig. 24). Rings were tested at 5-mm/s loading displacement rate to 1.7-mm crosshead displacement and at 5-mm/s unloading displacement rate. Test temperatures were: 150°C for ring H, 120°C for ring J, 90°C for C, and 24°C for ring 605C6E. The test sequence was 150°C (highly ductile), 24°C (low ductility), 90°C (intermediate ductility), and 120°C (intermediate ductility). Load displacement curves for these four RCTs are shown in Figs. 43-46.

Ring H, which was tested at 150°C, exhibited a small load drop after $\approx 3\%$ offset strain (see Fig. 43). Post-test examination of the ring indicated no cracking. The small load drop may have been associated with minor corrosion/hydride-rim layer cracking on the cladding outer surface, which would not be visible due to corrosion layer cracks formed during RHT. At 1.7-m total displacement, the traditional (using the loading slope K_L to unload the ring) offset strain was 11.1%, the corrected (using the reduced slope K_U to unload the ring) offset strain was 9.2%, and the permanent strain was 8.4%. The difference between the corrected offset strain and the measured permanent strain was $<1\%$. These data were used to develop the correlation (Eq. 3) for unloading slope as a function of loading slope and traditional offset strain.

Ring J, which was tested at 120°C, exhibited two small load drops prior to a 27% abrupt load drop after 6.7% offset strain (see Fig. 44). Post-RCT visual examination indicated a single crack at the 3 o'clock orientation, which extended from the outer surface through $45 \pm 5\%$ of the wall thickness. Although this is slightly below the $>50\%$ wall crack criterion, it was used to determine the corrected offset strain of 6.7%. If the point at 1.7-mm total displacement had been used, there would have been considerable uncertainty in the unloading slope. The post-test permanent strain was 8.7%, which set the upper bound on offset strain.

Ring C, which was tested at 90°C, exhibited a load drop of about 12% prior to the 50% load drop just after 5.0% offset strain (see Fig. 45). Post-RCT visual examination indicated a through-wall crack at the 3 o'clock orientation and a partial crack at the 9 o'clock orientation. Metallographic examination (see Fig. 47) showed that these cracks initiated at the outer cladding wall, propagated in a brittle mode through the hydride rim, and then propagated along radial and circumferential hydrides to a depth of about 70% of the wall.

Ring E, which was tested at 24°C, exhibited two small load drops prior to a 51% load drop just after an offset strain of 2.8% (see Fig. 46). Post-RCT visual examination indicated major cracks at the 3 ($65 \pm 10\%$) and 9 ($80 \pm 20\%$) o'clock orientations. The offset strain was 2.8%.

The DBTT for this set of ring tests was assessed to be $<20^\circ\text{C}$ based on the observed 2.8% ductility at 24°C. The DBTT was low due to the low RHCF ($9 \pm 5\%$) and the location of the short radial hydrides (near the cladding mid-wall).

Fig. 43. Load-displacement curve for ring H from high-burnup Zry-4 rodlet 605C6 following 5°C/h cooling from 400°C and 110-MPa hoop stress. RCT was conducted at 150°C. Offset strain was 9.2% for this ring with 9±5% RHCF and 520±90 wppm C_H .

Fig. 44. Load-displacement curve for ring J from high-burnup Zry-4 rodlet 605C6 following 5°C/h cooling from 400°C and 110-MPa hoop stress. RCT was conducted at 120°C. Offset strain was 6.7% for this ring with 9±5% RHCF and 520±90 wppm C_H .

Fig. 45. Load-displacement curve for ring C from high-burnup Zry-4 rodlet 605C6 following 5°C/h cooling from 400°C and 110-MPa hoop stress. RCT was conducted at 90°C. Offset strain was 5.0% for this ring with 9±5% RHCF and 520±90 wppm C_H.

Fig. 46. Load-displacement curve for ring E from high-burnup Zry-4 rodlet 605C6 following 5°C/h cooling from 400°C and 110-MPa hoop stress. RCT was conducted at 24°C. Offset strain was 2.8% for this ring with 9±5% RHCF and 520±90 wppm C_H.

(a) 9 o'clock orientation

(b) 3 o'clock orientation

Fig. 47. Post-RCT cracks observed in ring C tested at 90°C from high-burnup Zry-4 rodlet 605C6 subjected to 110-MPa hoop stress at 400°C for 8 hours prior to cooling at 5°C/h: (a) 9 o'clock orientation and (b) 3 o'clock orientation.

4.2.5 RCT Results for High-Burnup Zry-4 from Rodlet 605C2

Following RHT at 400°C and 140-MPa hoop stress, three RCT rings were sectioned from high-burnup Zry-4 rodlet 605C2 (see Fig. 27). Rings were tested at 5-mm/s loading displacement rate to 1.7-mm crosshead displacement and at 5-mm/s unloading displacement rate. Test temperatures were: 150°C for ring C, 90°C for ring F, and 30°C for ring G. The test sequence was 150°C (intermediate ductility), 30°C (brittle), and 90°C (low ductility). Load-displacement curves for these three RCTs are shown in Figs. 48-50.

Ring C, which was tested at 150°C, exhibited a small load drop and a 12% load drop prior to a 27% load drop just after 5.8% offset strain (see Fig. 48). Post-RCT visual observation indicated a single wall crack (60%) at the 3 o'clock orientation. Metallographic examination was used to determine that the crack depth was 70% of the wall thickness (see Fig. 51). The crack originated at the outer cladding surface, propagated in a brittle mode through the hydride rim and substrate though about 40% of the cladding wall, and then propagated in a zigzag manner along radial and circumferential hydrides an additional 30% of the wall. Based on this crack pattern, it appears reasonable to determine the offset strain based on displacement just prior to the 27% wall crack.

