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**OVERVIEW OF LANL PROGRESS IN PROCESS DEVELOPMENT,
ADVANCED CHARACTERIZATION METHODS AND PROTOTYPE
FABRICATION**

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ABSTRACT

Recent progress at LANL in process development, advanced characterization method development, prototype fabrication and alternate process development for monolithic LEU fuel will be discussed. Process development studies included HIP (Hot Isostatic Pressing) can design development and plasma spraying of zirconium. Progress on design and testing of formed HIP cans will be presented. Advanced characterization method development concentrated on interface bond strength measurement. Fracture energy results from both bulge testing and notched minicantilever testing will be presented. Maps of zirconium thickness on cold rolled LEU foils were performed using x-ray fluorescence. Residual stress measurements were made using synchrotron x-ray radiation and the incremental slitting method. The processing which produced two prototypical LEU foils will be discussed.

Introduction

This paper provides an overview of work at LANL in process development, development of bond strength measurement, residual stress measurements and prototype development over the last year. Process development was oriented towards producing full scale prototypes, resolving scale up issues, and developing process variations such as canless HIP which have potential for fuel production cost savings. The process development overview will include casting, rolling, HIP can fabrication, canless HIP, plasma spraying, laser ablation and promotion of aluminum grain growth across the HIP (Hot Isostatic Pressing) bond line. The bond strength measurement method overview will include the controlled bulge test and mini-cantilever test. The residual stress overview will include recent synchrotron measurements, and incremental slitting (crack compliance method) measurements. Selected topics will be discussed in more depth in separate posters and papers.

Casting Development for RERTR-FE Foil Fabrication

The initial casting done at LANL was done with the minimization of LEU machining in mind. The original plan was to cast a large ingot, homogenize the ingot, and bare hot roll the ingot to a thickness which was suitable for machining into separate co-rolling coupons. A number of issues with the shape of the hot rolled ingot and subsequent machining prompted interest in casting closer to the final coupon thickness. A combination of heat flow and fluid flow modeling (Flow3D computational fluid dynamics) and in-situ monitoring of casting and mold temperatures with more than ten thermocouples lead to an improved three part mold design for three zone vacuum induction melting (VIM) and casting process. Table 1 shows the mold geometry iterations used and the high (> 90%) U-10Mo metal yield achieved for the optimized mold geometry. Figure 1 shows an ingot from the optimized mold with thickness measurements. This ingot was cast to a very thin and uniform dimension (5 mm).

Table 1 Iterations in mold geometry and resulting

Nominal Compositi	Mold Dimensions			Hot Top Dimensions			Charge Wt (kg)	Casting Wt (kg)	Yield fraction
	Height (mm)	Width (mm)	Thickness (mm)	Height (mm)	Width (mm)	Thickness (mm)			
DU-7Mo	203	203	13	51	227	18	9.606	9.006	0.938
DU-10Mo	203	203	13	51	227	18	9.502	8.960	0.943
DU-10Mo	206	155	17	99	188	23	13.918	12.965	0.932
DU-10Mo	206	155	17	99	188	23	13.900	13.275	0.955
LEU-10Mo	108	148	14	83	148	18	4.395	3.625	0.825
DU-10Mo	105	140	14	72	140	18	4.399	4.256	0.967
DU-10Mo	115	295	5	80	295	10	4.404	3.878	0.881
DU-10Mo	115	295	5	80	295	10	4.379	4.150	0.948
DU-10Mo	115	295	5	80	295	10	4.401	4.101	0.932
DU-10Mo	115	295	5	80	295	10	4.411	4.099	0.929
DU-10Mo	115	295	5	80	295	10			
DU-10Mo	115	295	5	80	295	10			

