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Update On Monolithic Fuel Fabrication Development

27th International Meeting on Reduced Enrichment for Research and Test Reactors (RERTR)

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ABSTRACT

Efforts to develop a viable monolithic research reactor fuel plate have continued at Idaho National Laboratory. These efforts have concentrated on both fabrication process refinement and scale-up to produce full sized fuel plates. Advancements have been made in the production of U-Mo foil including full sized foils. Progress has also been made in the friction stir welding and transient liquid phase bonding fabrication processes resulting in better bonding, more stable processes and the ability to fabricate larger fuel plates.

Introduction

The Reduced Enrichment for Research and Test Reactors (RERTR) advanced fuel development program was reinitiated by the United States Department of Energy in the mid 1990's with the goal of developing fuel that will allow conversion to low enriched uranium (LEU) of the remaining research reactors which have fissile atom density requirements too high to be met by existing fuel types[1].

The primary focus of the RERTR US development effort has been shifted from the standard dispersion fuel to monolithic fuel, where the fuel meat is composed of a single piece of fuel in the form of a thin foil. Monolithic fuel is currently being tested in the Advance Test Reactor (ATR) in Idaho[2].

Unlike dispersion fuel plates, which have been fabricated for decades using the roll bonding technique[3], monolithic fuel requires a new fabrication method to achieve bonding while leaving the fuel foil intact. The three methods that have been pursued at the advance fuel development laboratory at the Idaho National Laboratory[*] (INL) have all undergone continued development: friction stir welding, transient liquid phase bonding and hot isostatic pressing. These efforts cover both process testing/improvement and also scale-up to allow production of full sized fuel plate fabrication. The US RERTR effort is also working on fabrication development of U-Mo foil of sufficient size to provide feedstock for the full-sized fuel plates.

In addition to monolithic fuel, INL is also pursuing magnesium matrix dispersion fuel. For this fuel type the aluminum matrix (which has shown detrimental interaction with the dispersed U-Mo fuel particles under certain irradiation conditions) is replaced by magnesium (which does not react with metallic uranium). This fuel fabrication development effort is pursuing a hybrid process to fabricate the Mg matrix fuel. A friction stir welding process is used to bond the fuel

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[*] The former Argonne National Laboratory-West was, in February 2005, combined with research elements of the former Idaho National Engineering and Environmental Laboratory to form the new Idaho National Laboratory.

compact into an aluminum can which is hot rolled to final thickness.

Friction Stir Welding

Friction Stir Welding (FSW), a solid state mechanical action bonding process, was developed in Great Britain in the early 1990's.[4, 5] This process is commonly used to weld butt or lap joints in aluminum alloys and was modified by the RERTR project to encase a fuel foil inside an aluminum plate.[6] This process has undergone a number of changes and improvements since work was begun in 2003. This method is, to date, the most promising of the three targeted monolithic fabrication processes.

FSW employs a rotating tool comprised of a small diameter pin mounted concentrically below a larger shoulder. A conventional milling machine is used to rotate the tool and force it into the surface of the metal being processed. The heat and pressure generated by the tool contact induce plastic deformation in the region near the pin. The tool is plunged into the material until the shoulder rests on the surface. The shoulder contact serves to control the depth of the weld, to keep the process material from migrating away from the process area and to give added heat and pressure. Movement of the pin through the process piece forms a weld bead. The process is repeated with overlapping welds to cover the entire area of the fuel plate. The plate is then turned over and the FSW process is again performed to bond the fuel foil to the bottom cladding plate (Figure 1).