Ring F, which was tested at 90°C, exhibited two small load drops prior to a 26% abrupt load drop after 3.3% offset strain (see Fig. 49). Subsequent load drops of 26%, 39% and 63% indicated significant cracking at multiple orientations. Post-RCT visual examination indicated major cracks at the 3 (70%) and 9 (95±5%) o'clock orientations, as well as a minor crack at the 6 o'clock orientation (20±10%). The 3.3% offset strain prior to the 27% load drop at 776 N may be a slight over-estimate of the ductility. However, it is quite likely that the actual offset strain would be >2%. As such, the ring is assessed as having low ductility.

Ring G, which was tested at 30°C, exhibited a 90% load just after 1.2% offset strain (see Fig. 50). This load-displacement curve is relatively straightforward to analyze. The 90% load drop is characteristic of multiple cracks propagating deep into the cladding wall. Post-RCT visual examination indicated major cracks at the 4 (90%), 6 (50%) and 9 (70%) o'clock orientations.

The DBTT for this set of ring tests was assessed to be about 55°C based on the observed embrittlement at 30°C and the 3.3% offset strain at 90°C.

Fig. 48. Load-displacement curve for ring C from high-burnup Zry-4 rodlet 605C2 following 5°C/h cooling from 400°C and 140-MPa hoop stress. The RCT was conducted at 150°C. Offset strain was 5.8% for this ring with 16±4% RHCF and 615±82 wppm C_H.

Fig. 49. Load-displacement curve for ring F from high-burnup Zry-4 rodlet 605C2 following 5°C/h cooling from 400°C and 140-MPa hoop stress. RCT was conducted at 90°C. Offset strain was 3.3% for this ring with 16±4% RHCF and 615±82 wppm C_H.

Fig. 50. Load-displacement curve for ring G from high-burnup Zry-4 rodlet 605C2 following cooling 5°C/h from 400°C and 140-MPa hoop stress. RCT was conducted at 30°C. Offset strain was 1.2% for this ring with 16±4% RHCF and 615±82 wppm C_H.

Fig. 51. Post-RCT image of crack extending through 70% of the wall of ring 605C2C, which was tested at 150°C following 5°C/h cooling from 400°C and 140-MPa hoop stress.

4.2.6 Summary of RCT Results for High-Burnup Cladding following RHT

Table 12 summarizes the RHT conditions, C_H , RHCF, and RCT results for high-burnup cladding rodlets and rings sectioned from those rodlets. The datasets for ZIRLO™ rodlets 648D and 648C were combined as the only difference in RHT was the hold time at 400°C: 1 hour for 648D and 24 hours for 648C. The RHCF values for these rodlets were comparable: 27±10% for 648D and 33±13% for 648C. The combined RHCF value for the two rodlets was 30±12%. Figure 52 shows the offset strain data in Table 12 plotted as a function of RCT temperature. The trend curves for high-burnup ZIRLO™ are represented by hyperbolic tangent functions. Based on a ductile-to-brittle transition strain of 2% (see Discussion), the DBTT values can be approximated for each cladding material and set of test conditions: 185°C for high-burnup ZIRLO™ with 65±17% RHCF, 125°C for high-burnup ZIRLO™ with 30±12% RHCF, 55°C for high-burnup Zry-4 with 16±4% RHCF, and <20°C for high-burnup Zry-4 with 9±5% RHCF.

Table 12

Summary of RCT results for RHT high-burnup rodlets subjected to simulated drying-storage temperature histories with a maximum temperature of 400°C, 400°C hoop stresses of 110 MPa or 140 MPa, and cooling at 5°C/h with decreasing hoop stress.

Material	ID	RHT σ_θ at 400°C, MPa	C_H , wppm	RHCF, %	RCT T, °C	Corrected Offset Stain, %	Permanent Strain, %
ZIRLO™	648G	140	650±190	65±17			
	5				150	0.0	---
	10				150	1.4	---
	3				195	4.5	---
ZIRLO™	648D	110	425±63	27±10			
	3				30	0.6	---
	5				150	9.5	---
	8				150	8.6	---
	10				150	9.0	---
ZIRLO™	648C	110	350±80	33±13			
	5				30	0.3	---
	3				90	0.4	---
	10				120	1.7	---
	8				150	10.2	9.4
Zry-4	605C6	110	520±90	9±5			
	E				24	2.8	---
	C				90	5.0	---
	J				120	6.7	---
	H				150	9.2	8.4
Zry-4	605C2	140	615±82	16±4			
	G				30	1.2	---
	F				90	3.3	---
	C				150	5.8	---

Fig. 52. Offset strain (measure of ductility) as a function of RCT temperature for high-burnup ZIRLO™ and Zry-4 cladding following simulated drying-storage conditions: heating pressurized rodlets to 400°C and to the indicated hoop stresses in the legend, holding at 400°C for 1 to 24 hours, and cooling at 5°C/h with decreasing pressure and stress.

5. DISCUSSION

5.1 Hydride Dissolution, Diffusion, Solubility, and Precipitation

According to Kearns [2, 18, 19], dissolution of hydrides and hydrogen diffusion during the heating phase are very rapid in non-irradiated Zr, Zry-2, and Zry-4. These results apply to cladding without a hydride rim. The terminal solubility in these alloys was determined to be 207 wppm at 400°C. The results for Zry-4 are consistent with the results of other researchers such as Kammenzind et al. [3]. Also, Kearns and Kammenzind et al. measured relatively low diffusion-coefficient activation energies. For the one-cycle temperature history and the materials used in the Argonne tests, hydrogen would have ample time to dissolve and diffuse across the cladding wall (0.54-0.70 mm) during the heating and hold-temperature phase of the cycle.