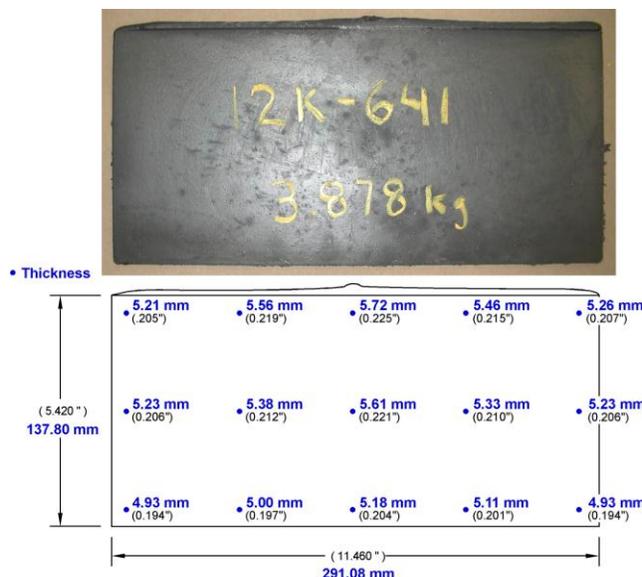


Figure 1 Photograph of DU-10Mo Ingot 12K-641 and schematic showing ingot thickness measurements. Pour temperature 1328C. Mold top was 1200C. Bottom of mold cavity was 750C. R. Aikin

RERTR-FE Foil Fabrication

An LEU U-10Mo casting (11K-635) was bare hot rolled using a salt bath (650 C), flattened at room temperature, homogenized in vacuum (1000C for four hours), machined into individual co-rolling coupons. Two

machined coupons were canned, co-rolled (650C), de-canned and cold rolled using INL rolling procedure [1]. The co-rolled LEU foils are shown in Figure 2a after decanning. Foil 11K-635-A had a split or defect which was sheared off prior to cold rolling. Figures 2b and 2c show the two foils after cold rolling. Foil 11K-635-A was cold rolled to a fuel meat thickness of 0.33 mm, and foil 11K-635-B1 was cold rolled to a fuel meat thickness of 0.5 mm. No edge defects or cracking was observed. It should be noted that cold rolled foil 11K-635-A was stored in a plastic bag in a laboratory with naturally low humidity for over eight months and showed no indications of cracking or bowing.

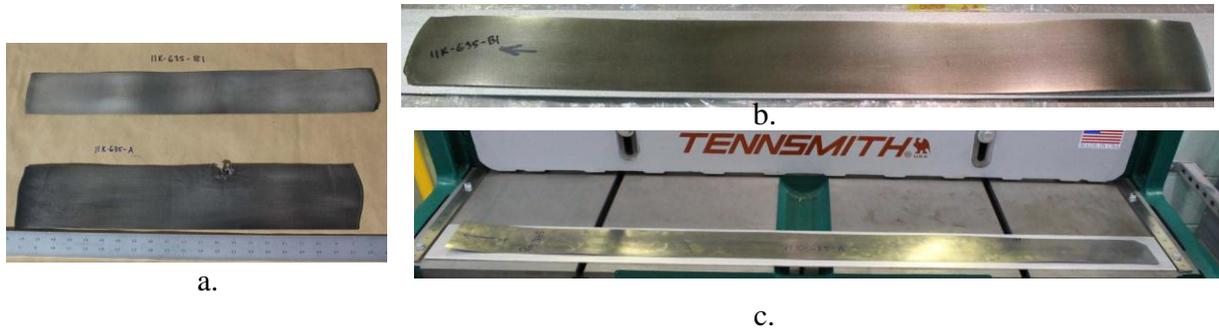


Figure 2 Photographs of LEU U-Mo foils. a. Co-rolled foils after decanning. b. Cold rolled foil 11k-635-B1. Overall dimensions are 0.737 m x 81 mm x 0.67 mm. c. Cold rolled foil 11k-635-A. Overall dimensions are 1.26 m x 74 mm x 0.38 mm. D. Hammon, K. Clarke, J. Scott

Figure 3 shows the thickness of the roll bonded zirconium (Zr) layer on one surface of foil 11K-635-A as measured using a calibrated, handheld x-ray fluorescence unit. Zr thickness varied between 0.020 and 0.022 mm. Some of the mapping shows low Zr thickness at the edges, but this is an artifact attributed to the overlap of the foil and tabletop detector in the detector field of view. Greater accuracy resolution of the Zr thickness at the edges will be obtained by use of the macro-XRF unit.