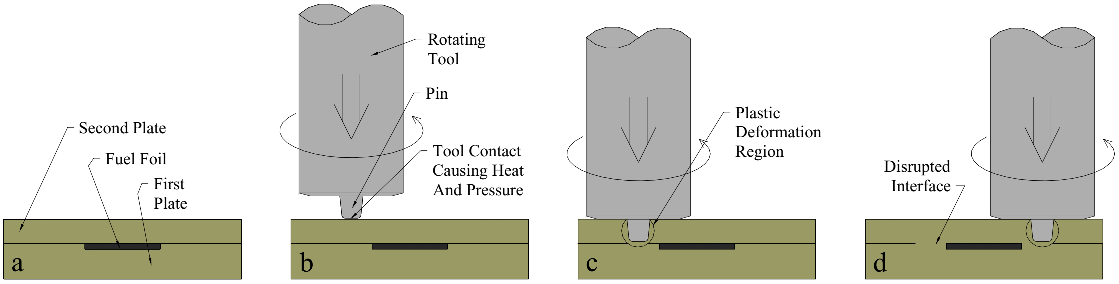

Figure 1. Friction stir welding schematic cutaway (vertical scale is exaggerated to show detail). a) Shows the plate assembly for FSW of monolithic plates. In b) the rotating pin contacts the surface of the plate causing heat and pressure. In c) the tool has been plunged into the surface of the material up to the shoulder of the tool. The rotating tool is stirring the metal in the plastic deformation region. In d) the tool has been dragged across the weld plane causing a disrupted interface and bonding to occur between the plates and between the cladding and the foil.

The RERTR-6 miniplate irradiation test inserted into the Advance Test Reactor (ATR) in April of 2005 contained 12 FSW monolithic fuel miniplates. To date, the experiment has been irradiated to over 20% U^{235} burnup with no indication of problems.

The geometry of the fuel plates is the overriding factor in dictating FSW parameters. The limited thickness of the assembled fuel plates (~1.47 mm to obtain a 1.40 mm final thickness) facilitates rapid heat transfer through the thickness of the plate and exacerbates any temperature excursions. A high process temperature can result in buckling of the assembly (aided by the thin plate geometry) or an excessive plunge of the tool into the assembly—which results in disruption of the fuel foil and a failed plate.

Initially, the temperature was visually gauged by watching the quality of the weld. To compensate for temperature variances, the operator was required to slow down the horizontal feed or repeat a pass to heat up the weld and speed up the horizontal feed or stop the weld and allow the equipment to sit idle to cool down the weld.

To mitigate the temperature excursions, a cooled tool holder was designed and built by INL staff. A recirculating chiller is used to pump an ethylene glycol-water mix through the tool (including a plenum behind the tool face). A thermocouple (TC), fed into the cooling plenum is used to monitor the process temperature (as the TC is immersed in the coolant flow, the reading is not an accurate indication of the welding temperature but does serve as an effective empirical gauge) (Figure 2).

