RERTR 2009 — 31^{st} International Meeting on Reduced Enrichment for Research and Test Reactors

November 1-5, 2009 Kempinski Hotel Beijing Lufthansa Center Beijing, China

MONOLITHIC FUEL FABRICATION PROCESS DEVELOPMENT AT THE IDAHO NATIONAL LABORATORY

 G. A. Moore, F. J. Rice, N. E. Woolstenhulme, J-F. Jue, B. H Park, S. E. Steffler, N. P. Hallinan, M. D. Chapple, M. C. Marshall, B. L. Mackowiak, C. R. Clark and B. H. Rabin Fuel Fabrication Idaho National Laboratory Idaho Falls, ID 83415 USA

ABSTRACT

Full-size/prototypic U10Mo monolithic fuel-foils and aluminum clad fuel plates are being developed at the Idaho National Laboratory's (INL) Materials and Fuels Complex (MFC). These efforts are focused on realizing Low Enriched Uranium (LEU) high density monolithic fuel plates for use in High Performance Research and Test Reactors. The U10Mo fuel foils under development afford a fuel meat density of ~16 gU/cc and thus have the potential to facilitate LEU conversions without any significant reactor-performance penalty. An overview is provided of the ongoing monolithic UMo fuel development effort, including application of a zirconium barrier layer on fuel foils, fabrication scale-up efforts, and development of complex/graded fuel foils. Fuel plate clad bonding processes to be discussed include: Hot Isostatic Pressing (HIP) and Friction Bonding (FB).

Introduction

The processes used for fabrication of monolithic fuel plates are significantly different than those used for fabricating traditional dispersion fuel plates; given that the metallic fuel foil is prepared to the desired thickness prior to application of the aluminum cladding. Integral to the current fuel foil processing scheme is the incorporation of a zirconium barrier layer for the purpose of controlling UMo-Al interdiffusion at the fuel-meat/cladding interface. [1, 2] A hot "co-rolling"

This manuscript has been authored by Battelle Energy Alliance, LLC under Contract No. DE-AC07-05ID14517 with U.S. Department of Energy. The United States Government retains and the publisher, by accepting the article for publication, acknowledges that the United States Government retains a nonexclusive, paid-up, irrevocable, world-wide license to publish or reproduce the published form of this manuscript, or allow others to do so, for United States Government purposes.

process is employed to establish a $\sim 25 \mu m$ thick zirconium barrier layer on each face of the ~ 0.3 mm thick U10Mo fuel foil, Figure 1

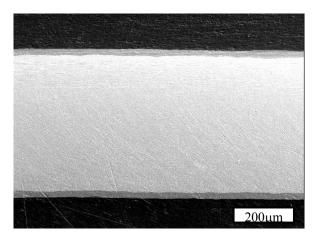


Figure 1. Cross sectional scanning electron microscope (SEM) micrograph of a Zr co-rolled U10Mo fuel foil.

Currently, two processes for cladding monolithic UMo fuel foils are being developed at the INL: 1) friction bonding (FB) and 2) hot isostatic pressing (HIP).[3-5] The FB process makes use of thermo-mechanical energy imparted via the application of a contacting rotary-tool. As the tool is traversed along the Al-foil-Al layup, down-force is applied to an extent that facilitates clad bonding, but not tool-contact with the fuel foil. Water cooling within the tool, rotational speed, translation feed rate, and down force are parameters used to controlled the FB process; a programmable computer interface is utilized for equipment control and data collection. The HIP process involves the application of heat and pressure upon a hermetically sealed, evacuated sample canister containing a stack of cladding/fuel foil sheets.

This paper described the fabrication steps utilized to prepare U10Mo Zr co-rolled fuel foils, discussion of the two clad bonding processes utilized, fuel plate characterization, and on-going process development activities.

2. Experimental

The processing sequence developed at the INL for the preparation of U10Mo Zr co-rolled monolithic foils is shown in Figure 2. A six step process is used to obtaining monolithic fuel foils suited to either the FB or HIP clad bonding process. A description of these processing steps and important attributes/considerations is provided in this section.

