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# Burst and Tensile Testing of Hydrided Fuel Cladding

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## Abstract

The U.S. Department of Energy Office of Nuclear Energy, Office of Fuel Cycle Technology has established the Used Fuel Disposition Campaign (UFDC) to conduct the research and development activities related to storage, transportation, and disposal of used nuclear fuel (UNF) and high-level radioactive waste. Within the UFDC, the storage and transportation task has been created to address issues of extended storage and transportation. One gap in knowledge identified as high priority by UFDC includes hydrogen effects in zirconium alloy cladding and reorientation of clad hydrides. During reactor operations, hydrogen, generated from the oxidation of the surface of zirconium alloy materials in the coolant of light water reactors, can be absorbed into the cladding and form zirconium hydrides. At high temperatures and hoop stresses, the normally circumferential-oriented hydrides in High Burnup UNF (HB-UNF) may reorient in the radial direction and affect the mechanical properties of the clad. Understanding these changes in mechanical properties will aid in analyzing how UNF will perform during storage and transportation. Separate effects testing to measure the mechanical properties of un-hydrided, pre-hydrided, and irradiated HB-UNF is currently underway at Pacific Northwest National Laboratory (PNNL). In order to measure these mechanical properties, staff at PNNL are employing tensile, compression, and burst testing. Each of these test stands are capable of testing the clad at temperatures up to 400ºC. The development of these test stands and future testing of pre-hydrided and HB-UNF cladding are described herein.

## Introduction

The U.S. Department of Energy Office of Nuclear Energy, Office of Fuel Cycle Technology established the UFDC to conduct research and development activities related to storage, transportation, and disposal of UNF and high-level radioactive waste. Within the UFDC, the storage and transportation task was created to address issues of extended storage and transportation. This task's near-term objectives are to develop the technical basis which supports the continued safe and secure storage, retrieval, and transportation of UNF. In 2012, the UFDC identified specific research and development gaps that needed to be filled to develop this basis [1]. One high-priority gap identified concerned hydrogen effects in HB-UNF, specifically the reorientation of cladding hydrides, which may negatively affect cladding performance [2].

Multiple laboratories within the UFDC are performing separate effects tests to determine the role that various parameters (e.g., hydride content and distribution, radiation damage, and oxide thickness) play in determining the mechanical properties of irradiated cladding and how they change over time during extended storage. Separate effects tests at PNNL were initiated to develop the relationship between the properties of as-manufactured cladding and cladding from irradiated used fuel. PNNL procured as-manufactured cladding and began characterizing its mechanical properties prior to the addition of hydrogen and irradiation. At the same time, Hanson et al. [3] and Shimskey et al. [4,5] developed a means for pre-hydriding cladding such that the concentration, distribution, and morphology are the same as in high burnup cladding at temperatures below the short-term annealing point for radiation damage in cladding

(<350°C). This paper outlines the progress PNNL has made in developing systems which are capable of pre-hydriding cladding specimens, mechanical testing of as manufactured and hydrided cladding, the current development of systems that can reorient circumferential hydrides to a radial orientation, and future testing of irradiated cladding.

## BACKGROUND

During light water reactor operations, zirconium cladding is in direct contact with reactor coolant. The surface of the cladding oxidizes in this environment and produces hydrogen as a by-product. Some fraction of this hydrogen is subsequently absorbed into the cladding. This fraction is commonly referred to as the hydrogen uptake ratio. Once the hydrogen solubility is exceeded in the metal matrix, zirconium hydrides begin to form. Because the hydrogen is taken up on the waterside of the cladding and because the temperature, and thus hydrogen solubility is much lower in this region, a localized high concentration of hydrogen on the surface results in the formation of a hydride rim with circumferential oriented hydrides as shown in Figure 1.