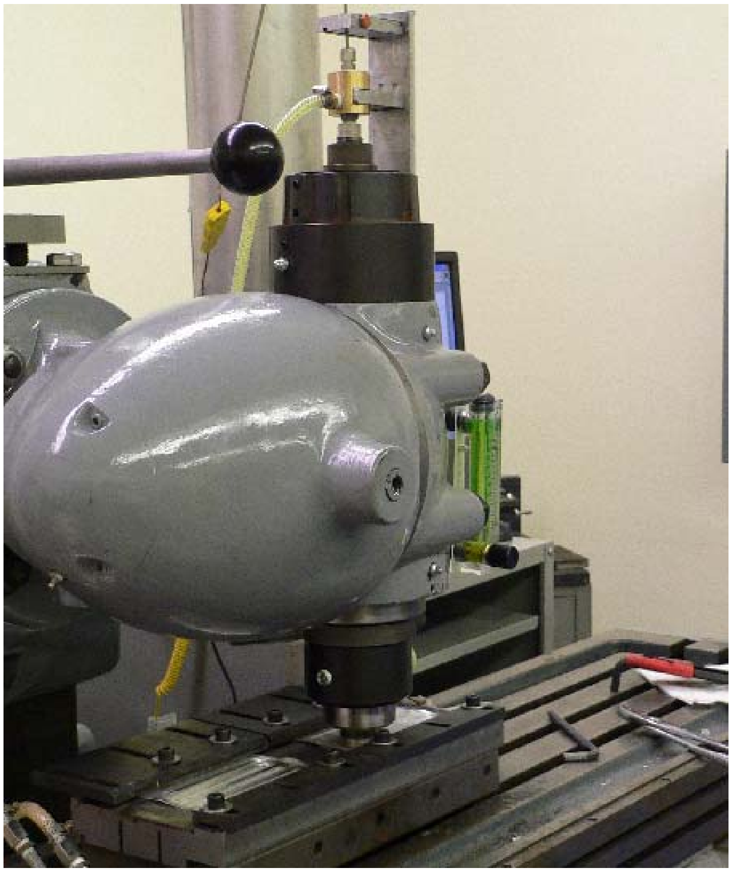

Figure 2. Kearney & Trecker friction stir welding setup. The cooled tool holder is the collar around the tool (cooling can be adjusted by a flow meter). Process temperature is measured by the thermocouple entering the top of the spindle.

Using the cooled tool holder system with a 1.3 cm diameter tool pin, the optimum processing temperature (as measured by the embedded TC) was found to be 120-125ºC. This resulted in a plate fully bonded across the fuel region.

Efforts over the past year have resulted in several improvements in the tool design. The small geometry (6.4 mm) tool used for the fabrication of RERTR-6 has been replaced by a larger diameter pin (12.7 mm). Advancements have also been made in the weld pattern. For the

RERTR-6 experiment, a one-way raster was performed (Figure 3) that covers the plate with a series of parallel welds running in the same direction. This method required a plunge and an extraction for every welding pass and added time for the tool to travel back across the assembly. The plunge is the most time consuming portion of the weld (a dwell time is needed during the plunge to allow the weld area to raise to the proper temperature). This pattern has been replaced by a true raster. The raster speeds up the FSW process by eliminating all but one of the plunges and reducing the travel time for the one-way pass to return to the starting point.

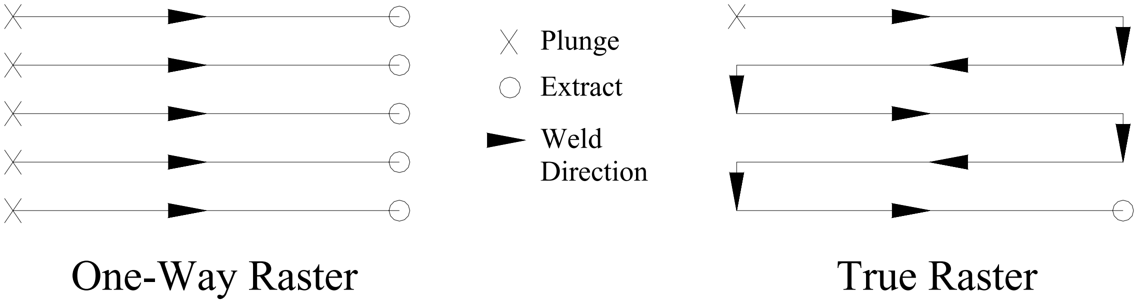

Figure 3. Weld patterns used for FSW of fuel plates.

The changes outlined here have led to dramatic improvements in bond quality. The ultrasonic scans of these fuel plates show a distinct lessening of debonded areas (Figure 4).

