Neutron Diffraction Measurement of Residual Stresses in “OSU Plate.”

D.W. Brown1, M. A. Okuniewski2, T. A. Sisneros1, B. Clausen1

1Los Alamos National Laboratory, Los Alamos, New Mexico, 87545

2Idaho National Laboratory, Idaho Falls, Idaho 83415

Introduction

Aluminum-clad U10Mo (including zirconium diffusion layer) fuel assembly plates are bound to be in a state of residual stress due to the mismatch of thermal expansion of the constituent phases. Due to the geometry of the plates, i.e. thin beams, it is expected that the stress will predominantly be in-plane. The thermal expansion of the aluminum cladding, zirconium diffusion layer and U10Mo foil are roughly $\alpha_{Al}=22\times10^{-6}/K$, $\alpha_{Zr}=5\times10^{-6}/K$, and $\alpha_{U10Mo}=10\times10^{-6}/K$. Note that the coefficient of thermal expansion of the zirconium is quoted as that in the a direction in the hexagonal close packed crystal structure because of the strong texture in the rolled zirconium foil, in which the a-axis are strongly preferred in the in-plane direction. During cooling from the hot isostatic pressing (HIP’ing) temperature, the zirconium contracts considerably less than the U10Mo, which in turn, contracts less than the aluminum. Because the aluminum dominates in terms of volume fraction, it is expected that the U10Mo foil and zirconium bonding layer will be in a residual stress state of in-plane compression at room temperature after bonding. This has been verified for the U10Mo foil in a mini fuel plate using x-ray diffraction at the Advanced Photon Source, but measurements of the stress state of the aluminum and zirconium were not possible at the APS. Also, the APS measurements were limited to mini size fuel plates.

Neutron diffraction experiments of residual stress on the “OSU plate” were completed on the Spectrometer for Materials Research at Temperature and Stress (SMARTS) at the Lujan Center at LANSCE between 2/19-21/2013. Longitudinal and transverse residual strain profiles were measured on an Al-clad U10-Mo OSU plate. Residual strains from the aluminum cladding, the zirconium bonding layer and the U10Mo fuel foil were recorded.

Experimental Details

Sample Preparation

Depleted uranium and molybdenum feedstock were arc-melted in an inert atmosphere and cast into a 2.5 mm thick coupon and hot rolled at 650°C. A zirconium diffusion barrier (~0.025mm thick) was co-rolled onto the fuel foil during the hot rolling process. The foil was annealed at 650°C for 45 minutes followed by cold rolling to the final thickness of 0.28mm. The finished foils were hand polished with 15μm diamond paste and cleaned with ethanol before being bonded to the aluminum cladding (Al 6061 in the T6 condition). A recess which loosely fit the U-10Mo foil was machined into one side of the aluminum cladding, thus the cladding is thinner
on one side of the foil. The plates were sealed in stainless steel can and heated to the HIP’ing temperature of 560°C at 4.8°C/min after which 104MPa of pressure was applied during the 90 minute soak time. Subsequently, the HIP assembly was cooled at ~6.7°C/min to room temperature.

The dimensions of the complete fuel assembly were 660.5mm longitudinal, 127.4mm transverse, and 2.03mm thick. Those of the U-10Mo foil were 580mm longitudinal, 88.9mm transverse, and nominally 0.25mm thick. The Zr diffusion layer was 0.015mm thick.

Residual strain measurement

The residual strain measurements were completed on the SMARTS diffractometer at LANSCE. SMARTS accepts a white neutron beam (continuous wavelength spectrum from 0.5Å - 4.0Å) from a 283K water moderator located roughly 31m from the sample position. A schematic of the diffraction geometry and picture of the OSU fuel plate sample in position are shown in figure 1. Two thin foils of fully annealed copper were attached to the sample symmetrically on the front and back face (see figure 1) to be used for “on-board” calibration of the instrument.

The incident neutron beam was defined using computer controlled motorized boron-nitride slits (set to 2x5mm) and impinged on the sample mounted with is longitudinal (2) direction oriented 45° from the incident beam. The neutrons are scattered from the illuminated portion (called the “gauge volume”) of the sample and collected in two fixed angle detector banks situated at ±90° from the incident beam each defining a unique diffraction vector (Q); one parallel to the 2-direction of the sample (Bank 2), the other parallel to the normal (3) direction (Bank 1).