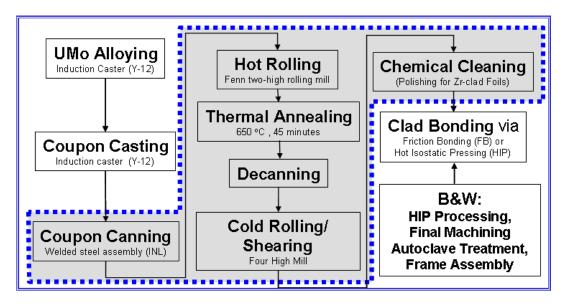


Figure 2. Monolithic U10Mo foil fuel plate fabrication process.

U10Mo Foil Preparation

The starting point of INL full size fuel foil fabrication is the preparation of cast U10Mo coupons for co-rolling. Coupons were provided by the Y-12 National Security Complex, Oak Ridge, TN. Face-machined and/or rolled-to-thickness coupons, \sim 75 mm x 100 mm x 3.25 mm, were filed to remove sharp edges and corners; in order to prevent "can breaching" in subsequent processing steps. The U10Mo coupons were chemically cleaned, to remove surface oxide contamination, using a \sim 30% nitric acid solution followed by a deionized water (DI) rinse.

Rolling assembly preparation involves laminating a U10Mo coupon with zirconium foil on each coupon face, Figure 3. The Zr foil contacting faces of the steel rolling assembly plates were coated with colloidal graphite paint "Neolube" and dried to establish "no-stick" surfaces; allowing the foil to be removed after co-rolling. After lay-up of the low carbon steel picture frame/cover components, U10Mo coupon, and Zr foils, the assembly was edge-welded inside of an argon atmosphere glove box to produce a "canned" rolling assembly.

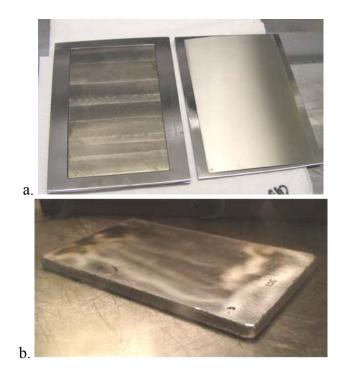


Figure 3. a) Full size U10Mo coupon hot rolling assembly lay-up, b) welding hot rolling assembly.

Rolling can assemblies were preheated in a box furnace at 650 °C for 30-45 minutes prior to rolling. Assemblies were then individually removed from the box furnace, and given one-four passes, as quickly as possible, through a two high rolling mill. Each assembly was then placed back in the furnace and reheated at 650 °C for 5-15 minutes. After the reheat step, the process was repeated. A total of 20-40 passes was typical for preparation of a 0.25-0.50 mm thick U10Mo foils. Figure 4, provides an example of the assembly thickness reduction over time The maximum per pass force was monitored and recorded during rolling operations.

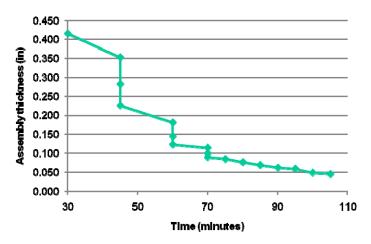


Figure 4. Graph showing rolling assembly thickness reduction during hot U10Mo Zr co-rolling process.

After hot co-rolling reduction was complete, foil-assemblies were placed back in the furnace for 45 minutes at 650 °C. Heat treated foils were then de-canned by removing the perimeter of the rolling assembly using a hand held power shear, Figure 5.



Figure 5. a) Heat treated co-rolling assembly, b) Full size Zr co-rolled U10Mo fuel foil (~85 mm x 750 mm x 0.5 mm).