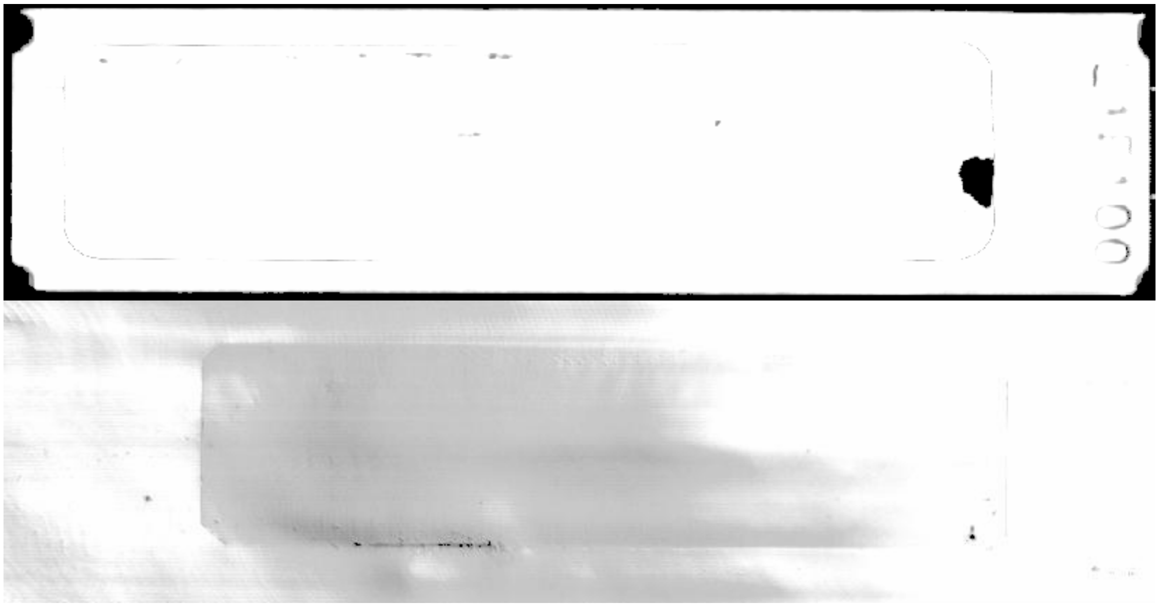

Figure 4. Ultrasonic scans of friction stir welded plates. At top is a plate included in the RERTR-6 irradiation test (note the region of debond on the fuel zone). At bottom shows a foil bearing plate fabricated more recently. The debond areas in the bottom plate are much smaller, less severe and not over the bulk of the fuel meat.

One of the major factors in development of a new research reactor fuel fabrication technique is the ability to scale this method to enable commercially viable fuel production. The RERTR-6 fuel plates were "miniplates" measuring 2.54 by 10 cm. A process capable of fabricating full sized research reactor fuel plate is required to convert any reactor.

Many of the process modifications listed above have the dual advantage of providing both process improvement and allowing for better scale-up of the FSW process. While the FSW method used to produce the RERTR-6 experiment could be used to fabricate a full sized fuel plate, the time required would be prohibitive. Significant improvements have been made in weld area speed and in duty cycle of the FSW process. The parameters used for the RERTR-6 experiment give a weld coverage of 4 mm^2/s; this has since been improved to over 18 mm^2/s by using a larger pin diameter and faster table feed. This speed advantage does not take into account the time savings a true raster weld pattern gives in comparison to the one-way raster. The cooled tool holder keeps the welding surface of the tool from overheating, allowing the welding to continue nearly indefinitely while the older setup required the process to be halted periodically to allow the temperature of the tool to drop.

To aid in FSW scale-up and process control a purpose-built FSW machine has been procured and is being installed at INL. This machine is computer controlled and will make the FSW process much less operator intensive. Unlike the old equipment at INL the new machine will allow fabrication of fuel plates for any reactor (the ATR core measures 1.22 m in length). In addition, the machine will provide in-process data acquisition to properly characterize the effect of welding parameters on bond quality and fuel performance as well as provide a higher level of quality control. It is anticipated that this machine will be operational by mid November of this year.

Transient Liquid Phase Bonding

Transient liquid phase bonding (TLPB) is the second candidate for cladding monolithic fuel plates. TLPB relies on a eutectic forming interlayer material to diffuse into the bonding interfaces and join the materials together. By application of a suitable material between the cladding plates, a eutectic liquid phase is formed in this interface. This temporary liquid phase spreads across the interface, acts as a flux to dissolve the oxide surface layer on the aluminum, diffuses into the cladding and forms a metallurgical bond, joining the two plates together. TLPB with silicon has been used previously to fabricate fuel plates[7]. Silicon and aluminum form a eutectic at 577ºC, significantly lower than the melting point of aluminum (660ºC).