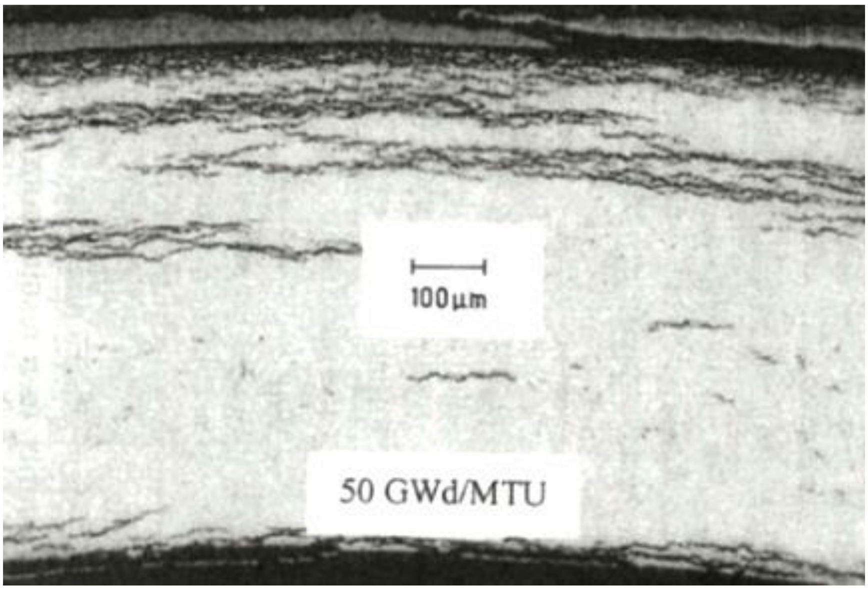

**Figure 1: Cladding sample with hydride rim**

After reactor operations, UNF is stored for some time period in spent fuel pools, and then moved to dry storage. As a part of the dry storage process, UNF is placed into canisters, which are evacuated and back-filled with helium. During this drying process, the decay heat of the fuel causes the rod internal pressure (from initial helium backfill and fission gas released from the fuel during irradiation) to increase, this in turn increasing the cladding hoop stress. The combination of high temperatures and high hoop stresses has the potential to allow for hydrides to dissolve and reprecipitate in the radial direction. If sufficient radial hydrides form, they can severely weaken the structure of the cladding and impact the mechanical properties of the cladding.

## HYDRIDING EQUIPMENT

The hydriding systems consist of a tube furnace capable of pre-hydriding cladding specimens and reorienting cladding hydrides and surface conditioning equipment.

### Hydrogen Tube Furnace

Unirradiated cladding specimens are pre-hydrided using a 24 inch tube furnace designed specifically for heat treating materials in hydrogen gas (Figure 2). The furnace has three heat zones that heat the interior stainless tube where specimens are heat treated. The furnace can be connected to the building argon supply or a high purity hydrogen gas bottle to heat treat materials in either gas. Each zone of the furnace is controlled by an exterior thermocouple that comes into contact with the tube furnace. In addition, a profile thermocouple is placed in the center of the tube furnace during operation to ensure that the internal temperature was adequately controlled. During operation, up to four 18 inch cladding specimens can be attached to the profile thermocouple and simultaneously pre-hydrided. Figure 3 provides a system diagram of the tube furnace being used for pre-hydriding.