Next, Zr co-rolled foils were cold rolled using a four high rolling mill. This step was employed in order to precisely establish the fuel foil thickness and to smooth out fuel meat thickness variations imparted via the co-rolling process, figure 6. Cold rolling reduction passes of 0.015-0.025 mm per were used. The fuel meat thickness variation/texture established during the co-rolling process is believed to be associated with hardness variations in the co-rolling assembly components, i.e. carbon steel, zirconium foil, and U10Mo coupon. After cold rolling, thickness-variation along the foil length were reduced to \sim 0.01-0.02 mm.

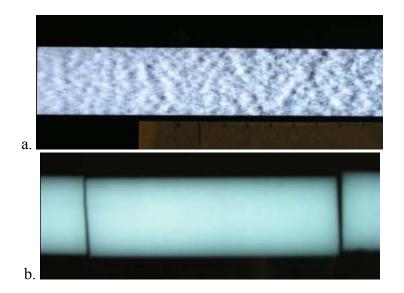


Figure 6. a) Radiographic image of Zr co-rolled foil, b) Radiographic image of Zr co-rolled foil after cold rolling operation.

After cold rolling to final thickness, fuel foils were sheared to length and sheared or slit to width, Figure 7.

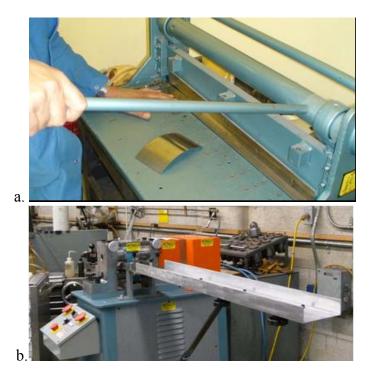


Figure 7. a) Hand operated shear, b) Foil slitting machine.

Prior to cladding application via the HIP or FB process, U10Mo foils were polished using lint free cloths and water soluble diamond paste, 30 µm, in order to remove any surface oxidation.

Removal of polishing compound residue was accomplished via a thorough water rinse and ethanol soak/wipe down, Figure 8.



Figure 8, Polished Zr co-rolled U10Mo fuel foils ready for cladding application.

Cladding Preparation

The cladding material used for both the HIP and FB processes was Al 6061 sheet stock. Two cladding sheets were prepared for each fuel plate assembly, i.e. a top/cover plate (\sim 0.6 mm) and a bottom/pocket plate (\sim 1.0 mm). Both sides of each cladding sheet were brushed using a stainless steel bristle brush chucked in a milling machine tool holder. A foil retention pocket was machined in the bottom cladding sheet to prevent foil shift during processing. For HIP plate processing, cladding sheets were additionally chemically cleaned as outlined in Table 1.

Chemical Cleaning Step	Method	Temp (°C)	Time (sec)
Degrease (Acetone/ethanol)	Hand	RT	-
Basic Etch (2M NaOH solution)	Bath	80-85	20-120
Water Rinse/Desmut	Bath	RT	15-30
Desmut (sponge, cloth, or brush)	Hand		
Pickle (30% nitric acid solution)	Bath	RT	120
Water Rinse	Bath	RT	15-30
Hot Rinse	Bath	80-85	≈15-30
Wipe (Lint Free)	Hand		
Vacuum Seal (if stored)	Hand		

Table 1. Aluminum chemical cleaning processing steps and conditions.

Fuel Plate Fabrication using the Friction Bonding Process

Cladding was bonded to prepared fuel foils using the Friction Bonding (FB) process developed at the Idaho National Laboratory (INL). The process utilizes a water cooled rotating tool configured in the TTI workstation, Figure 9. FB run set-up consisted of laying up a cladding/foil "pack", comprised of the polished fuel foil and two sheets of 6061 aluminum. The lay-up was placed on the working anvil of the TTI FB machine and clamped firmly in place along each long-edge.



Figure 9. TTI Friction Bonding workstation at the INL.