The sample was mounted on a computer controlled translator table with ±30cm and ±60cm of travel in both horizontal and vertical directions, respectively, and ±0.01mm positioning repeatability. The sample was located and oriented with respect to the known center of the diffractometer to better than ±0.1mm accuracy using computerized theodolites. The sample was moved with respect to the “gauge” volume and diffraction patterns collected at selected individual positions (indicated in figure 2). Following the completion of the measurements in the 2 and 3 directions, the sample was re-mounted in a vertical orientation such that the diffraction vector associated with Bank 2 was parallel to the sample transverse (1) direction. In this orientation, diffraction patterns in Bank 1 give a repeat measurement in the 3 direction.

Figure 2 shows a schematic of the sample with an overlaid plot of the measurement positions on the foil. The aspect ratio of the schematic is correct. The origin of the axis used throughout this report is indicated. The transverse (1) and longitudinal (2) directions of the sample are labeled in the figure and the sample normal direction (3) is out of the page. Diffraction patterns were collected at many points (see figure 2) on the plate including areas on the U10Mo meat (blue), areas solely on the aluminum (black), and areas on the aluminum as well as copper foil calibrant (red). In this coordinate system, the relevant edges of the U10Mo foil are at y = 19.1mm and x = 40.3mm. We will assume the sample is symmetric about the center. Then the results observed in
the aluminum and copper at \( y = 118.3 \text{mm} \) (which are 9.1mm from the edge of the assembly), will be placed at \( y = 9.1 \text{ mm} \). Several of the data points collected near the corner of the foil (40.3, 19.1) with the sample horizontal had to be partially discarded because neutrons scattered from the aluminum clamp on the fixture interfered with the aluminum diffraction patterns. The U10Mo diffraction patterns at these points were usable.

Diffraction patterns were recorded for 30-120 minutes depending on incident beam size and purpose of the measurement. In order to profile the residual strains, the incident beam was collimated to 2(H) x 5(V) mm, optimizing the spatial resolution in the scanning direction. In this condition 30 minutes of data collection was required to get sufficient statistics to determine the lattice parameters of the aluminum and U10Mo with sufficient accurately to determine residual strains. However, because the relatively small amount of zirconium resulted in large uncertainties in the determination of the residual strains, the slits were opened to 5x5mm and count time increased to 1 hour to optimize for accuracy in determination of the zirconium lattice parameter at a few select points.

The purpose of the on-board calibrant is to detect and correct fictitious strains which would arise if the sample were not aligned perfectly with the motion of the translator table, causing the sample to deviate from the center of the diffractometer. This also allows for cross calibration of the two detector banks, that is the two detectors are calibrated to each other, but not necessarily “absolutely” calibrated.

Data analysis The diffraction data were analyzed using Rietveld analysis of the full pattern using the General Structural Analysis Software (GSAS) developed at Los Alamos to determine the lattice parameter at each measurement position. The elastic lattice strain at position \((x,y)\) is determined from the lattice parameter by \( \varepsilon(x, y) = \frac{a(x, y) - a_0}{a_0} \), where \( a_0 \) represents a reference, stress-free, lattice parameter. No good reference sample available for either the aluminum, the U10Mo or the zirconium. In the case of the aluminum, \( a_0 \) was taken to be the observed value far from the U10Mo foil (near a corner), where stresses should be minimal. The reference lattice parameter of the U10Mo was determined by forcing the normal component of the stress \( (\sigma_{33}) \) to zero at the center of the plate, far from any boundary. The zirconium will be discussed more thoroughly when the data is presented.

These are not ideal assumptions, but it should be recognized that they only affect the zero of the strain (and stress), and affect neither the trends as a function of position, nor the direction dependence (1, 2, or 3 direction) as these have been independently calibrated by the attached copper foil. The following plots of strain and stress are based on these assumptions.

