RERTR 2008 — 30th INTERNATIONAL MEETING ON REDUCED ENRICHMENT FOR RESEARCH AND TEST REACTORS

October 5-9, 2008
Hamilton Crowne Plaza Hotel
Washington, D.C. USA

MONOLITHIC FUEL FABRICATION PROCESS DEVELOPMENT AT THE IDAHO NATIOANAL LABORATORY

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
Fuel Fabrication
Idaho National Laboratory
P.O. Box 1625, Idaho Falls, ID – USA

ABSTRACT

Within the Reduced Enrichment for Research and Test Reactors (RERTR) program directed by the US Department of Energy (DOE), UMo fuel-foils are being developed in an effort to realize high density monolithic fuel plates for use in high-flux research and test reactors. Namely, targeted are reactors that are not amenable to Low Enriched Uranium (LEU) fuel conversion via utilization of high density dispersion-based fuels, i.e. 8-9 gU/cc. LEU conversion of reactors having a need for >8-9 gU/cc fuel density will only be possible by way of monolithic fuel forms. The UMo fuel foils under development afford fuel meat density of ~16 gU/cc and thus have the potential to facilitate LEU conversions without any significant reactor-performance penalty.

Two primary challenges have been established with respect to UMo monolithic fuel development; namely, fuel element fabrication and in-reactor fuel element performance. Both issues are being addressed concurrently at the Idaho National Laboratory. An overview is provided of the ongoing monolithic UMo fuel development effort at the Idaho National Laboratory (INL); including development of complex/graded fuel foils. Fabrication processes to be discussed include: UMo alloying and casting, foil fabrication via hot rolling, fuel-clad interlayer application via co-rolling and thermal spray processes, clad bonding via Hot Isostatic Pressing (HIP) and Friction Bonding (FB), and fuel plate finishing.

1. Introduction

Traditional aluminum clad dispersion fuel plates, consists of a particulate fuel-phase distributed in an aluminum matrix phase. Fabrication of plate-type fuel elements involves preparation of a fuel/matrix powder compact, encapsulation of the fuel-form compact in an aluminum assembly, followed by hot and cold rolling to obtain a "bonded" fuel plate.

The process of fabricating monolithic fuel plates is significantly different than that of the traditional roll-bonding process; given that the metallic fuel foil is prepared to the desired thickness prior to application of an aluminum cladding process. Thus, when working with a

monolithic fuel form, i.e. a metallic UMo foil, the bonding of the aluminum clad bonding must be accomplished using other clad-bonding techniques.[1-3]

Currently, two processes for cladding monolithic UMo fuel foils are being investigated at the INL: 1) friction bonding (FB) and 2) hot isostatic pressing (HIP). The FB process involves the application of a traversing and rotating tool for plasticizing and forge-bonding the aluminum cladding material to an underlying fuel foil. The HIP process consists the application of heat and pressure to a hermetically isolated cladding/fuel foil lay-up. These two processes impart significantly different time, temperature, and pressure conditions on the materials being bonded. Thus, foil properties and cladding-stock preparation must be tailored to each process. For example, foil annealing treatments are utilized in order to establish foil ductility conducive to the friction bonding process and co-rolled zirconium barrier-layer application can be used for circumventing UMo-Al interfacial reactions that occur during HIP processing.

This paper described the preparation of U10Mo fuel foils and aluminum cladding for used in the HIP and FB plate fabrication processes; including zirconium clad UMo foils and 6061 Al cladding having a silicon-rich aluminum thermal-spray coatings at the fuel-clad interface for the purpose of stabilizing the irradiation-induced UMo-Al reaction product.[4, 5]

2. Experimental

The processing sequence developed at the INL for the preparation of U10Mo monolithic foils is shown in Figure 1. A seven step process is employed prior 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.