As the TLPB process was originally implemented, silicon powder was blended with a mixture of ethanol and glycerin to facilitate application of the TLPB interlayer. A thin film of this Si “paint” was applied to the aluminum plates (the aluminum plates were mechanically cleaned), allowed to air dry, and then the glycerin “burned off” using a hot plate. The aluminum plates were assembled with the silicon powder in the interface. The assembly was loaded into a hot press and heated to 590ºC under a load of 6.9 MPa. The temperature and pressure were maintained above the eutectic temperature for 30 minutes. The plate was then cooled below the eutectic temperature and removed.

Despite initial promise, the process outlined above was ultimately unsuccessful. TLPB bonded plates were slated for inclusion in the RERTR-6 test matrix but had to be removed after serious problems with bond integrity (Figure 5).

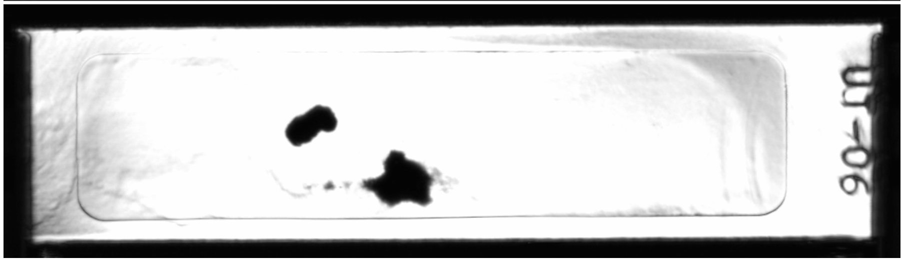

Figure 5. Early TLPB U-Mo plate. The black regions indicate debonded areas.

In order to improve the bonding, a study was conducted to improve the fabrication process. The major areas of development were aluminum cleaning and silicon application.

Originally, the plates were cleaned prior to silicon application by a rotating ScotchBrite pad to remove gross impurities from the surface. This method has been abandoned due to the plastic residue ScotchBrite leaves on a cleaned surface. The rotating pad was replaced by a rotating stainless steel brush. This was tested and showed some promise but was ultimately supplanted by a more rigorous cleaning method.

To obtain an even cleaner aluminum surface, a chemical cleaning procedure has been employed after the mechanical brushing. This cleaning procedure is taken directly from standard fabrication practices for roll bonding aluminum research reactor fuel plates[3]. After cleaning, the plates are to have the silicon applied and undergo TLPB within 48 hrs.

The original Si application method (outlined above) led to non-uniform powder application and the compositional quality of the powder after organic burnout was questionable. The new approach is to thermally spray the silicon powder (99.9% pure) directly to the mating surfaces of the aluminum. The silicon is applied with a SG 100 gas torch located at the INL (Figure 6).