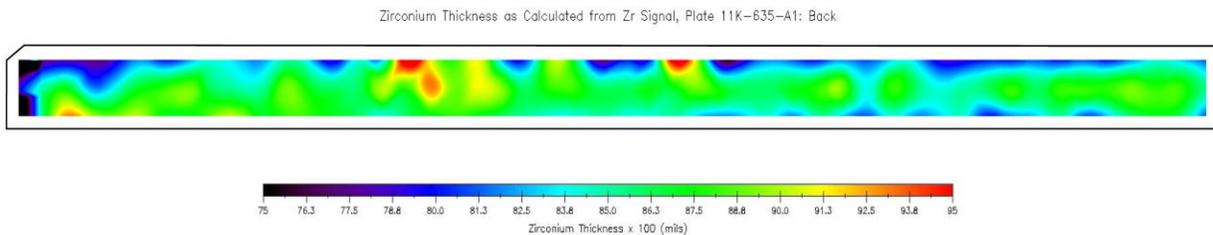


Figure 3 Thickness of the roll bonded Zr layer on one surface of foil 11K-635-A. D. Summa

HIP Can Development

Several initiatives have been started to reduce the cost of HIP can fabrication, including use of formed HIP cans and canless HIP. The present method of HIP can fabrication in the program is to have a trained welder build each six sided HIP can by hand from six sheets of stainless steel. This is a labor intensive process and the weld volume and location often leads to significant dimensional distortions even before the can is loaded with foils and cladding. One method of minimizing the weld volume and labor time is to use a can which uses a stainless steel sheet mechanically formed into a five sides of a HIP can. The contents could be loaded from the open

top, and only the top would have to be welded on to complete the HIP can. An example of a formed can is shown in Figure 4. This example and results are described in a separate poster and paper at the conference [2].



Figure 4 Example of a formed HIP can. a. Round formed HIP cans used to demonstrate feasibility b. Schematic of hydroforming press used to make round formed HIP cans. K. Clarke

Canless HIP is an attempt to decrease HIP can fabrication costs and bonding force distribution issues by eliminating the stainless steel HIP can altogether. The concept is to seal the U-10Mo foil in aluminum cladding under vacuum before it is placed in the HIP. Electron beam welding is used to weld the perimeter of the top and bottom aluminum cladding together. Since the electron beam welding process is performed in vacuum, there is a vacuum between the U-10Mo foil and the cladding after the aluminum cladding pieces have been welded together. A schematic of the process is shown in Figure 5a. Electron beam welding trials were made on 610 mm long 6061 aluminum cladding and surrogate foil (stainless steel) assemblies. Figure 6b shows two plates after e-beam welding and HIP. The plates show a small amount of thickness decrease and no inflation. This indicates that there was no major leak at the electron beam welds. Metallographic examination will be used to confirm aluminum to aluminum bonding and aluminum to stainless steel bonding. More details are available in a separate poster and paper at the conference [2]

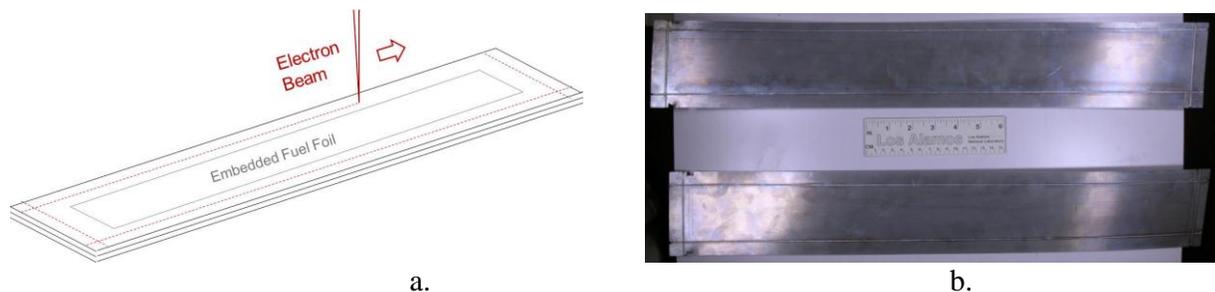


Figure 5 Canless HIP a. Process schematic b. Two plate assemblies (Al/stainless steel/ Al) after electron beam welding and HIP. C. Cross