**Figure 2: Hydrogen tube furnace**

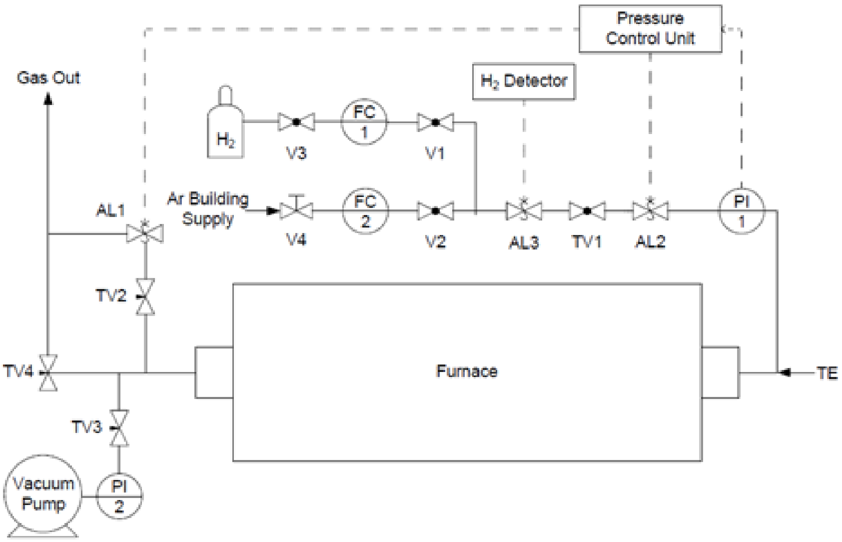

**Figure 3: Pre-hydriding system diagram**

The tube furnace can also be configured to reorient hydrides. This is done by including a manifold system capable of simultaneously internally pressurizing four clad specimens up to 3750

PSI with argon. During this mode of operation, the tube furnace is filled with argon, the cladding specimens are pressurized and heated to temperatures up to 400°C. Specimen internal pressure is maintained at a constant pressure during heating and cooling with a dome style back pressure regulator. It is also possible to pressurize the specimens and allow the pressure to vary as the sample heats up and cools down. Throughout this process, system pressure and tube furnace temperature are digitally recorded. The tube furnace manifold is shown in Figure 4, and the system diagram is shown in Figure 5.

**Figure 4:** Tube furnace manifold system

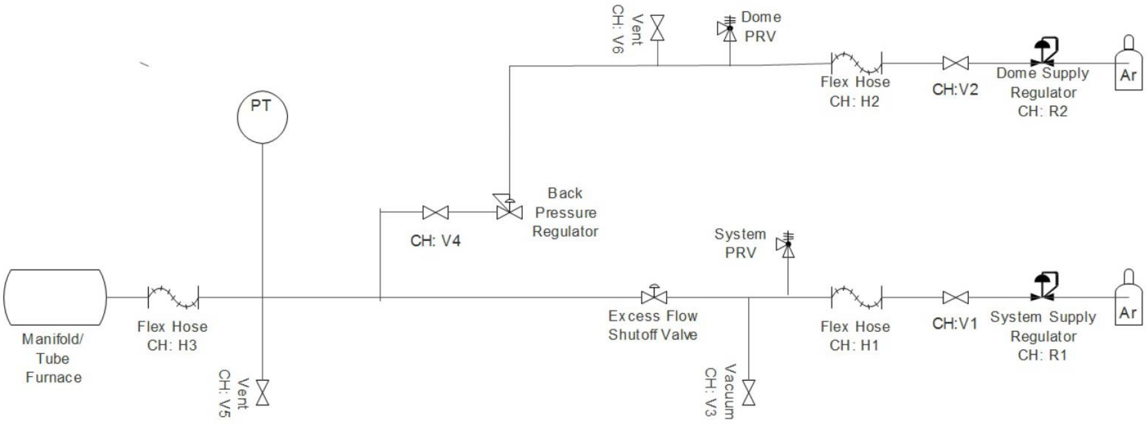

**Figure 5: Hydride reorientation system diagram**

Currently, the pre-hydride and hydride reorientation systems at PNNL are fully operational. Figure 6 shows optical metallography images of a pre-hydrided specimen. Results from the hydride reorientation system are shown in a subsequent section.