Results
Calibration Figure 3 shows the copper lattice parameter as a function of position resulting from the scan along the longitudinal direction of the copper foil attached to the OSU plate to be used as a calibration of the detector. The copper calibrant has 2 purposes. The first is to detect errors due to the sample being imperfectly aligned with respect to the translation axis of the sample. The positive slope indicates that the sample is moving with respect to the gauge volume as it is translated from one end to the other. The slope corresponds to roughly 250 microstrain (strain x 10⁶ or \( \mu \varepsilon \)), which in turn corresponds to roughly 0.5mm offset over the 300mm translation of the sample. The second purpose is to calibrate the two detector banks against each other. The offset between the copper lattice parameter in the normal (bank 1) and transverse (bank 2) directions indicate that the center of diffraction of the sample is slightly off the defined center of diffraction of the instrument. This offset again is roughly 250\( \mu \varepsilon \), or roughly 0.5mm. The copper calibrant does not allow for absolute calibration of the diffractometer because lattice parameters are very sensitive to chemical impurities. Rather, absolute calibration will be completed for each phase independently by making assumptions about “stress free” conditions of the sample.

These corrections are usually not necessary because focusing diffracted beam optics are typically used to define the volume that the detectors “see”. That is not possible with these particular samples because they are so thin. Both of these errors were anticipated and the copper data was collected specifically to enable corrections of same. The actual corrections are relatively small compared to the mechanical strains which will be discussed in the followin.

Lattice parameters Figures 4a and b show the lattice parameters determined for (a.) aluminum and (b.) U10Mo along a line at the transverse center of the fuel assembly (at y = 63.7mm). The U10Mo foil starts at x = 40.3mm, indicated by the green line. The aluminum lattice parameters are relatively direction independent at x less than 40.3mm from the edge of the assembly, that is off of the U10Mo foil. Immediately upon reaching the edge of the U10Mo foil, a clear separation between the aluminum lattice parameter in the normal direction and the longitudinal/transverse directions is apparent. The lattice parameters in the longitudinal and transverse directions are significantly larger than in the normal direction. The separation is roughly 1000\( \mu \varepsilon \), or 4 times larger than the corrections made to the raw data. The two independent measurements of the aluminum lattice parameter in the normal direction (made with the sample vertical and horizontal) are in good agreement with each other.

The U10Mo lattice parameter likewise is very different in the normal direction and close in the longitudinal and transverse directions. In this case, the difference between normal and the in-plane directions is roughly 3000\( \mu \varepsilon \), or an order of magnitude larger than the necessary corrections. In contrast to the aluminum, in the U10Mo, the lattice parameter in the normal direction is larger than the in-plane directions.

Residual Strains
Figures 5-11 show the residual strain profiles in the aluminum on various longitudinal loci, starting from the transverse center and moving towards the edge of the assembly. Each figure includes a schematic which indicates the location of the profile relative to the fuel assembly. The long edge of the U10Mo foil is at y = 19.1 mm. Over the U10Mo meat, a tensile strain is observed in the aluminum cladding in both in-plane (longitudinal and transverse) directions, compressive in the normal direction. Moving in the x-direction (longitudinally) from the center across the edge of the U10Mo foil and subsequently toward the edge of the assembly, the longitudinal strain drops very quickly and actually becomes compressive. The transverse strain drops to zero more slowly. Moving in the y direction, the transverse strain rapidly changes from tensile to compressive at the edge of the U10Mo foil, at x=19.1mm. The longitudinal strain remains constant within uncertainty, decreasing only a small amount when near the edge of the assembly, i.e. at y = 9.1mm.

Figures 12-15 show the residual strain profiles in the U10Mo foil. Everywhere in the foil, the in-plane strain components are compressive while the normal strain component is tensile. The transverse strain slowly decreases near the long edge of the foil.

The residual strain in the zirconium is not plotted because no variation in the zirconium lattice parameter was observed as a function of position and because we have no reliable reference, i.e. we would have to rely on an absolute calibration of the diffractometer and compare to a literature value for the lattice parameter. However, it is reasonable to consider the c/a ratio of the zirconium because absolute calibration uncertainty in the diffractometer (both in SMARTS and the reference value from literature) will cancel in the c/a ratio.

The literature value of c/a is 1.59270 for pure Zr. Near the center of the foil, at (300mm, 63.7mm), in the dominant crystal orientation, i.e. with the c-axis in the sample normal (3) direction and the a-axis in the sample longitudinal (2) direction, we measure a $c_3/a_2$ of 1.5972 ± 0.0002, significantly larger (0.29 ± 0.01%) than the literature value. Moreover, in the weak crystal orientation, i.e. with the a-axis in the sample normal (3) direction and the c-axis in the sample longitudinal (2) direction, we measure a $c_2/a_3$ of 1.589 ± 0.0011, which has a much larger uncertainty, but is still significantly different (-0.25 ± 0.06%), lower in this case, than the literature value.