Grain growth across the Al-Al bond line in HIP'd monolithic fuel plates is desirable but has yet to be attained; at least 25% grain growth across the bond line is desired. To improve this, studies of the effect of aluminum cladding surface preparation on the bond lines of HIP'd Al-Al

sandwiches are being performed. Electron back scatter diffraction (EBSD) is being used to characterize the aluminum grain growth. A comparison of two different cleaning processes on the bond line is shown in Figure 6. More details are available in a separate poster and paper at the conference [2]

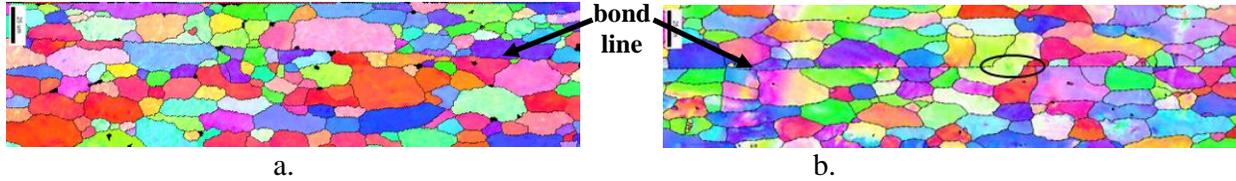
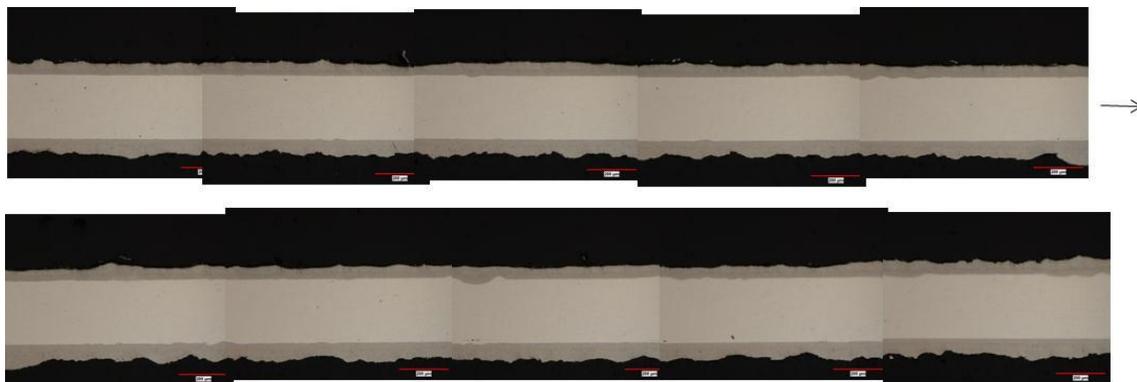


Figure 6 EBSD characterization of bond line for two different surface cleaning processes. a. LANL cleaning produced 0.4% grain growth at bond line (none visible here) b. Babcock & Wilcox cleaning process produced 1.8% grain growth seen in circled region. R. Hackenberg

Plasma Spraying a Diffusion Barrier Coating on U-Mo Foils

The standard way of applying a zirconium diffusion barrier coating on U-Mo foils is a roll bonding method called co-rolling. Applying these coatings by plasma spraying may decrease U-10Mo recycling costs because all the rolling scrap would be uncoated U-Mo. Uncoated U-Mo is less expensive to recycle than zirconium coated U-Mo. Use of plasma spraying would also remove the canning costs associated with co-rolling. Low pressure plasma spraying (LPPS) has been used to produce zirconium coatings and molybdenum coatings on stainless steel foils and U-10Mo foils. Uniformity of a zirconium coating on DU-10Mo foil is shown by optical metallography in Figure 7. Spot measurements of zirconium thickness were made using a handheld x-ray fluorescence (XRF) unit. These are shown in Figure 8. Details on surface roughness, grain size and morphology, and scale up to a 610 mm long foil are available in a separate poster and paper at the conference [3].