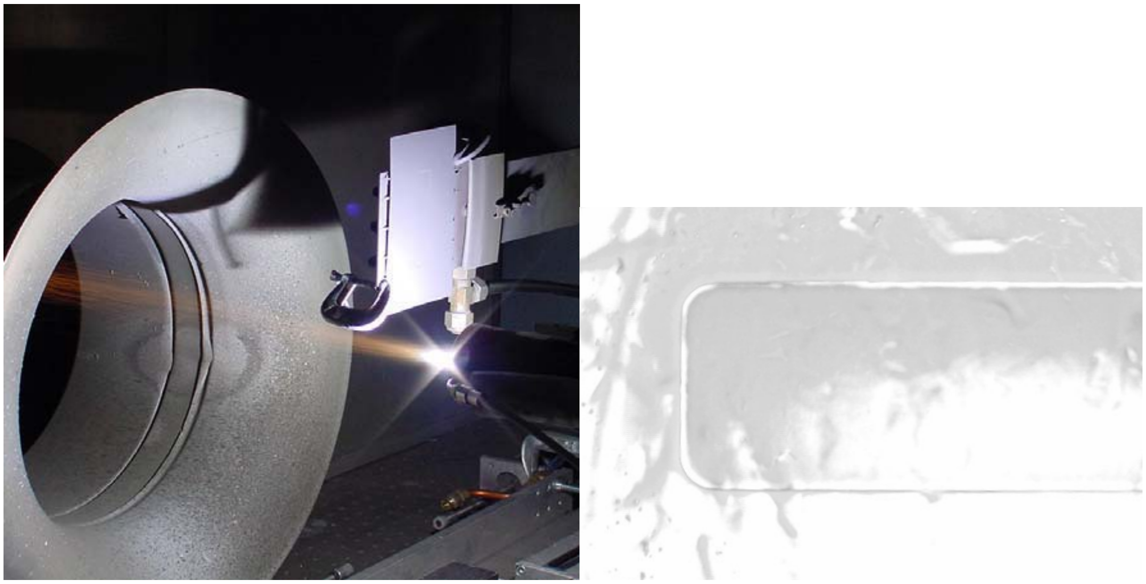

Figure 6. l) thermal spray process used for Si application. r) Close-up of ultrasonic scan of TLPB bonded plate with U-10Mo foil.b

These process improvements have led to full bonding of U-Mo fuel plates (Figure 6-r). Examination of the fuel-cladding interface (the older TLBP method showed significant formation of a brittle reaction layer) is underway but results were unavailable for this paper.

Hot Isostatic Pressing

Hot isostatic pressing (HIP) was invented by the Battelle Institute in the 1950s in order to perform diffusion bonding of Zircaloy cladding to uranium oxide nuclear fuel. HIP uses high temperature and isostatic pressure simultaneously to bond similar or different materials, which are otherwise difficult to bond[8]. Diffusion bonding by HIP is one of the monolithic bonding methods being investigated at INL.

Since no appropriate domestic hot isostatic press was available (being either non-operational or not able to handle radioactive materials) this method has yet to be tested with a U-Mo foil. A surrogate test was performed using stainless steel surrogate fuel foils (This study was conducted by Bodycote under a subcontract for BWXT). This work showed that the aluminum-aluminum bonding could be achieved in Al 6061 cladding material.

Recently, a reconditioned Eagle 8 Hot Isostatic Press (Figure 7) has been purchased from American Isostatic Press and is being installed at INL. After the installation is complete, the instrument will be used to conduct studies to ensure proper aluminum-aluminum and fuel-aluminum bonding.

Figure 7. The Hot Isostatic Press being installed at the Idaho National Laboratory.

Given the thermal cycle required by the HIP process, the reaction between uranium and aluminum is a primary concern[9, 10, 11]. Significant reaction between the stainless steel and aluminum cladding was also observed in the previous HIP experiment. In order to address the concern on the reaction between the U-Mo fuel foil and aluminum cladding, a diffusion-bonding experiment was carried out using a uniaxial hot press (which approximates isostatic conditions for the plate geometry of research reactor fuel). Aluminum foils (6061-aluminum) and U-10Mo foils were used for these experiments. A niobium foil with a thickness of 25µ was introduced between the U-10Mo foil and the aluminum as a diffusion barrier. The hot press experiment was conducted under similar conditions to those performed in the surrogate foil HIP test.

Figure 8 is a back-scattered scanning electron micrograph showing the reaction layers found in the hot pressing experiment. The bonding between the U-10Mo and aluminum has some void regions and a reaction zone up to 20 µm was observed. Since the reaction layer between the U-10Mo and aluminum is expected to be an order of magnitude thicker at this temperature, more experiments are underway to verify this result. The bonding between aluminum and niobium is very good with no apparent reaction layer between them. The bonding between the U-10Mo and niobium is not very good as large areas of voids are apparent. A better surface cleaning technique is being worked on to improve the bonding.