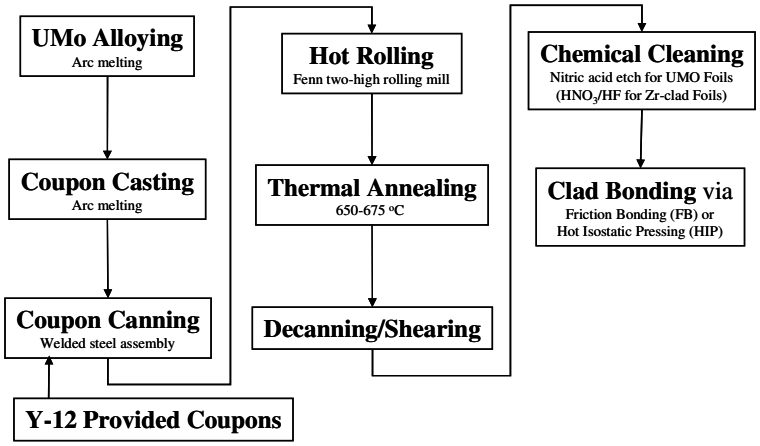


Figure 1. Monolithic UxMo foil fuel plate fabrication process.

U10Mo Foil Preparation

Uranium feedstock is prepared for alloying via a 30% nitric acid solution etch/cleaning step in order to remove surface oxide deposits. Cleaned uranium metal and 99.95% pure molybdenum foil is alloyed using a Centorr Vacuum Industries Model 5SA single arc furnace that resides in an

argon glove box having <50 ppm oxygen. U10Mo alloy buttons are turned/flipped and remelted three or four time in order insure homogeneity.

After the alloying sequence is complete, the arc melter is configured with a casting-hearth assembly and the alloy "button" is drop cast into a rectangular "coupon" mold comprised of copper, steel, and/or graphite construction. Subsequently, the casting sprue is removed and edges dressed to yield a 30-35g coupon, ~38mm x 25mm x 3mm.

A hot-rolling process is used to prepare UMo fuel-foils from cast alloy coupons. Rolling assembly preparation involves laminating UMo coupon(s), with or without zirconium or Niobium foils on each face, inside of a carbon steel picture-frame assembly, Figure 2. After the bottom and cover plates are applied, the assembly is perimeter-welded inside of an argon glove box, thus, producing a "canned" assembly that can be heated and rolled in air. The "canning process has also been successfully performed using larger, ~65mm x 115 mm x 3 mm, machined U10Mo "full size" coupons prepared at the Y-12 National Security Complex.

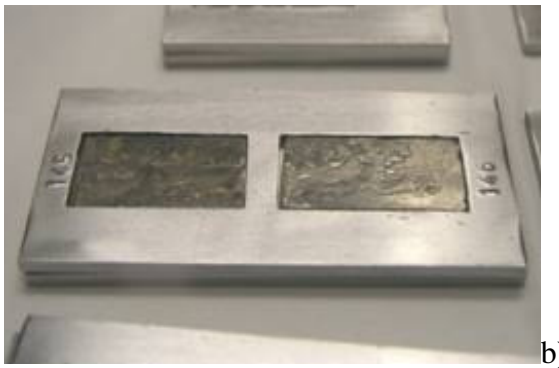
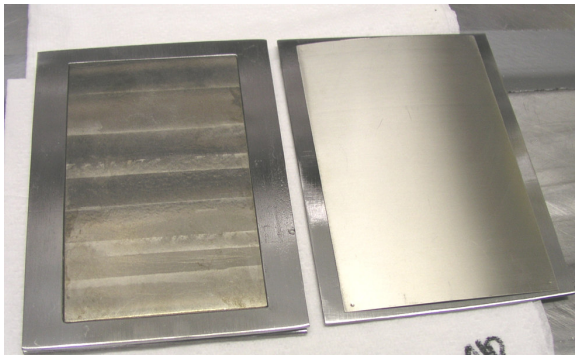
a)  b) 

Figure 2. UMo coupons hot rolling assemblies in process, a) minifoil size coupons and b) full size coupon.