While hydride dissolution is very fast during increasing temperatures, precipitation of hydrides is more difficult and results in a supersaturated hydrogen content prior to initiation of precipitation. As Zr hydrides have lower density than Zr metal, precipitation causes a local volume increase that has to be accommodated by the cladding metal. For precipitation of isolated hydrides, the hydride thickness grows under increasing compressive stress, while the metal experiences increasing tensile stress. The difference between dissolution (higher T_s) and precipitation (lower T_p) temperatures is referred to as hysteresis and is characterized by the temperature gap ($T_s - T_p$). McMinn et al. [20] observed a temperature gap of 46°C in Zry-2 and Zry-4 for dilute hydrogen contents (≤ 80 wppm). Kammenzind et al. [3] studied the same phenomenon for higher hydrogen contents (< 500 wppm) in Zry-4. Based on interpolation of their data, the measured temperature gap was 65°C for 210 wppm hydrogen dissolved at 400°C. The materials tested in both of these studies were in the stress-free condition.

For stress free Zr-alloys, the hysteresis gap is about $55 \pm 10^\circ\text{C}$ between dissolution and initiation of precipitation of circumferential hydrides for 100% dissolution of hydrogen at peak temperature. However, if hydrides are present at the peak temperature, precipitation can occur during cooling from the peak temperature with no hysteresis gap. For this relevant case, the hysteresis gap applies to precipitation of new hydrides at locations other than those of the hydrides present at the peak temperature. Regardless of the magnitude of the hysteresis gap, the stress level in high-burnup cladding rodlets at the initiation of precipitation of new hydrides would decrease by $< 10\%$ for a temperature gap of $< 65^\circ\text{C}$, neglecting the impact of thermal creep on the free volume of rodlets.

5.2 High-Burnup Cladding vs. Pre-Hydrided Cladding

The purpose of tests with PH Zry-4 and ZIRLO™ was to map out peak drying-storage hoop stresses vs. hydrogen content for which radial hydrides either did or did not embrittle the cladding at 150°C RCT temperature. Reference conditions for these tests were: 1-h hold time and 150-MPa hoop stress at 400°C. For hydrogen contents (350-650 wppm) of interest for high-burnup ZIRLO™ and Zry-4 cladding, rings sectioned from PH rodlets exhibited high ductility ($> 10\%$) at 150°C with all but one of the rings surviving the RCT with no indication of cracking. However, these high-ductility matrix samples with uniform distributions of hydrides across the radius proved to be poor surrogates for high-burnup cladding. The high-burnup ZIRLO™ rodlet

(650-wppm hydrogen) subjected to the same thermal cycle at slightly lower stress (140 MPa at 400°C) behaved in a highly brittle manner at 150°C RCT temperature. The primary difference in test materials causing such a dramatic difference in results was the pre-test radial distribution of hydrides: uniform for pre-hydrided cladding and highly non-uniform for high-burnup cladding.

5.3 *Metric for Radial-Hydride Precipitation*

As discussed in Section 3.1, the RHCF is assessed as being a better metric than RHF or HCC for correlating hydride morphology and orientation with extent of cracking during RCT loading. It represents the length of continuous radial-circumferential hydrides across the cladding wall (normalized to the wall thickness) within a circumferential band of 0.15 mm. However, the RHCF does not distinguish radial-circumferential hydrides emanating from the inner cladding surface from those located near the cladding mid-wall or those located near the cladding outer wall. In the RCT, the hoop tensile stress is highest at the cladding inner surfaces below the applied load and above the support plate. Continuous radial-circumferential hydrides emanating from this inner surface would induce crack initiation and propagation with very small or no plastic deformation. Continuous radial-circumferential hydrides emanating from the outer surface just under the hydride rim would participate in unstable crack growth initiated in the rim. However, as shown by Billone [21], the elastic tensile stress at the outer surfaces located at the 3 and 9 o'clock orientations are only 60% of those located at the 12 and 6 o'clock inner-surface orientations. Thus, it is possible that these inner-surface locations could experience some plastic deformation prior to crack initiation at the 3 and 9 o'clock outer-surface orientations. Continuous radial-circumferential hydrides located near the cladding mid-wall would be subjected to very low hoop-stress levels during RCT loading. As such, they might participate in crack propagation following some plastic deformation. These radial hydrides are the least degrading with respect to embrittlement. Also, for the initial RCT point loading, the maximum hoop stress at this inner-surface location is highly localized. For 17×17 cladding, at arc lengths of ±0.5 mm (about the length of one 100X micrograph) from the loading point, the maximum elastic tensile hoop stress decreases by 18%. A more complete metric or set of metrics would take into account RHCF, radial-hydride location, radial-hydride spacing, and perhaps radial-hydride thickness in order to better indicate the susceptibility of RHT cladding to radial-hydride induced embrittlement.

5.4 *Brittle-to-Ductile Transition Strain for Ring Compression Tests*

Several factors were included in the selection of 2% offset strain as the ductile-to-brittle transition for RCTs. Based on extensive experience, rings with $\geq 1\%$ permanent strain (d_p/D_{mo}) were classified as ductile, while rings with $< 1\%$ permanent strain were classified as brittle because these low values were within the measurement uncertainty band. Secondly, there is an inherent error in using the traditional offset strain (δ_p/D_{mo}) method to determine ductility because the unloading slope is always less than the loading slope. As shown in Fig. 53, this error increases as δ_p/D_{mo} increases. For AF cladding, the error was only 0.3% for traditional offset strains of about 2%. Thus, the offset strain should be $\geq 1.3\%$ for AF cladding to be classified as ductile. However, the offset-strain error for high-burnup cladding with a corrosion layer and a hydride rim was found to be greater than the error for AF cladding. Also, as shown by Billone [21], the relative displacement determined by δ_p/D_{mo} is less than the maximum material strain

Fig. 53. Inherent error in the determination of ductility from RCT load-displacement curves using the traditional offset displacement and strain method. AF and HB are as-fabricated and high-burnup, respectively, cladding rings.

in the elastic deformation regime. Finally, there are uncertainties in determining low values of offset strain for samples that exhibit small load drops indicative of crack formation prior to 2% offset strain. As such, the ductility criterion based on offset strain is $\geq 2\%$. Although a correction was made in the current work for better determination of offset strains $> 2\%$, the correction does not apply to offset strains $\leq 2\%$.