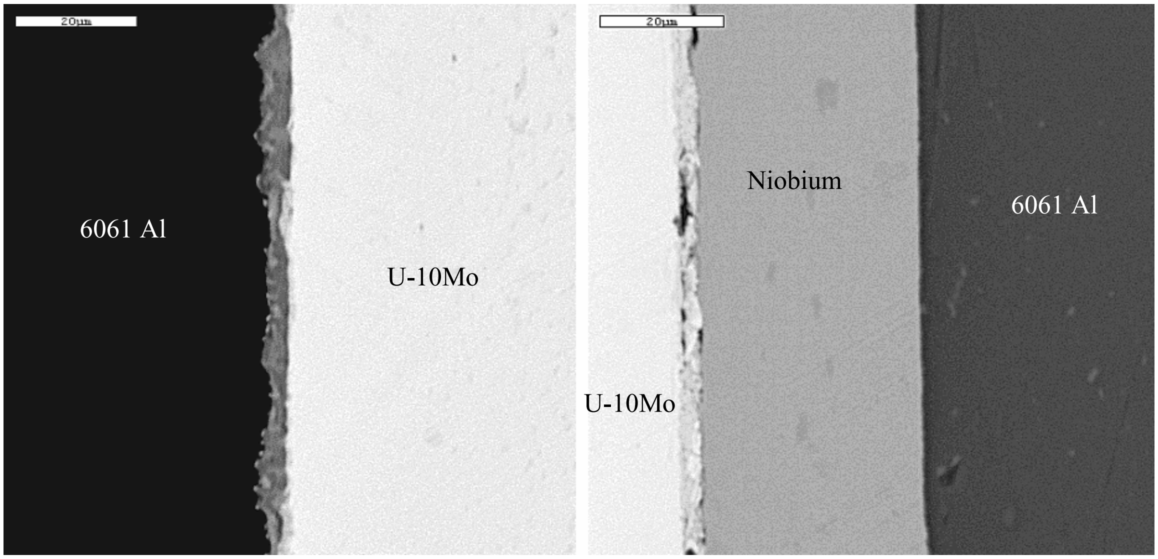

Figure 8. Pseudo-HIP back-scattered electron micrographs. left) 6061 Al/U-10Mo interface showing interaction region. Nb foil showing interaction layer with U-10Mo and clean interface with Al.

Foil Production

In order to meet the needs of upcoming full sized fuel plate fabrication campaigns, the existing foil production method[6] is being scaled up. Foil widths of up to 10 cm are being fabricated to obtain foils of the correct size.

Arc melting is used to provide the molten material for the casting. The melt is bottom poured from the crucible into a mold. The mold consists of two copper plates clamped together on either side of a U-shaped insert (also copper). The insert determines the final size of the coupon and can be changed out as different sized coupons are needed. The tops of the cover plates are flared outward to funnel the material into the mold. Using this arrangement U-10Mo has been cast into coupons (Figure 9-left).