Canned coupon assemblies are preheated in a box furnace at 650-700 ^{o}C for 45-60 minutes prior to rolling. Assemblies are individually removed from the box furnace, and receive two-four passes, as quickly as possible, through a two high rolling mill. The assembly is then placed back in the furnace and reheated at 650 ^{0}C for several minutes. Ten to twenty reheat cycles in conjunction with 20-40 passes is typical for preparation of a 0.25-0.50 mm thick UMo foils. Figure 4, provides an example of the thickness reduction over time during a typical hot rolling session. The maximum per pass force is monitored and recorded during rolling operations.

For the rolling schedule data shown in figure 3, two passes per sequence are represented for each of the first nine data points. The remaining eleven data points reflect single passes made between each furnace reheat. A total of 29 rolling passes were performed. It is observed, that the rolling force incrementally increases until a 30 minutes reheat is performed at the 165 min mark. Thereafter, 15 min reheats were performed after each pass. The significant reduction in pass force after the 30 minute reheat is due to less aggressive reduction; employed in order to prevent rippling of the rolling assembly/foil.

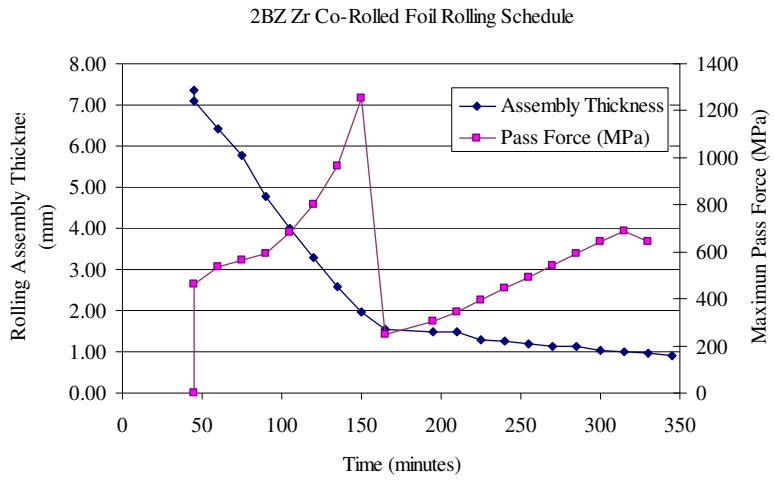


Figure 3. Graph of rolling assembly thickness and pass force throughout Zr co-rolled U10Mo foil processing.

After rolling is complete, foil-assemblies are returned to the box furnace and annealed at 650-675 ^{o}C for 30-120 minutes, in order to impart ductility. Heat treated foils are subsequently de-canned and sheared to the desired size, Figure 4.

a.  b. 

Figure 4. a) Decanned and coarse sheared full size Zr co-rolled U10Mo fuel foils (~500 mm x 240mm x 0.3 mm), b) sheared minifoils (83mm x19mm x 0.25 mm).

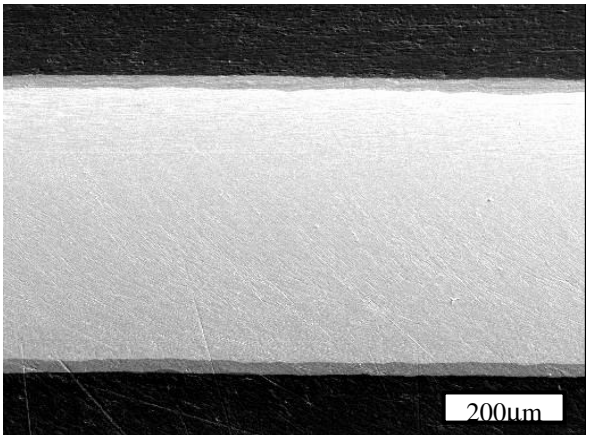


Figure 5. Cross sectional scanning electron microscope (SEM) micrograph of a nominally 0.010" thick U10Mo fuel foil with Zr co-rolled barrier layer.