Another important consideration in the ductile vs. brittle classification is the use of the 50% wall-crack limit. This conservatism was incorporated into the criterion because displacement rates may be higher than the one used in the RCTs and loads other than pinch-type loads are present during normal and accident conditions for dry-cask storage and transport. The internal gas pressure, although low at low fuel-rod temperatures, would contribute a tensile hoop stress. Bending in the axial direction would contribute tensile and compressive axial stresses, as well as hoop stresses due to radial loads applied to the cladding by the fuel. For cladding without fuel, the cross section would deform from an initial circular shape to an elliptical shape. For fueled cladding, the fuel resists decreases in cladding inner diameter by applying a secondary radial load to the cladding. Ductility of homogeneous metals tends to decrease as the biaxiality ratio (σ_θ/σ_z) $\rightarrow 1$ [22] and may decrease at higher displacement-rate loading. As such, use of offset strain determined just prior to development of a 100% wall crack due to RCT loading would lead to overly optimistic results. While conservative for RCT loading, testing of post-RHT samples for a relevant range of σ_θ/σ_z values and strain rates would be needed to determine if the $\leq 50\%$ wall-crack criterion is conservative enough for high-burnup cladding during

transport. Given that the ductility of irradiated Zry-4 is relatively strain-rate insensitive [1] and high-burnup fuel cladding with radial hydrides is not homogeneous, it is likely that the failure criterion used in the current work would still be conservative at higher strain-rate loading and for biaxiality ratios in the range of 1 to 2.

It should be noted that DBTT in the current work corresponds to the RCT temperature for which the corrected offset strain is 2% (ductility limit). Literally, it is the temperature at which the material transitions from brittle to ductile behavior. This definition differs from the one used to determine DBTT under impact test conditions (e.g., Charpy test). For impact tests, materials exhibit lower and upper shelf failure energies. DBTT for these tests is defined as the average of the highest lower-shelf temperature and the lowest upper-shelf temperature.

5.5 DBTT for RHT Cladding

RCT results indicate a significant difference in extent of radial hydride formation (i.e., RHCF) and DBTT for the high-burnup ZIRLO™ rodlets tested as compared to the high-burnup Zry-4 rodlets tested following simulated drying-storage for the same temperature and stress histories. For 140-MPa hoop stress at 400°C, the RHCF and DBTT values were: a) 65±17% and about 185°C for ZIRLO™ and b) 16±4% and 55°C for Zry-4. For 110-MPa hoop stress at 400°C, the RHCF and DBTT values were: a) 30±12% and about 125°C for ZIRLO™ and b) 9±5% and <20°C for Zry-4. For high-burnup ZIRLO™ with 140-MPa RHT, additional RCT data are needed in the range of 150°C (two brittle data points) and 195°C (one ductile data point) to better determine the DBTT. Several factors were considered in evaluating the DBTT results: a) pre-test radial distribution of hydrides, b) texture, c) residual stresses, and d) alloy effects. Based on metallographic examination of sibling pre-test samples, the hydride rim was denser and more localized for high-burnup ZIRLO™ cladding as compared to the rim in high-burnup Zry-4 cladding. More significantly, the average hydrogen content below the hydride rim appeared to be higher for Zry-4 than for ZIRLO™. Based on optical-microscopy images, it was estimated that the average hydrogen content in the inner two-thirds of the Zry-4 segments was >200 wppm, while the average hydrogen content in the inner two-thirds of ZIRLO™ segments was <150 wppm. This estimate did not include the likely presence of microscopic hydrides (≈100 to 200 nm long and ≈30 to 50 nm long [23]) and hydrogen trapped in irradiation defects, neither of which are considered to be damaging to the material. As shown in Fig. 13 differences in pre-test hydrogen content can have a significant effect on RHCF, especially if circumferential hydrides are present at the peak drying temperature. For high-burnup Zry-4, precipitation of hydrogen at these circumferential hydrides would occur during cooling from 400°C to T_p , while precipitation of new hydrides (circumferential and radial) would initiate at T_p . The radial distribution of hydrides in pre-RHT, high-burnup Zry-4 would favor precipitation of circumferential hydrides at the RHT stresses used in this test program. However, the hydrogen content for high-burnup Zry-4 below the hydride rim decreased significantly from about 67% of the cladding wall to the inner surface. The absence of pre- and post-RHT hydrides at the Zry-4 inner surface suggests that the difference between results for high-burnup ZIRLO™ and Zry-4 was not due solely to differences in the pre-test radial distribution of hydrides. To resolve this issue, additional data would be needed for high-burnup Zry-4 with 300-400 wppm hydrogen for which the hydride rim is more concentrated and the hydrogen content is <200 wppm within the inner two-thirds of the cladding wall. Also, local regions of radial hydrides were observed emanating from the inner surface of

as-irradiated ZIRLO™ while none were observed in extensive metallographic examination of axial regions from seven Zry-4-clad fuel rods irradiated in the same assembly to high burnup. The results suggest that high-burnup ZIRLO™ is more susceptible than high-burnup Zry-4 to radial-hydride precipitation during reactor shut down, as well as during simulated drying-storage conditions.

Microstructure (i.e., orientation of grains and grain boundaries) has been shown to have a significant impact on the susceptibility of cladding materials to radial-hydride precipitation during cooling under tensile hoop stress. Aomi et al. [6] observed that high-burnup RXA Zry-2 formed radial hydrides at lower stress levels than high-burnup CWSRA Zry-4. The higher susceptibility of the RXA microstructure is likely due to the more random orientation of grains and grain boundaries as compared to CWSRA materials. The ZIRLO™ and Zry-4 used in the Argonne test program were CWSRA materials. Zry-4 fabrication details were provided to Argonne for this cladding manufactured in 1977 and used to fabricate the HBR fuel rods. However, no such details were provided for the ZIRLO™ cladding used to fabricate the North Anna rods, for which irradiation was initiated in 1987. It is possible that there were microstructural differences between these two cladding materials that may have contributed to differences in results.