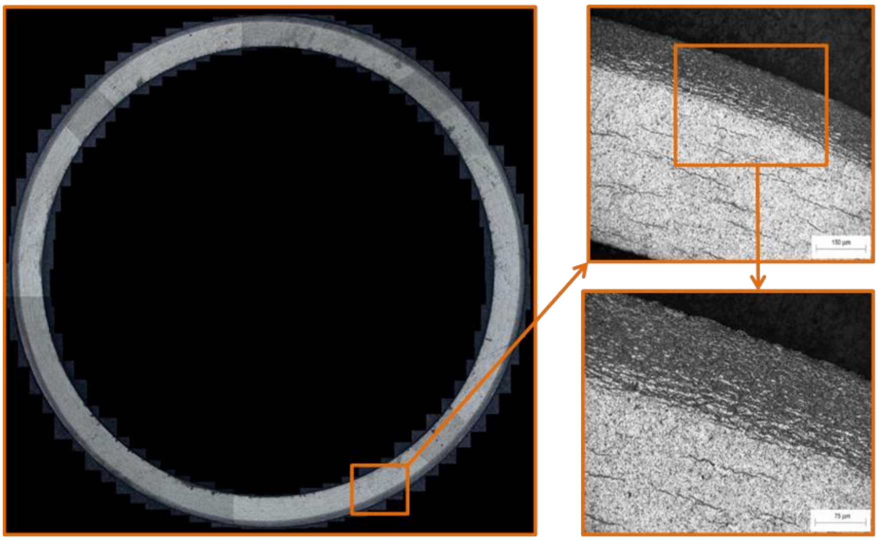

**Figure 6: Optical metallography images of a pre-hydrided specimen**

Surface Conditioning

The surface condition effects hydrogen diffusion into the cladding. Etching the inner surface limits the diffusion of hydrogen into the cladding inner diameter, and frit blasting the outer surface increases the diffusion of hydrogen into the cladding outer diameter.

To chemically etch the inner surface of the cladding specimens prior to pre-hydriding, a system that pumps a solution of 4% HF / 30% $HNO_3$ has been designed and fabricated. After the inner surface of the specimen has been sufficiently etched, the system is reconfigured to direct water through the inner diameter (ID). After a final rinse, the specimens are soaked in deionized water to achieve a uniform oxide coating. The etching system is shown in Figure 7 and a system diagram of the etching system is shown in Figure 8.

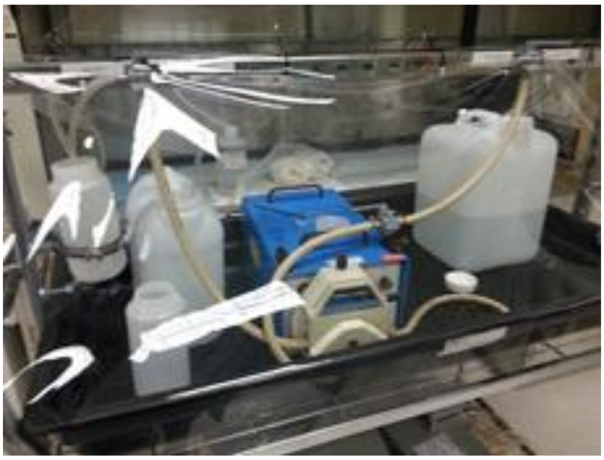

**Figure 7: Cladding etching system**

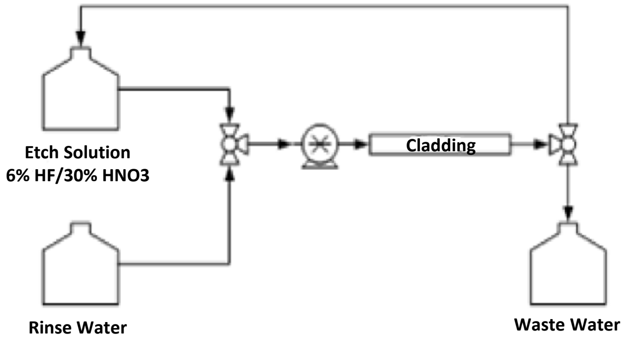

**Figure 8: Cladding etching system diagram**

After the specimens are dried, the outer diameter (OD) of each specimen is blasted using glass frit to strip the surface oxide layer. A system has been designed to perform this frit blasting process in a highly repeatable manner, in order to ensure that all of the hydrided specimens are prepared in a consistent manner. The blasting system rotates the specimen at high speed while simultaneously blasting it along its length using a linear actuator.  Both rotation and linear travel are controlled with variable speed drives. The frit blasting system is shown in Figure 9.