Prior to cladding application via the HIP or FB process, UMo foils are wet sanded and chemically cleaned in order to remove oxidation and to provide a roughened surface conducive to bonding. Sanding is performed using 80-160 grit emery cloth and deionized water. During sanding, foil edges are lightly sanded to remove small burs and sharp edges. Chemical cleaning is then conducted as follows: 1) Immerse foil in 30-40% nitric acid solution for ~15-60 seconds; spot brush as needed, 2) Rinse foil in deionized water for ~20-30 seconds, 3) Rinse foil in absolute ethyl alcohol for 20-30 seconds, 4) Dry foil with clean lint free cloth, 5) Repeat process, as needed, until a uniform metallic finish is obtained on areas of the foil. In the case of co-rolled foils having a zirconium barrier layer, cleaning is performed using a mixture of nitric and hydrofluoric acid (~2.5% HF, ~35% HNO_3, 62.5% H_2O), followed by a thorough water rinse and ethanol soak/wipe down.

Cladding Preparation

Al 6061 sheet stock is used in both the HIP and FB processes; two cladding sheets/plates are prepared for each fuel plate assembly, i.e. a top/cover plate (~0.6 mm) and a bottom/pocket plate (~1.0 mm). Both sides of each cladding sheet were brushed using a 75 mm diameter stainless steel bristle brush chucked in a milling machine tool holder The thickness of each cladding plate is prepared such that a final nominal cladding thickness after final surface finishing is 0.6 mm. A foil retention pocket is machined in the bottom cladding sheet to prevent foil shift during processing. Prior to clad bonding, the cladding sheets are then 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.

Thermal Spray Barrier Coating Application.

The thermal spray coating process is utilized to apply a silicon containing barrier layer to prepared aluminum cladding. Specifically, to the areas to the cladding sheet that will be in direct contact with the fuel foil. The process consists of generating a hot gas jet by passing argon or an argon-helium mixture through an electric arc discharge where it is heated and ionized to form a thermal plasma. The gas rapidly expands and exits the torch at 12000K and 1.5 Km/sec. Metallic or ceramic particles are injected into the jet where they are heated and accelerated to be deposited on the substrate. A thin, 0.01mm to 0.002mm thick coating is formed

In the case of cladding designated for thermal spray application, the following steps are performed:

- Using a template/mask and a micro grit blaster, the area to be coated "foil pocket and opposing cladding-plate region were roughened using high purity alumina abrasive. After the surface preparation step, the plates were cleaned using a wet acetone/ethanol wipe, dried, and placed in a clean plastic bag.
- Cladding plates were next mounted in a holding fixture and a template/mask aligned and secured over the front face. A 0.025-0.050 mm coating of Si, Si-Al, or aluminum alloy is applied using a Praxair SG-100 plasma spray gun utilizing 30 standard liters per minute (slm) of argon. The coating was formed by traversing the cladding plate in front of the thermal spray plume at 200-350 mm/sec.
- Coated cladding sets are wrapped with lint free clothes and vacuum sealed in plastic bags until needed.

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 involves the use of a water cooled rotating tool configured in a Kearny & Trecker milling machine, Figure 6.

Figure 6. Friction bonding on the K&T milling machine.

A cladding/foil "pack" is prepared for the friction bonding process by placing a fuel foil into the machined "foil retention pocket" and then placing the cladding cover sheet . The cladding/foil lay-up "pack" was then placed on a water cooled anvil, located on the mill table, and clamped firmly in place along each longer edge of the pack.

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 thermo-mechanical softening of the cladding material under the tool.
- Traversing the work piece "clad/foil pack" and Table.
- Stopping the traverse and drawing out the rotating tool from the work piece.
- Returning to the start position and offsetting work piece for the next pass.
- Removing any protruding flash material from the upcoming pass region using a scraping tool.
- Repeating steps above until one side of fuel plate was been friction bonded.
- Sanding and/or scraping protruding flash material from the work piece.
- Flipping the clad/fuel pack over and securing in anvil.
- Repeating the friction bonding process on the second side.