A chemistry difference between the Zr layer (i.e. dissolved Al, U, or Mo) could easily cause a change in c/a ratio from the accepted literature value, but it could not be direction dependent (that is it would be a scalar similar to a temperature difference). Relative calibration error of the two detectors could also cause an erroneous disparity of c/a in the two directions described; the lattice parameter is always larger in the sample normal direction. However, the measured shift is too large (by 10x) to be due to instrument calibration and the relative values of the aluminum and U10Mo lattice parameters in the opposite detector banks agree with expectations.
The observed c/a ratios are consistent with a (large) longitudinal compressive stress in the zirconium bonding layer resulting in compressive longitudinal strains and tensile normal (Poisson’s) strain. While the current data is not enough to determine actual stresses, they can and will be compared to FE models for the purpose of cross validation.

**Residual Stresses**

The residual strains are determined in three orthogonal directions. The residual stresses in the same directions are then determined from Hooke’s law:

\[
\sigma_{ii} = \frac{E}{1+\nu} \left\{ \varepsilon_{ii} + \frac{\nu}{1-2\nu} \left( \varepsilon_{11} + \varepsilon_{22} + \varepsilon_{33} \right) \right\},
\]

where we have used \(E=70\)GPa and \(\nu=0.33\) for the aluminum and \(E=90\)GPa and \(\nu=0.35\) for U10Mo.

Figure 16 and 17 show residual stress profiles for both the aluminum cladding and U10Mo foil over the area mapped. The scale for the aluminum profiles goes from -50MPa to -100MPa in 10MPa increments, while that of the U10Mo profile ranges from -325MPa to 100MPa in 25Mpa intervals. The * on the figure mark the actual diffraction positions. The white * indicate positions where the aluminum diffraction pattern was affected by neutrons scattered from the aluminum clamps and the data has been interpolated on these points in order to have a uniform grid for the contour plot.

In both cases the normal stress components are small compared to the in-plane stress. In the aluminum, the normal stress is generally tensile over the meat and compressive elsewhere, but is within 20MPa of zero. The in-plane stress components are more significant. The longitudinal stress in the aluminum is roughly 60MPa everywhere over the fuel foil. It drops very quickly to zero and slightly negative when crossing the short edge, i.e. when the stress component is normal to the edge. In contrast, the longitudinal stress component drops more slowly when crossing the long edge, that is when the stress component is parallel to the boundary. The transverse stress is similar, reaching 80MPa near the center of the foil. Similar to the behavior of the longitudinal stress, when crossing the edge that is normal to the stress component, in this case the long edge, the transverse stress decreases rapidly to zero or even slightly compressive. In contrast to the longitudinal stress, the transverse stress also drops relatively quickly when crossing the short edge, when the stress component is parallel to the boundary. The difference in the behavior is likely due to the fact that the edge effects will be more dramatic on the short side, where the edge is much closer.

The normal stress in the U10Mo hovers near zero, with possibly one measurement point reaching 50MPa. The in-plane stress in the U10Mo are significant and compressive, roughly -200MPa near the center of the foil and reaching an absolute maximum at roughly -275MPa closer to the corner.

**Summary**
Residual stresses were measured as a function of position over a relatively course grid covering a single quadrant on a large (660 mm by 127 mm) aluminum clad U10Mo monolithic fuel plate assembly. On-board calibration via an annealed copper foil allowed for measurements in an atypical configuration due to the very thin (for neutron measurements) sample. The stresses were largest in the in-plane directions in the area where the cladding was directly over the U10Mo foil, reaching 70MPa in tension in the aluminum and -250MPa in compression in the U10Mo foil. Unsurprisingly, the normal components of the stress were considerably lower; the stress field is roughly equi-biaxial. Evidence of a large in-plane compression stress in the zirconium bonding layer is observed.