Zr on SS thickness montage, mean porosity <math><0.5\%</math>, 200 μm marker, 40-90 μm Zr

Figure 7 Optical metallography image of plasma sprayed zirconium thickness and porosity. K. Hollis

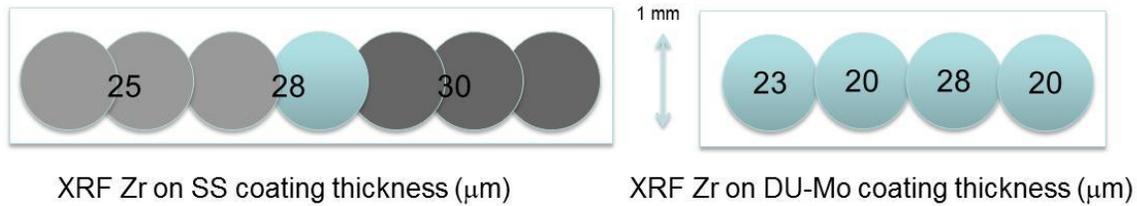


Figure 8 Spot measurements of plasma sprayed zirconium thickness on two different foil materials. K. Hollis, D. Summa

Integrated Intelligent Machining

Machining of monolithic fuel plates to final thickness can be difficult and costly if there are deviations in foil position or shape, or aluminum thickness. In the worst case, these factors could combine and result machining of the aluminum below the minimum aluminum cladding thickness (Al MinClad) which is required for safe reactor operation. Integrated Intelligent Machining combines x-ray fluorescence, pulsed echo, time-of-flight and ultrasonic phased array measurement methods to provide real time aluminum thickness (Al MinClad) guidance during plate machining. For aluminum thicknesses above 0.8 mm, ultrasonic testing provides fast, accurate results. An ultrasonic probe such as the ultrasonic squirter system shown in Figure 9 can be directly attached to the machining head and used for real time measurements on aluminum thickness. Such a system could with appropriate calibration use normal machining coolants. For aluminum thicknesses below 0.8 mm, a portable x-ray fluorescence source as shown in Figure 9 can provide more accurate aluminum thickness readings because the ultrasonic methods have more scatter in their measurements due to surface roughness. Calibration of XRF measurements is done using attenuation curves as shown in Figure 10. Aluminum composition affects calibration. For very thin aluminum ($< .125$ mm), fluorescence absorption is dominated by intervening Zr layer. For aluminum thickness $> .65 - .75$ mm, internal scattering becomes more pronounced

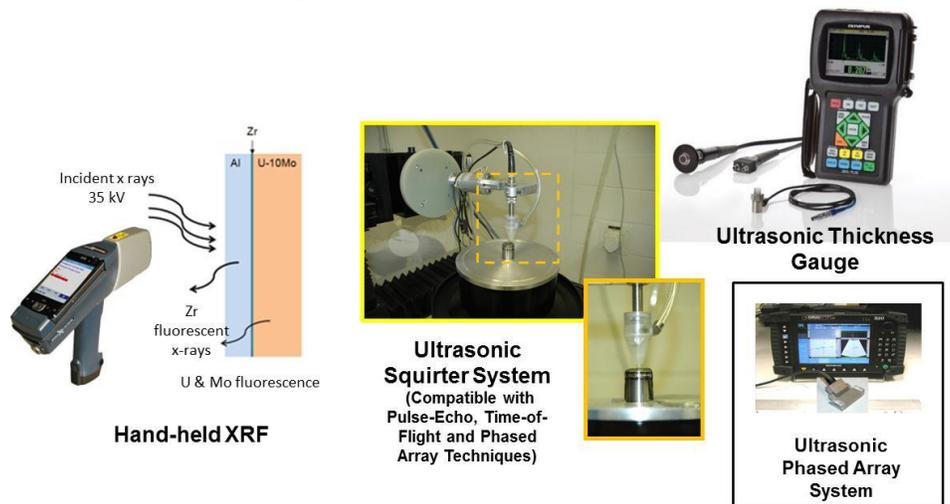


Figure 9 Integrated Intelligent Machining combines x-ray fluorescence, pulsed echo, time-of-flight and ultrasonic phased array measurement methods for real time MinClad guidance during plate machining. D. Summa.