Residual tensile hoop stresses from fabrication and irradiation can impact radial hydride precipitation. Following stress-free hydrogen pre-charging of AF RXA Zry-2 with a Zr liner on the inner surface, Billone and Burtseva observed radial hydrides emanating from the cladding outer surface in local regions around the circumference. These radial hydrides were relatively long ($\leq 30\%$ of the Zry-2 thickness). The results suggest that additional processing steps were used following cladding-liner co-extrusion and recrystallization. If local residual stresses were present in AF Zry-2, they would have partially relaxed during irradiation due to creep. Aomi et al. did not observe the presence of these radial hydrides in as-irradiated Zry-2 cladding. Although residual stresses can enhance radial-hydride precipitation, no information is available to suggest that residual tensile hoop stresses are higher at the cladding inner surface for either AF or high-burnup ZIRLO™ as compared to those for AF or high-burnup Zry-4. However, during irradiation and reactor shut down, non-uniform pellet-cladding mechanical interaction (PCMI) can result in post-irradiation residual stresses. The radial hydrides observed in as-irradiated ZIRLO™ cladding [17] may have been caused by higher and more non-uniform PCMI loading than experienced by the high-burnup Zry-4 [14] used in this study.

In terms of alloy composition, North Anna ZIRLO™ differed from HBR Zry-4 in Sn content (about 1 wt.% vs. 1.4 wt.%) and Nb content (about 1 wt.% vs. 0 wt.%). Other Nb-bearing zirconium alloys used in nuclear reactors are RXA M5® (Zr-1%Nb) cladding and CWSRA Zr-2.5%Nb pressure tube material. Data are not publically available for the behavior of M5® under simulated drying-storage conditions. However, even if such data were available, high susceptibility to radial-hydride formation would likely be the result of the RXA microstructure and low hydrogen content (≈ 100 wppm or less) rather than the presence of Nb. Data reported by Wallace et al. [11] indicate that a relatively high tensile stress is required to induce significant radial-hydride precipitation during cooling of CWSRA Zr-2.5%Nb with 60 to 80 wppm hydrogen. Although possible, it is unlikely that the presence of Nb in ZIRLO™ is

responsible for the higher susceptibility to radial hydride precipitation observed in high-burnup ZIRLO™.

5.6 Comparison of Data to Other Work

A number of mechanical test methods may be used to study the influence of radial hydrides on cladding embrittlement: RCT, uniaxial (hoop) ring-stretch test [21], expansion-due-to-compression test [21], and pressurized-tube test ($\sigma_\theta/\sigma_z = 2$). All four test techniques induce tensile hoop stresses. Except for the RCT, the other three test methods induce relatively uniform tensile hoop stresses across the cladding wall, which result in a more severe loading condition that may lead to higher DBTT values. For the RCT ($\sigma_\theta/\sigma_z = 2.8$ at RT for 17×17 cladding within the elastic deformation regime), stresses transition from tensile to compressive across the wall and non-uniform bending moments in the hoop direction result in peak stress regions at and around four orientations (i.e., 12, 3, 6, and 9 o'clock). The RCT was used in the current work because it better simulates pinch-type loading. Aomi et al. [6] did similar testing at RT and a slow displacement rate (2 mm/min) for high-burnup Zry-2 and Zry-4. Their RHT differed from the current work in that pressure and stress were held constant during cooling. Also, most data were generated for the faster cooling rate of 30°C/h. RXA Zry-2 with a Zr-liner showed a high susceptibility to radial hydride precipitation at relatively low stress levels (e.g., 70 MPa) and cooling from 400°C. Based on published metallographic images, the hydrogen content in the cladding outside the liner appears to be quite low (<100 wppm). For high-burnup Zry-4 tested by Aomi et al., hydrogen contents appear to be low relative to the samples used in the current study and the maximum annealing temperature was lower (250°C to 340°C vs. 400°C). The only Zry-4 data point that indicated brittle behavior (<2% offset strain) was for the sample annealed at 340°C and at about 140-MPa hoop stress during 30°C/h cooling. The result is consistent with the Argonne RT data point for high-burnup Zry-4 subjected to 400°C anneal at 140-MPa hoop stress prior to cooling. The Aomi et al. data point for Zry-4 subjected to a 340°C anneal at about 100 MPa indicated ductile behavior at RT. Again, this result is consistent with the Argonne data point for Zry-4 subjected to an RHT with 400°C and 110-MPa hoop stress prior to cooling. For other data in Ref. 6, RHT conditions (250°C to 300°C) were too low to allow a one-to-one comparison.

6. CONCLUSIONS

A test procedure was developed to simulate the effects of elevated temperatures, pressures and stresses during transfer-drying operations and early storage, as well as slow cooling under decreasing pressures and stresses during both drying and storage. Sealed and pressurized rodlets were heated and held at 400°C for 1 to 24 hours and slow cooled at 5°C/h. The procedure was applied to PH and high-burnup ZIRLO™ and Zry-4. It was effective in determining stress levels that resulted in radial-hydride precipitation and the extent of that precipitation. Following this simulated drying-storage thermal cycle, rings sectioned from rodlets were subjected to RCT at temperatures in the range of 20°C to 200°C. The RCT was used as a ductility screening test and to simulate pinch-type loading during cask drops from rod/grid-spacer and rod/rod mechanical interactions. RCT results were used to determine the DBTT of cladding alloys as a function of the peak drying-storage hoop stress. A strong correlation was found between the extent of radial hydride formation across the cladding wall and the extent of wall cracking during RCT loading.

The RHCF, which accounts for the connectivity of radial and circumferential hydrides, was used to assess the extent of radial-hydride precipitation.