<u>Fuel Plates Fabrication using the Hot Isostatic Pressing (HIP) process</u>

Fuel plate samples are HIP processed in a evacuated stainless steel "can" that can hold up to six miniplate size samples; each being separated by a "strong backs/steel plates" that keep each sample flat during processing. Additionally, grafoil sheets are used to prevent fuel plate samples from sticking to the HIP can and strong backs.

The following steps are conducted for the preparation of a HIP can.

- Chemically cleaned, as described above, aluminum cladding sheets and UMo foils are loaded into a partial HIP can, as shown in Figure 7a.
- The loaded HIP can is clamped and moved to an argon glove box, Figure 7b.
- All edges of the HIP can are edge welded.
- The HIP can is helium leak checked.
- The HIP can/samples as vacuum degassed, using a roughing pump, at 315 ^{o}C for 3-4 hours.
- The stem of the HIP can is crimp welded so as to maintain the established vacuum.
- The HIP can is loaded into the HIP and heated, typically, to 560 ^{o}C and held for a period of 60 minutes at 100 MPa.
- Samples are removed after cutting away the sides to the HIP can.

Prior to final plate processing steps, both friction bonded and HIP processed fuel plates are characterized "qualitatively" for bonding and cladding thickness over the fuel zone region "min clad" using an ultrasonic testing (UT) work station.[6]

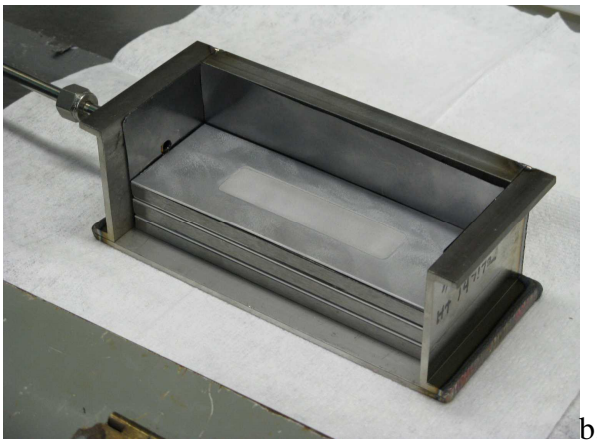
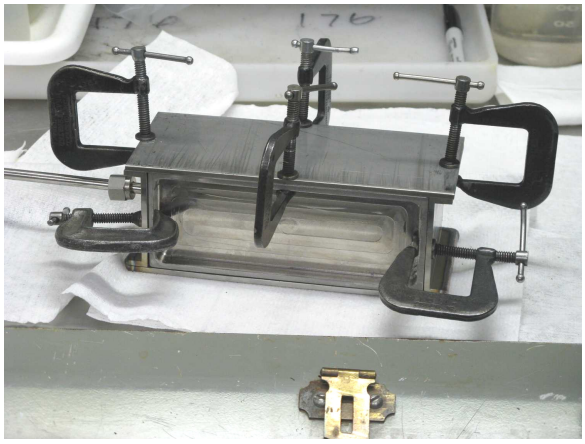
a.  b. 

Figure 7. HIP can preparation: a) HIP can during lay up step, and b) loaded and clamped HIP can ready for welding.

Final Processing Sequence

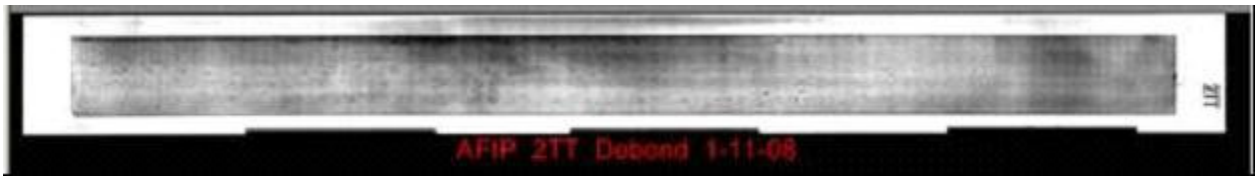
Based on a map of the plate thickness and UT characterization derived cladding thickness data, fuel plates are sanded, on both sides, in order to realize a fuel plate having the desired overall thickness and at least the minimum cladding thickness, 0.15 mm, over the fuel region. Once sanded, the final plate dimensions are established via shearing and final machining operations.