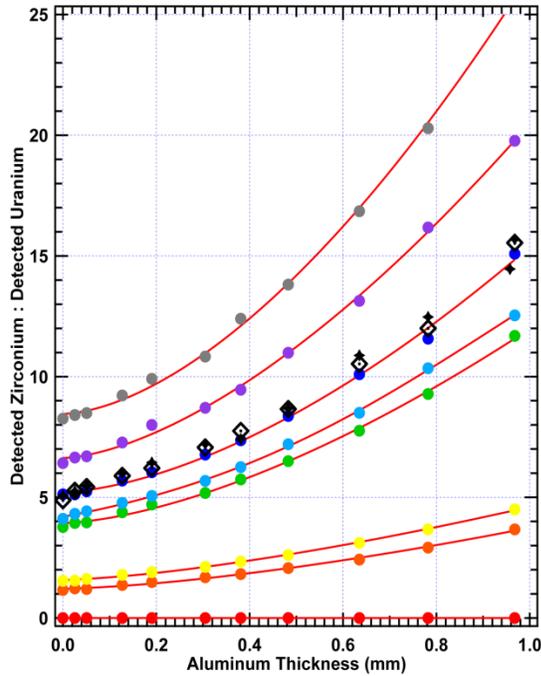


Figure 10 Al thickness calibration requires examination of *ratio* of detected Zr to detected U signals. D. Summa

Bond Strength Characterization

Bond strength measurements and ideally fracture toughness values are needed for fuel qualification as well as for development of quality control standards. Bond strength measurements for thin materials such as fuel plates are very challenging because there is not enough material to grip and apply mechanical force. There are three bond strengths needed: the aluminum to aluminum bond, the aluminum to zirconium bond, and the U-Mo to zirconium bond. Two methods have been under development at LANL, the controlled bulge test and the minicantilever test. The controlled bulge test uses a

fluid under an interface to cause a deflection in the plate and ultimately a delamination at the desired interface. That round upward deflection is quantitatively characterized by using Digital Image Correlation (DIC). An energy analysis yields bond strength and interfacial fracture. Figure 11 shows a schematic cross section of the fixture used for measurements, the initial data obtained, and the conceptual breakdown of total energy into components including the delamination energy. More details are available in a separate poster and paper at the conference [4].

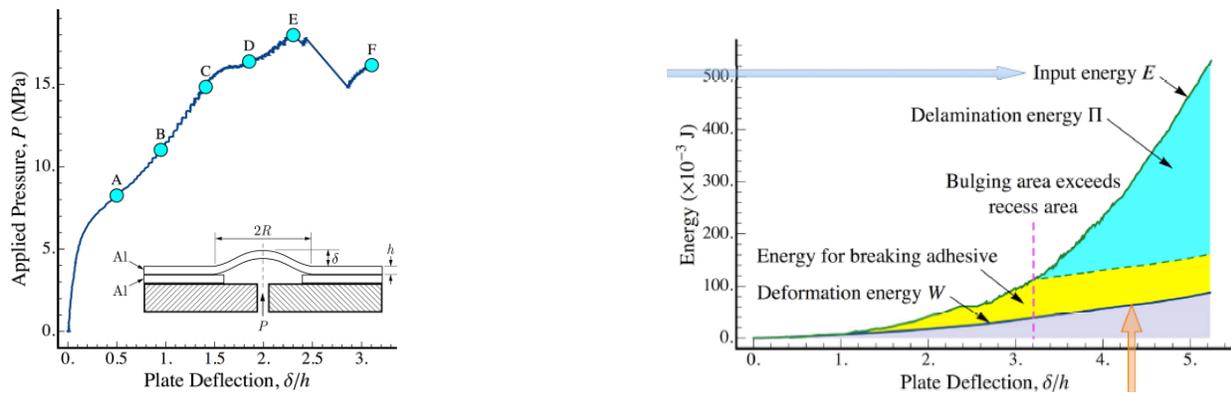


Figure 11 Controlled bulge test a. Schematic cross section of plate in testing fixture and pressure versus volume curve obtained from test b. Schematic description of energy analysis used to obtain the delamination energy. C. Liu, M. Lovato