Prior to conducting tests with high-burnup cladding, a large number of tests were conducted with non-irradiated PH Zry-4 and ZIRLO™ with uniform distributions of hydrides across the cladding wall. It was observed that the extent of radial-hydride precipitation and the mechanical performance of uniformly PH cladding following simulated drying-storage indicated that PH cladding was not a good surrogate for high-burnup cladding.

The RHCF is defined in the current work as the percentage of cladding wall within a 0.15-mm circumferential band containing continuous radial-circumferential hydrides along which an unstable crack may propagate. For high-burnup Zry-4, RHCF values were relatively low: $9\pm4\%$ for 110-MPa hoop stress at 400°C and $16\pm4\%$ for 140-MPa hoop stress at 400°C. For high-burnup ZIRLO™, RHCF values were relatively high: $30\pm12\%$ for 110-MPa hoop stress at 400°C (based on two rodlets) and $65\pm17\%$ for 140-MPa hoop stress at 400°C. The results indicate that the high-burnup ZIRLO™ tested was more susceptible to radial-hydride precipitation and embrittlement than the high-burnup Zry-4 tested. Along with material differences, differences in irradiation conditions may have contributed to the observed differences in susceptibility to radial hydride precipitation during cooling. The RHCF appears to be a good metric to evaluate the extent of hydride reorientation in these cladding alloys.

The DBTT values for high-burnup Zry-4 following simulated drying-storage at 110 MPa and 140 MPa were $<20^{\circ}\text{C}$ and about 55°C , respectively. Under the same simulated drying-storage conditions, the DBTT values for high-burnup ZIRLO™ were about 125°C and 185°C . Most radial hydrides observed in Zry-4 were relatively short and close to the cladding mid-wall where the RCT hoop bending stresses were very low. They participated in crack propagation but not in crack initiation. For high-burnup ZIRLO™, radial hydrides were found to be relatively long and to extend from the inner cladding surface, which is subjected to the maximum RCT hoop bending stress under the applied load and above the support plate. The DBTT was found to be a function of the cladding material, stress at maximum drying-storage temperature, hydrogen level, and pre-drying radial distribution of hydrogen. The hold time at maximum temperature did not appear to influence the DBTT.

Although additional data would be needed to better determine DBTT, the trend of the data generated in the current work clearly indicates that failure criteria for high-burnup cladding need to include the embrittling effects of radial-hydrides for drying-storage conditions that are likely to result in significant radial-hydride precipitation.

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Appendix A

Post-RHT Metallographic Images for High-Burnup ZIRLO™ Rodlet 648G

Fig. A.1 Sectioning diagram for high-burnup ZIRLO™ rodlet 648G following RHT (i.e., simulated drying-storage) at 400°C peak temperature and 140-MPa peak hoop stress.

Fig. A.2 Area 1 image for ring 648G7 sectioned from high-burnup ZIRLO™ rodlet with RHT stress of 140 MPa at 400°C. C_H was 720 ± 310 wppm for an adjacent ring. RHCF = 79%.

Fig. A.3 Area 2 image for ring 648G7 sectioned from high-burnup ZIRLO™ rodlet with RHT stress of 140 MPa at 400°C. C_H was 720 ± 310 wppm for an adjacent ring. RHCF = 81%.

Fig. A.4 Area 3 image for ring 648G7 sectioned from high-burnup ZIRLOTM rodlet with RHT stress of 140 MPa at 400°C. C_H was 720 ± 310 wppm for an adjacent ring. RHCF = 52%.

Fig. A.5 Area 4 image for ring 648G7 sectioned from high-burnup ZIRLO™ rodlet with RHT stress of 140 MPa at 400°C. C_H was 720 ± 310 wppm for an adjacent ring. RHCF = 38%.

Fig. A.6 Area 5 image for ring 648G7 sectioned from high-burnup ZIRLO™ rodlet with RHT stress of 140 MPa at 400°C. C_H was 720 ± 310 wppm from an adjacent ring. RHCF = 82%.

Fig. A.7 Area 6 image for ring 648G7 sectioned from high-burnup ZIRLO™ rodlet with RHT stress of 140 MPa at 400°C. C_H was 720 ± 310 wppm for an adjacent ring. RHCF = 67%.

Fig. A.8 Area 7 image for ring 648G7 sectioned from high-burnup ZIRLO™ rodlet with RHT stress of 140 MPa at 400°C. C_H was 720 ± 310 wppm for an adjacent ring. RHCF = 86%.

Fig. A.9 Area 8 image for ring 648G7 sectioned from high-burnup ZIRLO™ rodlet with RHT stress of 140 MPa at 400°C. C_H was 720 ± 310 wppm for an adjacent ring. RHCF = 71%.

Fig. A.10 Area 1 image for ring 648G8 sectioned from high-burnup ZIRLO™ rodlet with RHT stress of 140 MPa at 400°C. C_H was 640 ± 80 wppm for an adjacent ring. RHCF = 82%.

Fig. A.11 Area 2 image for ring 648G8 sectioned from high-burnup ZIRLO™ rodlet with RHT stress of 140 MPa at 400°C. C_H was 640 ± 80 wppm for an adjacent ring. RHCF = 50%.

Fig. A.12 Area 3 image for ring 648G8 sectioned from high-burnup ZIRLO™ rodlet with RHT stress of 140 MPa at 400°C. C_H was 640 ± 80 wppm for an adjacent ring. RHCF = 80%.

Fig. A.13 Area 4 image for ring 648G8 sectioned from high-burnup ZIRLO™ rodlet with RHT stress of 140 MPa at 400°C. C_H was 640 ± 80 wppm for an adjacent ring. RHCF = 64%.

Fig. A.14 Area 5 image for ring 648G8 sectioned from high-burnup ZIRLO™ rodlet with RHT stress of 140 MPa at 400°C. C_H was 640 ± 80 wppm for an adjacent ring. RHCF = 46%.