The experiments demonstrate that neutron diffraction can be used to give rapid turnaround (when the accelerator is available) measurements of residual stresses in Al-clad U10Mo foils, although they cannot hope to match the spatial resolution or mapping capabilities of x-ray measurements done at the APS. However, the neutron measurements do not suffer the limitations in terms of large samples size or activated samples that are inherent in APS (or x-ray in general) measurements. Thus, for full size fuel plates and/or irradiated fuel plates, neutron diffraction measurements provide a reasonable alternative to characterize the residual stress profile with a short turn-around time. Also, neutron diffraction provides the opportunity to sample the aluminum cladding and zirconium bonding layer, which is not available to high-energy x-rays because of statistical sampling issues. The x-ray and neutron measurements should be considered as complementary. We believe this demonstrates the veracity of the technique to the point that planning for measurements on irradiated fuel plates can be initiated.

Some potential improvements to future measurements were also learned. 1.) The aluminum clamps need to be placed more carefully or masked to prevent parasitic scattering which interfered with the aluminum diffraction data of interest. 2.) If the sample were rotated about a vertical axis as well as rotated from the horizontal to the longitudinal position, then the 2 repeat measurements of the lattice parameters in the normal direction, which would not be in the 2 different banks, could be used as a calibration between the two banks. This could potentially be a primary calibration, or just a secondary check on the copper calibration.

Acknowledgements We wish to thank Dave Alexander and Kester Clarke for preparation of the sample. The Lujan Center at the Los Alamos Neutron Science Center at LANSCE is funded by the U.S. DOE, Office of Basic Energy Sciences. Los Alamos National Laboratory is operated by Los Alamos National Security LLC under DOE contract DE-AC52-06NA25396.
Figure 1. Schematic of diffraction instrument and picture of same.
Figure 2. Schematic of the sample with an overlaid plot of the measurement positions on the foil. The origin of the axis used throughout this report is indicated. The longitudinal (2) and transverse (1) directions are indicated. The normal (3) direction is out of the page.
Figure 3. Lattice parameters determined from annealed copper foil used to calibrate the two detector banks and correct for mis-alignment.
Figure 4. Lattice parameters determined for a.) aluminum and b.) U10Mo along the central line of the fuel assembly.
Figure 5. Normal, longitudinal, and transverse residual strains in the aluminum cladding as a function of x along the center line of the sample (y = 63.8 mm). The schematic on the right indicates the loci of measurement points shown in the plot.
Figure 6. Normal, longitudinal, and transverse residual strains in the aluminum cladding as a function of x at y = 40.7. The schematic on the right indicates the loci of measurement points shown in the plot.
Figure 7. Normal, longitudinal, and transverse residual strains in the aluminum cladding as a function of x at y = 40.7. The schematic on the right indicates the loci of measurement points shown in the plot.
Figure 8. Normal, longitudinal, and transverse residual strains in the aluminum cladding as a function of x at y = 19.7. The schematic on the right indicates the loci of measurement points shown in the plot.
Figure 9. Normal, longitudinal, and transverse residual strains in the aluminum cladding as a function of x at y = 16.4. The schematic on the right indicates the loci of measurement points shown in the plot.
Figure 5. Normal, longitudinal, and transverse residual strains in the aluminum cladding as a function of $x$ at $y = 13.4$. The schematic on the right indicates the loci of measurement points shown in the plot.
Figure 11. Normal, longitudinal, and transverse residual strains in the aluminum cladding as a function of x at y = 118.3 (reflected to y = 9.1mm). The schematic on the right indicates the loci of measurement points shown in the plot.
Figure 12. Normal, longitudinal, and transverse residual strains in the U10Mo foil as a function of x along the center line of the sample y = 63.8mm. The schematic on the right indicates the loci of measurement points shown in the plot.
Figure 13. Normal, longitudinal, and transverse residual strains in the U10Mo foil as a function of x at y = 40.7mm. The schematic on the right indicates the loci of measurement points shown in the plot.
Figure 14. Normal, longitudinal, and transverse residual strains in the U10Mo foil as a function of x at y = 25.7mm. The schematic on the right indicates the loci of measurement points shown in the plot.
Figure 15. Normal, longitudinal, and transverse residual strains in the U10Mo foil as a function of x at y = 19.7mm. The schematic on the right indicates the loci of measurement points shown in the plot.
Figure 16. Contour plots of residual stress in aluminum cladding. Contours go from -50MPa to 100MPa in 10MPa increments. * represent measurement points.
Figure 17. Contour plots of residual stress in U10Mo foil. Contours go from -325MPa to 100MPa in 25MPa increments. * represent measurement points.