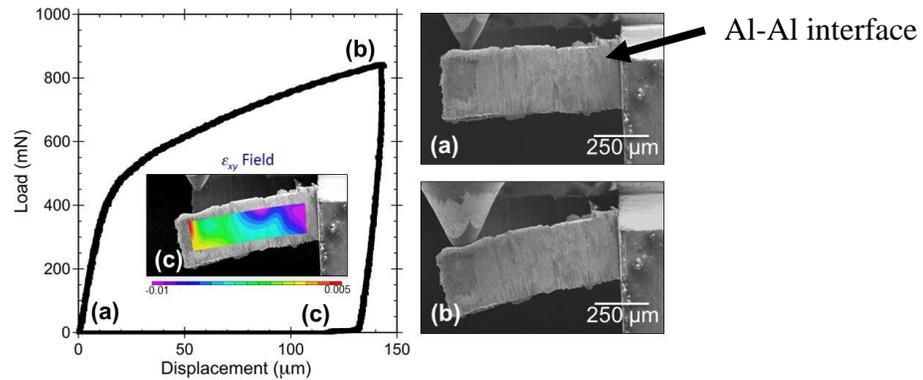


Figure 12 The load displacement curve for a minicantilever test is shown on the left. The inset shows a DIC strain map. a. The mini-cantilever test for an Al/Al interface begins with deflection of the cantilever with a nanoindenter. b. Final deflection or fracture. N. Mara, W. Mook

Previous work [5] had developed a micro-cantilever which was able to measure interface strength of all three interfaces. However that technique used a Focused Ion Beam to make the microcantilevers and nanoindenter to test the beams. It would not be feasible in present hot cells. The mini-cantilever test was designed to utilize much simpler equipment to machine the cantilever and test it. The method has been used for monolithic Al minicantilevers made from HIP'd Al, minicantilevers machined from an Al/Al interface and minicantilevers machined from Al/Zr interfaces. Although the minicantilevers can be tested in small load frames in a hot cell, initial work was done in-situ in an SEM for diagnostic purposes. Figure 12 shows a typical test for an Al/Al interface minicantilever. More details are available in a separate poster and paper at the conference [4].

Residual Stress Measurement

Residual stress measurements have been performed using synchrotron radiation and the incremental slitting (compliance) method. Synchrotron measurements were made of residual stress in INL miniplates. Each miniplate had a different combination of foil annealing (no annealing or annealed) and cooling rate at the end of a HIP cycle (slow cooling or fast cooling). Results are shown in Figure 13. In-plane strains are generally similar for the different combinations of process conditions with one exception. The high gradient of in-plane strain near the plate end which is observed for the fast cooling sample is not observed for slow cooling samples. More details are available in a separate poster and paper at the conference [4].

Residual stress measurements have been made as a function of depth using the incremental slitting method on two HIP'd plates containing DU-Mo foils. The plates are each nominally 610 mm long and 76 mm wide. The thin plate was nominally 1.6 mm thick and the thick plate was nominally 3 mm thick. The experimental set up is shown in Figure 14. A preliminary plot of residual stress versus depth has been generated. Final results will be available once microscopy confirms the actual thicknesses of Al, Zr and DU-Mo at the slit location for each plate. Preliminary results indicate that there is a pattern of compressive stress in the DU-Mo. The residual stress magnitudes appear to be limited by low strength of 6061 Al after thermal processing in the HIP. More details are available in a separate poster and paper at the conference [4].