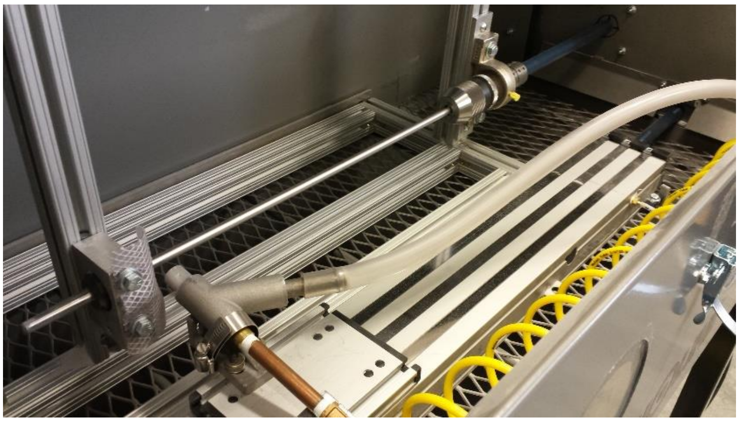

**Figure 9: Frit blasting system**

## MECHANICAL TEST EQUIPMENT

Mechanical testing (i.e., tensile, compression, and burst testing) will be performed on both pre-hydrided cladding and cladding with reoriented hydrides, and the results will be compared with those of as-manufactured cladding. Tensile and compression testing will be performed using a 5 kilo-pound load frame (see Figure 10); burst testing will be performed on a purpose-built test system (see Figure 11).

Strain is measured with a digital image correlation (DIC) system. DIC is a real time optical measurement technique that utilizes high-definition imagery to measure object deformation. This is done by first applying an optical pattern (i.e., speckle pattern) to an object’s surface and then using DIC software to quantify the displacement of the pattern. This method allows the strain tensor to be determined at any point on the tube’s surface. Figure 12 shows the DIC system during tensile testing.