Clad/Clad and Clad/Fuel bonding was accomplished via the following steps::

- Slowly bringing the rotating tool into contact with the cladding material
- Applying a controlled amount of down force and time; in order to generate thermomechanical softening of the cladding material under the tool.
- Traversing the anvil/bed/"cladding/foil pack" lengthwise end to end..
- Stopping the traverse and drawing out the rotating tool from the work piece.
- Returning to the start position and offsetting the work piece laterally in preparation for the next pass.
- Removal of flash material from the previous pass using a scraping tool.
- Repeating above sequence until one side of fuel plate was fully bonded.
- Then, flipping the "cladding/foil pack" over and repeating the friction bonding process on the second side.

Fuel Plates Fabrication using the Hot Isostatic Pressing (HIP) process

Fuel plate samples were HIP processed in a evacuated stainless steel "cans" having four AFIP-4 size samples, (~55 mm x 200 mm); each sample being separated by a "strong back/steel plate" and grafoil sheets. HIP can preparation has been previously reported.[6] HIP runs were conducted at 560 °C for a period of 60 minutes and 100 MPa.

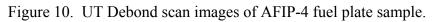
Final Processing Sequence

Prior to final plate processing steps, both friction bonded and HIP processed fuel plates were characterized "qualitatively" for bonding and cladding thickness over the fuel zone region "min clad" using an ultrasonic testing (UT) work station.[7] Figure 10 shows the UT transmission/Debond image of an AFIP-4 size monolithic fuel; overall plate dimensions, 56 mm x 190 mm x 1.3 mm.

Based on a map of the plate thickness and UT based cladding thickness measurements, fuel plates were sanded, on both sides, in order to realize the final fuel plate thickness. A minimum cladding thickness of 0.15 mm, over the fuel region, was maintained during processing. Once sanded, final plate dimensions were established via shearing and final machining operations.

An oxide film "boehmite" was applied to dimensioned fuel plates using an autoclave treatment consisting of a four hour treatment at 185 °C and ~160 psi (1.1 Mpa) in DI water. Prior to autoclaving, fuel plates were chemically cleaned using the sodium hydroxide solution etching process described above; including the nitric acid pickle and water rinse steps. Approximately 0.01-0.02 mm of cladding thickness was removed during the chemical cleaning step.





On going Technology Developments

A graded density fuel meat is required for the High Flux Isotope Reactor (HFIR). As such, the development of complex/grade monolithic fuel foils is being pursued at the INL.[8] In addition to the foil and fuel plate processing activities discusses above, the INL RERTR Fabrication Team has been experimenting with variations and/or enhancements of the monolithic fabrication processes being employed. Namely, casting of complex-shape ingots and hot rolling of surrogate graded fuel foils. Below is a brief discussion of the efforts and results to date.

Graded Fuel Foil Development

A pressure over vacuum induction casting system has been established within the Fuels and Applied Science Building (FASB) at the INL's Materials and Fuels Complex (MFC). Depleted U10Mo casting experiments were recently initiated. The development effort currently underway involves induction casting of a complex geometry ingot/coupon that is subsequently "canned" and hot rolled to produce a foil have thickness variation of 0.125-0.500 mm (0.005-0.020"), Figure 11. Full size foils, 570 mm x 56 mm, have been demonstrated with stainless steel using this method.