Fig. A.15 Area 6 image for ring 648G8 sectioned from high-burnup ZIRLO™ rodlet with RHT stress of 140 MPa at 400°C. C_H was 640 ± 80 wppm for an adjacent ring. RHCF = 60%.

Fig. A.16 Area 7 image for ring 648G8 sectioned from high-burnup ZIRLO™ rodlet with RHT stress of 140 MPa at 400°C. C_H was 640 ± 80 wppm for an adjacent ring. RHCF = 36%.

Fig. A.17 Area 8 image for ring 648G8 sectioned from high-burnup ZIRLO™ rodlet with RHT stress of 140 MPa at 400°C. C_H was 640 ± 80 wppm for an adjacent ring. RHCF = 67%.

Appendix B

Post-RHT Metallographic Images for High-Burnup ZIRLO™ Rodlet 648D

Fig. B.1 Sectioning diagram for high-burnup ZIRLO™ rodlet 648D following RHT (i.e., simulated drying-storage) at 400°C peak temperature and 110-MPa peak hoop stress.

Fig. B.2 Area 1 image for ring 648D7 sectioned from high-burnup ZIRLO™ rodlet with RHT stress of 110 MPa at 400°C. C_H was 425 ± 75 wppm for an adjacent ring. RHCF = 48%.

Fig. B.3 Area 2 image for ring 648D7 sectioned from high-burnup ZIRLO™ rodlet with RHT stress of 110 MPa at 400°C. C_H was 425 ± 75 wppm for an adjacent ring. RHCF = 30%.

Fig. B.4 Area 3 image for ring 648D7 sectioned from high-burnup ZIRLO™ rodlet with RHT stress of 110 MPa at 400°C. C_H was 425 ± 75 wppm for an adjacent ring. RHCF = 23%.

Fig. B.5 Area 4 image for ring 648D7 sectioned from high-burnup ZIRLO™ rodlet with RHT stress of 110 MPa at 400°C. C_H was 425 ± 75 wppm for an adjacent ring. RHCF = 20%.

Fig. B.6 Area 5 image for ring 648D7 sectioned from high-burnup ZIRLO™ rodlet with RHT stress of 110 MPa at 400°C. C_H was 425 ± 75 wppm for an adjacent ring. RHCF = 19%.

Fig. B.7 Area 6 image for ring 648D7 sectioned from high-burnup ZIRLO™ rodlet with RHT stress of 110 MPa at 400°C. C_H was 425 ± 75 wppm for an adjacent ring. RHCF = 34%.

Fig. B.8 Area 7 image for ring 648D7 sectioned from high-burnup ZIRLO™ rodlet with RHT stress of 110 MPa at 400°C. C_H was 425 ± 75 wppm for an adjacent ring. RHCF = 18%.

Fig. B.9 Area 8 image for ring 648D7 sectioned from high-burnup ZIRLO™ rodlet with RHT stress of 110 MPa at 400°C. C_H was 425 ± 75 wppm for an adjacent ring. RHCF = 23%.

Appendix C

Post-RHT Metallographic Images for High-Burnup ZIRLO™ Rodlet 648C

Fig. C.1 Sectioning diagram for high-burnup ZIRLO™ rodlet 648C following RHT (i.e., simulated drying-storage) at 400°C peak temperature and 110-MPa peak hoop stress. Hold time at 400°C was 24 hours. A silver line was put along the axial direction prior to sectioning to define the 12 o'clock orientation.

Fig. C.2 Area 1 image for ring 648C7 sectioned from high-burnup ZIRLO™ rodlet with RHT stress of 110 MPa at 400°C. C_H was 325 ± 72 wppm for an adjacent ring. RHCF = 28%.

Fig. C.3 Area 2 image for ring 648C7 sectioned from high-burnup ZIRLO™ rodlet with RHT stress of 110 MPa at 400°C. C_H was 325 ± 72 wppm for an adjacent ring. RHCF = 31%.

Fig. C.4 Area 3 image for ring 648C7 sectioned from high-burnup ZIRLO™ rodlet with RHT stress of 110 MPa at 400°C. C_H was 325 ± 72 wppm for an adjacent ring. RHCF = 21%.

Fig. C.5 Area 4 image for ring 648C7 sectioned from high-burnup ZIRLO™ rodlet with RHT stress of 110 MPa at 400°C. C_H was 325 ± 72 wppm for an adjacent ring. RHCF = 50%.

Fig. C.6 Area 5 image for ring 648C7 sectioned from high-burnup ZIRLO™ rodlet with RHT stress of 110 MPa at 400°C. C_H was 325 ± 72 wppm for an adjacent ring. RHCF = 54%.

Fig. C.7 Area 6 image for ring 648C7 sectioned from high-burnup ZIRLO™ rodlet with RHT stress of 110 MPa at 400°C. C_H was 325 ± 72 wppm for an adjacent ring. RHCF = 29%.

Fig. C.8 Area 7 image for ring 648C7 sectioned from high-burnup ZIRLO™ rodlet with RHT stress of 110 MPa at 400°C. C_H was 325 ± 72 wppm for an adjacent ring. RHCF = 26%.

Fig. C.9 Area 8 image for ring 648C7 sectioned from high-burnup ZIRLO™ rodlet with RHT stress of 110 MPa at 400°C. C_H was 325 ± 72 wppm for an adjacent ring. RHCF = 41%.

Appendix D

Post-RHT Metallographic Images for High-Burnup Zry-4 Rodlet 605C6

Fig. D.1 Sectioning diagram for high-burnup Zry-4 rodlet 605C6 following RHT (i.e., simulated drying-storage) at 400°C peak temperature and 110-MPa peak hoop stress. Hold time at 400°C was 8 hours. The black line indicates the 12 o'clock orientation and the red line indicates the clockwise direction.