An oxide film "boehmite" is applied to the fuel plate using an autoclave treatment. 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 is removed with cleaning and a like thickness reestablished during autoclaving. For autoclaving, fuel plates are fully immersed in deionized water within the

autoclave, with only their edges in contact with the plate holder. The oxide film is a four hour hold at 185 °C and pressure of ~160 psi (1.1 Mpa).

Following the oxide film treatment, immersion density measurements are obtained in order to obtain fuel plate volume. Subsequently, final dimensional inspection, final UT characterization, and a cleanliness inspection are performed. Figure 9 shows the UT transmission/Debond image of two full size monolithic fuel plates fabricated at the INL for the AFIP-2 experiment; overall plate dimensions, 570 mm x 56 mm x 13 mm.

2BZ



2TT

Figure 8. UT Debond scan images of AFIP-2 fuel plates.

On going Technology Developments

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 shaped geometry ingots, hot rolling of surrogate graded fuel foils, and thermal spray coating of neutron poison materials. Below is a brief discussion of the efforts and results to date.

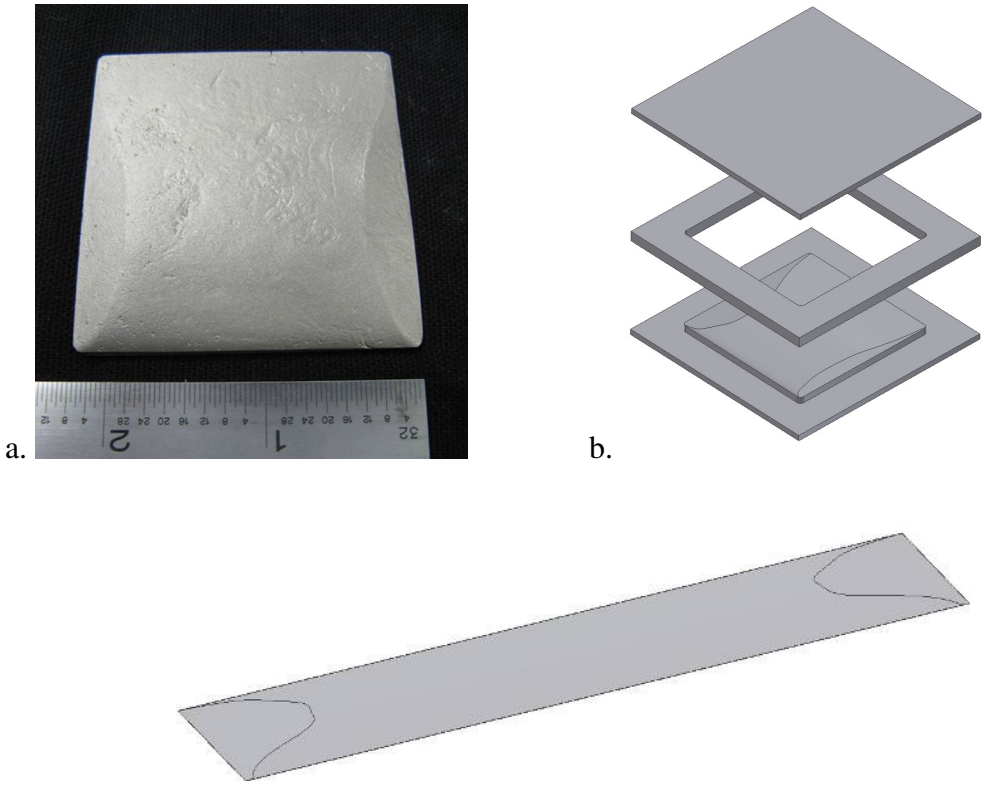
Graded Fuel Foil Development

A graded density dispersion fuel form is employed in the High Flux Isotope Reactor (HFIR). As such, the development of complex/grade monolithic fuel foil is being pursued at the INL.[7] The proof of concept effort, currently being performed using stainless steel, involves induction casting of a complex geometry ingot/coupon that is “canned” and hot rolled to produce a foil have thickness of 0.125-0.500 mm (0.005-0.020”), Figure 9. Full size foils, 570 mm x 56 mm, have been demonstrated using the method.

Thermal Spray Coating Containing Neutron Poison Materials

In the future, tailoring of monolithic fuel plate performance may be possible via selective deposition of burnable poison at the fuel cladding interface. Proof of concept experiments have