Figure 10: Load frame

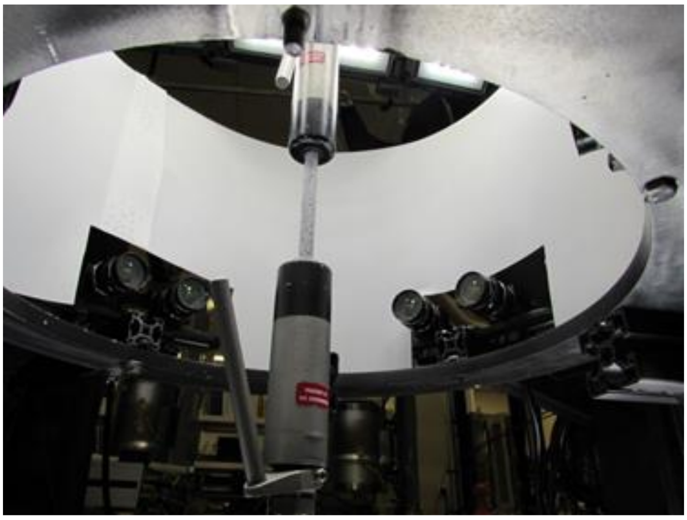

Figure 11: Burst system

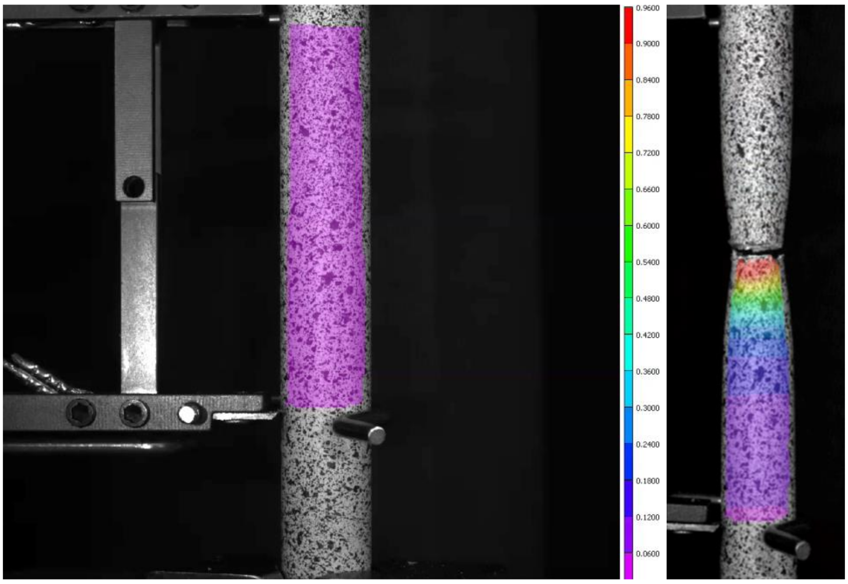

Figure 12: Tensile testing DIC results

## MECHANICAL TESTING RESULTS: POST PROCESSING

Results from the destructive testing are analyzed using statistical methods to evaluate the probabilistic nature of the mechanical properties of the cladding and to understand if variability increases or decreases from the hydrogen heat treatments being performed. Baseline testing of the as-manufactured material was performed to evaluate the initial variability of the test material from the effects of flaws that are introduced during the manufacturing process. One of the more common statistical distributions used in manufacturing is the Weibull distribution, commonly used in the ceramic industry for evaluation of flaws. The presence of zirconium hydride in Zircaloy behaves like flaws in the matrix causing reduction in strength where the flaws are present. For this reason, this type of analysis was selected to understand how variability in the mechanical properties changes when the hydrides are present, and how that differs with unirradiated and irradiated material. The results are normalized and plotted against each other on a Weibull plot. Figure 13 shows an example Weibull plot.

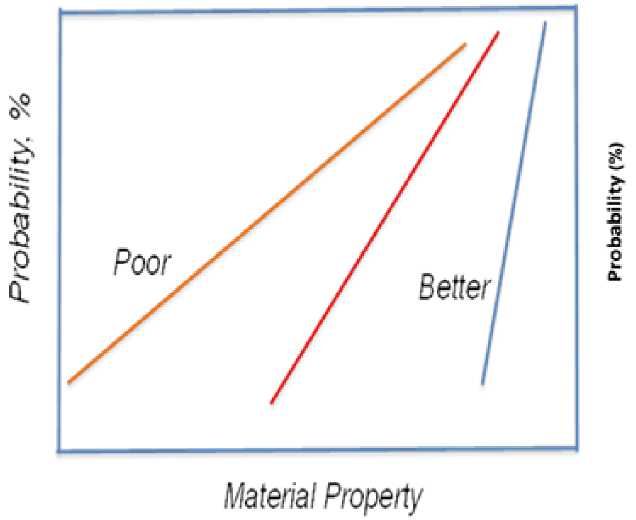

Figure 13: Example Weibull plot

The utility of the Weibull plot is that the distribution is plotted linearly. The slope from the plot is called the Weibull Modulus. As the Weibull Modulus increases, the material is considered to be more reliable and will behave in a consistent manner. As the Weibull Modulus decreases, material performance becomes less predictable which is likely the result of flaws being present. In order to ensure a sound statistical data set it is important that testing be conducted in a manner which does not exclude material with as manufactured flaws and include a statistical sufficient number of tests with each specimen type to ensure confidence in the result.

## HYDRIDE REORIENTATION RESULTS

PNNL has begun processing samples with the hydride reorientation system described herein. Runs have ranged from 325°C to 400°C, and 70 MPa hoop stress to 150 MPa hoop stress. To illustrate the transition from cases with no radial hydrides to cases with radial hydrides; metallography results from the 325°C/70MPa, 325°C/110MPa, and 325°C/150MPa cases are shown respectively in Figures 14-16. There are no radial hydrides present in Figures 14 and 15, and Figure 16 clearly shows the presence of radial hydrides.