Fig. D.2 Area 1 image for ring 605C6G sectioned from high-burnup Zry-4 rodlet with RHT stress of 110 MPa at 400°C. C_H was 512 ± 86 wppm for an adjacent ring. RHCF = 21%.

Fig. D.3 Area 2 image for ring 605C6G sectioned from high-burnup Zry-4 rodlet with RHT stress of 110 MPa at 400°C. C_H was 512 ± 86 wppm for an adjacent ring. RHCF = 10%.

Fig. D.4 Area 3 image for ring 605C6G sectioned from high-burnup Zry-4 rodlet with RHT stress of 110 MPa at 400°C. C_H was 512 ± 86 wppm for an adjacent ring. RHCF = 13%.

Fig. D.5 Area 4 image for ring 605C6G sectioned from high-burnup Zry-4 rodlet with RHT stress of 110 MPa at 400°C. C_H was 512 ± 86 wppm for an adjacent ring. RHCF = 4%.

Fig. D.6 Area 5 image for ring 605C6G sectioned from high-burnup Zry-4 rodlet with RHT stress of 110 MPa at 400°C. C_H was 512 ± 86 wppm for an adjacent ring. RHCF = 7%.

Fig. D.7 Area 6 image for ring 605C6G sectioned from high-burnup Zry-4 rodlet with RHT stress of 110 MPa at 400°C. C_H was 512 ± 86 wppm for an adjacent ring. RHCF = 11%.

Fig. D.8 Area 7 image for ring 605C6G sectioned from high-burnup Zry-4 rodlet with RHT stress of 110 MPa at 400°C. C_H was 512 ± 86 wppm for an adjacent ring. RHCF = 9%.

Fig. D.9 Area 8 image for ring 605C6G sectioned from high-burnup Zry-4 rodlet with RHT stress of 110 MPa at 400°C. C_H was 512 ± 86 wppm from an adjacent ring. RHCF = 9%.

Fig. D.10 Area 9 image for ring 605C6G sectioned from high-burnup Zry-4 rodlet with RHT stress of 110 MPa at 400°C. C_H was 512 ± 86 wppm for an adjacent ring. RHCF = 6%.

Fig. D.11 Area 10 image for ring 605C6G sectioned from high-burnup Zry-4 rodlet with RHT stress of 110 MPa at 400°C. C_H was 512 ± 86 wppm for an adjacent ring. RHCF = 9%.

Fig. D.12 Area 11 image for ring 605C6G sectioned from high-burnup Zry-4 rodlet with RHT stress of 110 MPa at 400°C. C_H was 512 ± 86 wppm for an adjacent ring. RHCF = 3%.

Appendix E

Post-RHT Metallographic Images for High-Burnup Zry-4 Rodlet 605C2

Fig. E.1 Sectioning diagram for high-burnup Zry-4 rodlet 605C2 following RHT (i.e., simulated drying-storage) at 400°C peak temperature and 140-MPa peak hoop stress. Hold time at 400°C was 8 hours. The black line indicates the 12 o'clock orientation and the red line indicates the clockwise direction.

Fig. E.2 Area 1 (12 o'clock orientation) image for ring 605C2E sectioned from high-burnup Zry-4 rodlet with RHT stress of 140 MPa at 400°C. C_H was 586 ± 78 wppm for an adjacent ring. RHCF = 11%.

Fig. E.3 Area 2 image for ring 605C2E sectioned from high-burnup Zry-4 rodlet with RHT stress of 140 MPa at 400°C. C_H was 586 ± 78 wppm for an adjacent ring. RHCF = 17%.

Fig. E.4 Area 3 image for ring 605C2E sectioned from high-burnup Zry-4 rodlet with RHT stress of 140 MPa at 400°C. C_H was 586 ± 78 wppm for an adjacent ring. RHCF = 24%.

Fig. E.5 Area 4 image for ring 605C2E sectioned from high-burnup Zry-4 rodlet with RHT stress of 140 MPa at 400°C. C_H was 586 ± 78 wppm. RHCF = 23%.

Fig. E.6 Area 5 image for ring 605C2E sectioned from high-burnup Zry-4 rodlet with RHT stress of 140 MPa at 400°C. C_H was 586 ± 78 wppm for an adjacent ring. RHCF = 16%.

Fig. E.7 Area 6 image for ring 605C2E sectioned from high-burnup Zry-4 rodlet with RHT stress of 140 MPa at 400°C. C_H was 586 ± 78 wppm for an adjacent ring. RHCF = 16%.

Fig. E.8 Area 7 image for ring 605C2E sectioned from high-burnup Zry-4 rodlet with RHT stress of 140 MPa at 400°C. C_H was 586 ± 78 wppm for an adjacent ring. RHCF = 13%.

Fig. E.9 Area 8 image for ring 605C2E sectioned from high-burnup Zry-4 rodlet with RHT stress of 140 MPa at 400°C. C_H was 586 ± 78 wppm for an adjacent ring. RHCF = 12%.

Fig. E.10 Area 9 image for ring 605C2E sectioned from high-burnup Zry-4 rodlet with RHT stress of 140 MPa at 400°C. C_H was 586 ± 78 wppm for an adjacent ring. RHCF = 17%.

Fig. E.11 Area 10 image for ring 605C2E sectioned from high-burnup Zry-4 rodlet with RHT stress of 140 MPa at 400°C. C_H was 586 ± 78 wppm for an adjacent ring. RHCF = 11%.

Fig. E.12 Area 11 image for ring 605C2E sectioned from high-burnup Zry-4 rodlet with RHT stress of 140 MPa at 400°C. C_H was 586 ± 78 wppm for an adjacent ring. RHCF = 14%.

Fig. E.13 Area 12 image for ring 605C2E sectioned from high-burnup Zry-4 rodlet with RHT stress of 140 MPa at 400°C. C_H was 586 ± 78 wppm for an adjacent ring. RHCF = 14%.

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