been performed at the INL using the thermal spray coating method discussed above. In this case, a mixture of aluminum powder and a burnable poison, i.e.B_4C, ZrB_2, were co-deposited on the fuel contacting regions of prepared cladding sets. Surrogate fuel plate samples were then prepared using the HIP process and with DU10Mo foils, Figure 10. Examination results indicate that in the case of the B_4C containing interlayer, particle size refinement is needed in order to enhance the uniformity of the poison phase.



a. b.

c.

Figure 9. Graded fuel foil fabrication: a) Image of cast, complex geometry, stainless steel coupon/ingot, b) drawing of three piece roll rolling can assembly, and c) drawing of resulting full size contoured foil.

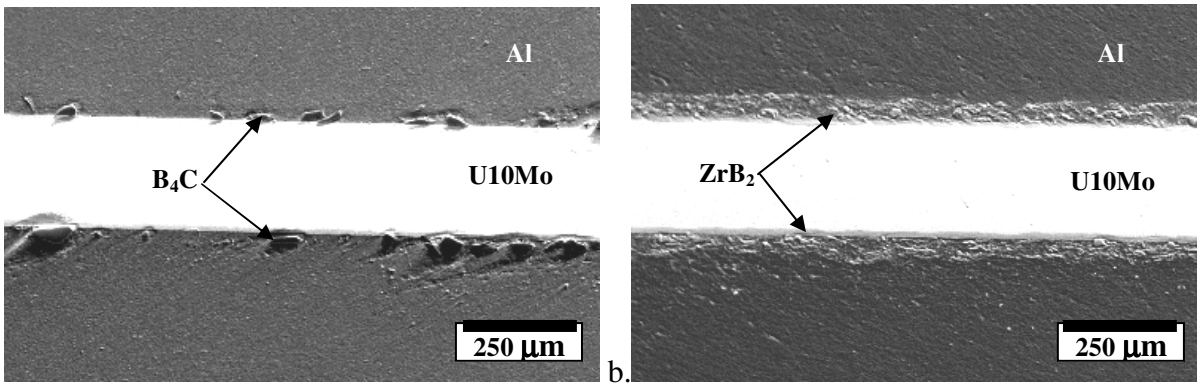


Figure 10. SEM micrographs of fuel plate sample cross sections showing thermal spray deposited interface layer at U10Mo interface: a) B4C/Al coated and b) ZrB2/Al coated.

3. Summary

Significant progress in the fabrication of monolithic fuel foils and fuel plates has been made during 2007 by the INL RERTR Fabrication group. Full size fuel plates incorporating 1) a zirconium barrier layer applied to U10Mo fuel foil and 2) a silicon rich interfacial layer applied to aluminum cladding via thermal spray techniques, have been fabricated using the friction bonding method. During 2007, two full size monolithic LEU fuel plates were irradiated in the INL Advanced Test Reactor (ATR) as part of the AFIP-2 (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.

References

[1] 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

[2] 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

[3] 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

[4] 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.

[5] 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.

[6] 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.

[7] 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.