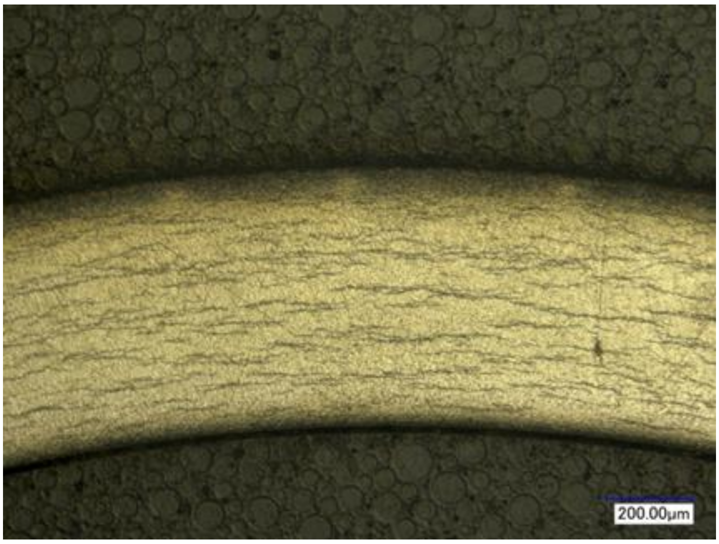

**Figure 14: 325°C/70Mpa**

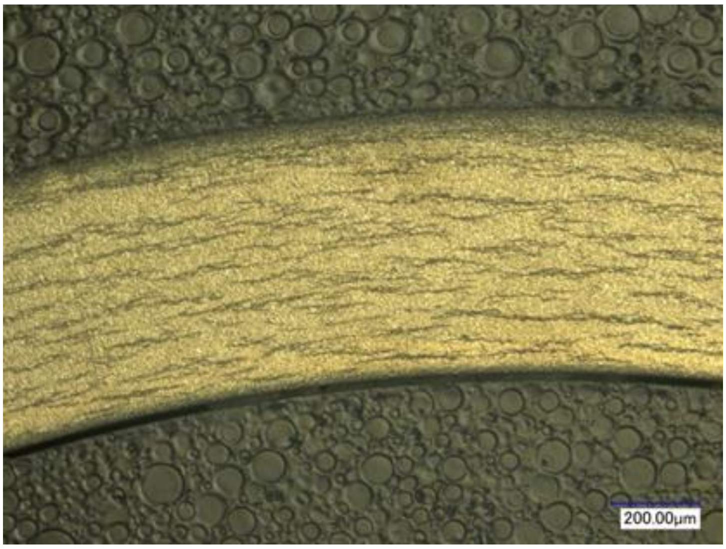

Figure 15: 325C/110Mpa

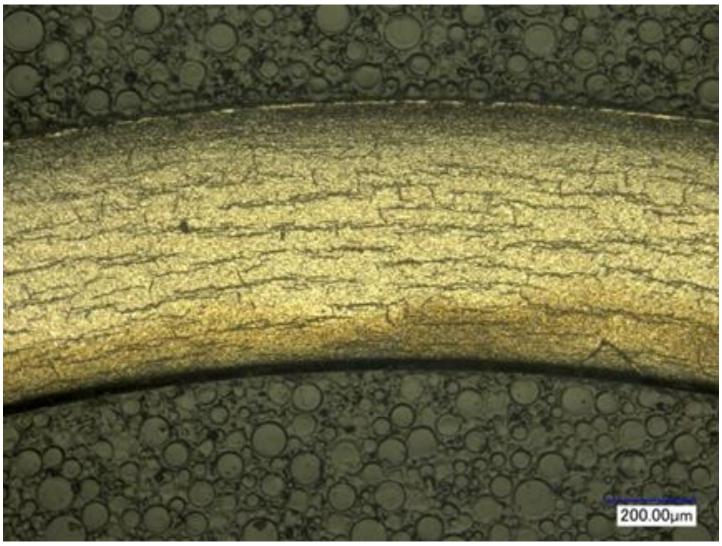

Figure 16: 325°C/150MPa

## CONCLUSION AND FUTURE WORK

The systems which are capable of introducing hydrides into cladding specimens, reorienting hydrides, and test frames capable of measuring the mechanical properties of these specimens have been presented. Baseline testing of as-manufactured cladding specimens and pre-hydrided cladding specimens has been conducted. It is planned that tensile, compression, and burst testing of cladding with reoriented hydrides will begin this year. This work will be used to guide future work with irradiated specimens.

## ACKNOWLEDGMENTS

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