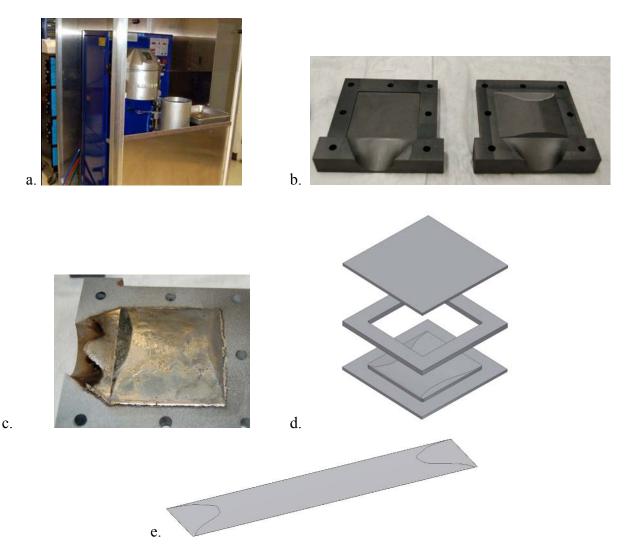


Figure 11. Graded fuel foil fabrication: a) Indutherm pressure over vacuum induction caster, b) graphite mold for complex-shape coupon, c) Image of cast, complex geometry, stainless steal coupon/ingot, d) drawing of three-piece rolling can assembly, and e) drawing of resulting full size contoured foil.

3. Summary

Significant progress in the fabrication of monolithic fuel foils and fuel plates has been made during 2009 by the INL RERTR Fabrication group. Fabrication of ATR fuel plate size Zr corolled U10Mo fuel foils has been demonstrated. During 2009, twelve monolithic LEU fuel plates were irradiated in the INL Advanced Test Reactor (ATR) as part of the AFIP-4 (ATR Full –size-plate in center flux trap Position) experiment.

4 Acknowledgements

Work supported by the U.S. Department of Energy, Office of Nuclear Nonproliferation and Security Affairs (NNSA) under DOE-NE Idaho Operations Office Contract DE AC07 05ID14517.

5. References

- [1] D. D. Keiser and J. F. Jue, "Characterization of Si-rich Layers in As-Fabricated RERTR Fuel Plates: Recent Results," Proceedings of RERTR 2008 — 30th International Meeting on Reduced Enrichment for Research and Test Reactors," Washington, D.C. USA, October 2008.
- [2] G. L. Hofman, Y. S. Kim, J. Rest, T. Totev and A. B. Robinson, "Post-Irradiation Analysis of U-Mo Miniplates Tested to Date," Proceedings of RERTR 2008 — 30th International Meeting on Reduced Enrichment for Research and Test Reactors," Washington, D.C. USA, October 2008.
- [3] C.R. Clark, et al., "Update on Monolithic Fuel Fabrication Development," Proceedings of the 2005 International Meeting on Reduced Enrichment for Research and Test Reactors," Boston, Massachusetts, USA, 2005
- [4] C.R. Clark, et al., "Update on Monolithic Fuel Fabrication Methods," Proceedings of the 2006 Reduced Enrichment for Research and Test Reactors International Conference, Cape Town, South Africa, November 2006
- [5] C.R. Clark, et al., "Full Sized Monolithic Fuel Fabrication Development," Proceedings of the 2006 International Topical Meeting on Research Reactor Fuel Management, Sofia, Bulgaria, 2006
- [6] G. A. Moore, F. J. Rice, N. E. Woolstenhulme, W. D. Swank, D. C. Haggard, J. Jue, B. H Park, S. E. Steffler, N. P. Hallinan, M. D. Chapple, and D. E. Burkes, "Monolithic Fuel Fabrication Process Development at the Idaho National Laboratory," Proceedings of RERTR 2008 — 30th International Meeting on Reduced Enrichment for Research and Test Reactors, Washington, D.C. USA, October 2008.
- [7] J.M. Wight, G.A. Moore, S.C. Taylor, N.E. Woolstenhulme, and S.T. McCormick, "Testing and Acceptance of Fuel Plates for RERTR Fuel Development Experiments," Proceedings of RERTR 2008 — 30th International Meeting on Reduced Enrichment for Research and Test Reactors, Washington, D.C. USA, October 2008.
- [8] N.E. Woolstenhulme, G.A. Moore, W.D. Swank, and D.C. Haggard, "Fabrication of Contoured Monolithic Foils," Proceedings of RERTR 2008 — 30th International Meeting on Reduced Enrichment for Research and Test Reactors," Washington, D.C. USA, October 2008.