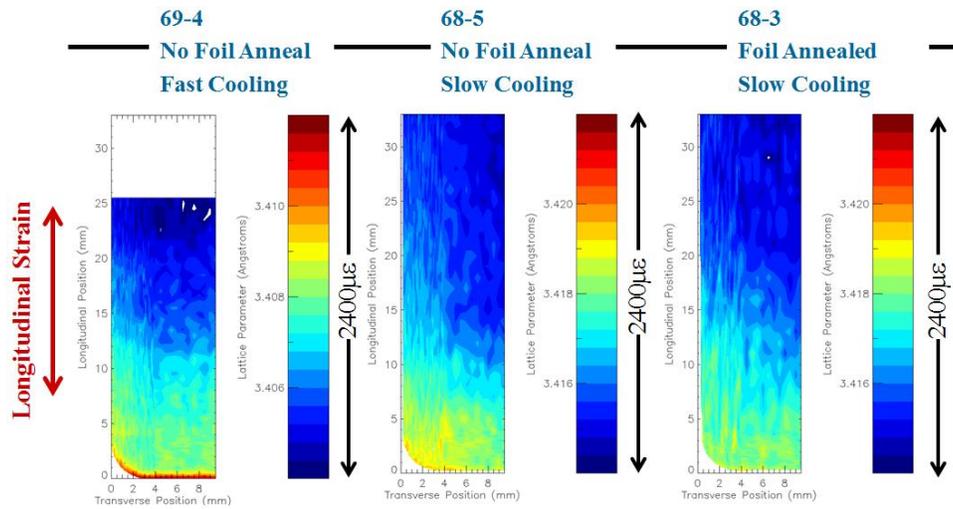


Figure 13 Residual stress maps calculated from synchrotron x-ray radiation of miniplates.

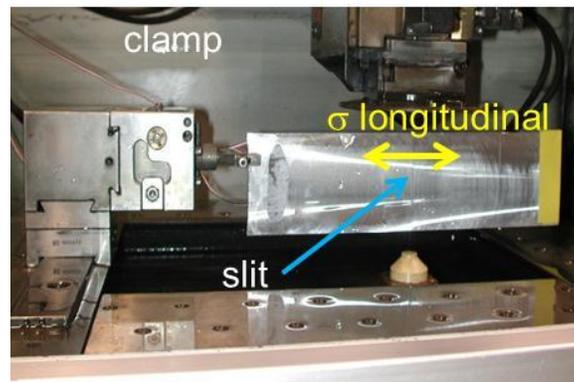


Figure 14 Experimental set up for performing incremental slitting measurements using electric-discharge machining (EDM). A pair of transducers on the side opposite the slitting is used for primary data acquisition.

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References

1. G. A. Moore, M. C. Marshall, Co-Rolled U10Mo/Zirconium-Barrier-Layer Monolithic Fuel Foil Fabrication Process, INLreport INL/EXT-10-17774, January 2010

2. Kester D. Clarke, Carl E. Cross, Robert E. Hackenberg, Rodney J. McCabe, Joel D. Montalvo, Matthew J. Dvornak, Randall L. Edwards, Justin M. Crapps, Ralph R. Trujillo, Beverly Aikin, Victor D. Vargas, Kendall J. Hollis, Bogdan Mihaila, Duncan Hammon, Richard W. Hudson, Tim J. Tucker, Jeffrey E. Scott, Andrew N. Duffield, Richard Y. Weinberg, David E. Dombrowski, Development of Aluminum-Clad Fuel Plate Processing Through Canned and Canless Hot Isostatic Pressing (HIP), and Studies of Aluminum Cladding Grain Growth during HIP, RERTR 2012, October 14-17 2012, Warsaw, Poland
3. K.J. Hollis, Plasma Spraying of Diffusion Barrier Coatings for LEU Monolithic Fuel, RERTR 2012, October 14-17 2012, Warsaw, Poland
4. C. Liu, M.L. Lovato, D.J. Alexander, K.D. Clarke, N.A. Mara, W.M. Mook, M.B. Prime, D.W. Brown, Experimental Investigation of Bonding Strength and Residual Stresses in LEU Fuel Plate, RERTR 2012, October 14-17 2012, Warsaw, Poland
5. N. Mara, J. Crapps, T. Wynn, K. Clarke, P. Dickerson, B. Mihaila, D. Dombrowski, Nanomechanical Behavior of U-10Mo/Zr/Al Fuel Assemblies, October 23-27, 2011, RERTR 2011, Santiago, Chile