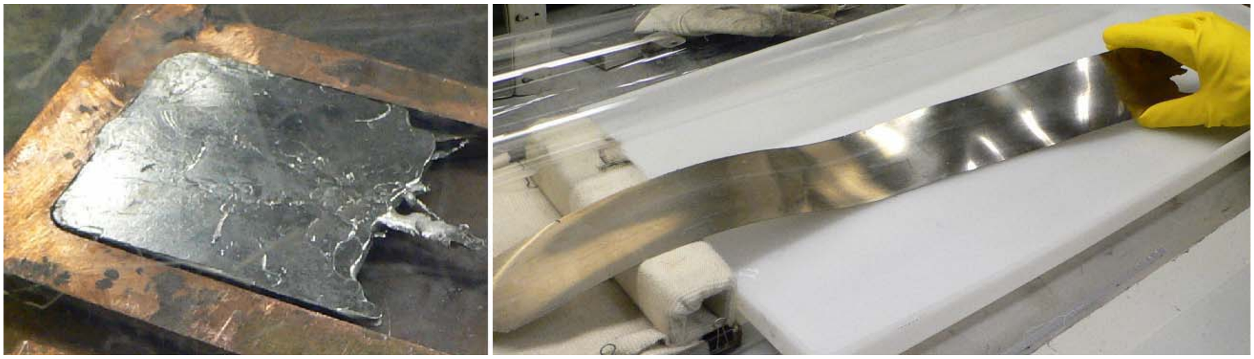

Figure 9. Cast coupon (left) and rolled U-10Mo foil.

While the jeweler's mill served to make foils suitable for miniplate sized fuel plates, the nature of this equipment dictates a practical limit of ~2.5 cm in width. This limit is imposed by the clampdown mechanism, the lack of bearings, the diameter of the rolls, and the absence of a drive motor.

A large 4-high rolling mill has been procured and installed at INL. The smaller roll diameter of the work rolls allows thinner stock to be rolled than with a standard 2-high mill. The foil is produced by rolling the bare coupon through the rolls. A hood-like housing was built around the entrance and exit side of the rolls. This Lexan box allows for handling of the foil at either side of the mill and for the foil to be passed from the exit side to the entrance side without leaving the containment. The box is connected to a ventilation system to insure net airflow into the containment (Figure 10). This mill has produced rolled U-10Mo foils over 10 cm in with.

Figure 10. Foil rolling mill. The containment box is shown on either side of the rolling mill.

As the cold worked foil will crack within a matter of hours after rolling to a 90% reduction, an annealing step is required that can be applied quickly while minimizing oxide formation. The resistance annealing method[6] as performed on the miniplate sized foils used for RERTR-6, has proven to be a suitable process in every regard. As this process was limited in size to foils less than 2.5 cm in width, a scale up of the equipment was undertaken. This scale up consists of a larger process tube (10 cm diameter by 100 cm in length) and a larger power supply (600 amp).

Magnesium Matrix Dispersion Fuel Fabrication

A primary limitation associated with the U-Mo dispersion fuel is the interaction formed between the fuel particles and the aluminum matrix. This interaction is seen as the precursor to void formation, coalescence and pillowing under certain irradiation conditions. One solution to the U-Mo/matrix Al interaction is to replace the aluminum matrix material with magnesium[12]. The US RERTR program irradiated one fuel plate containing a Mg matrix in the RERTR-3 experiment with promising results. The primary difficulty in fabrication of this fuel type is the need to maintain process-temperatures below 437ºC, the aluminum-magnesium eutectic temperature.

This temperature constraint necessitates the use of an alternative process to hot rolling; the process traditionally used for bonding aluminum-matrix dispersion fuel plate. The hybrid processing approach being pursued involves: 1) encapsulation of a Mg-matrix/U-Mo powder compact in a 6061 aluminum frame via friction stir welding, 2) warm rolling of the assembly to obtain the desired RERTR mini-plate test specimen[13]. Testing is done with the RERTR 2.5 x 10 cm miniplate specimens.

For the process development phase of this effort, spherodial or plasma-spherodized tungsten power is being utilized as the U-Mo fuel surrogate. The effort has been partitioned into the following elements: 1) compact processing, 2) friction stir welding (FSW) development, and 3) warm-rolling. At this time, compact processing and FSW experiments have been conducted.

Powder compacts, having ~85-90%, theoretical density are prepared in a uniaxial press using Mg and spherical tungsten powder; the tungsten volume fraction is adjusted to reflect a fuel loading of ~7 gU/cc with a U-6Mo alloy. This pellet is encapsulated in a two layer assembly. Unlike the standard dispersion fuel assembly where a “picture frame” is sandwiched between two cover plates, a single cover plate is used in the hybrid process (Figure 11) to reduce the number of bonding interfaces from two to one.

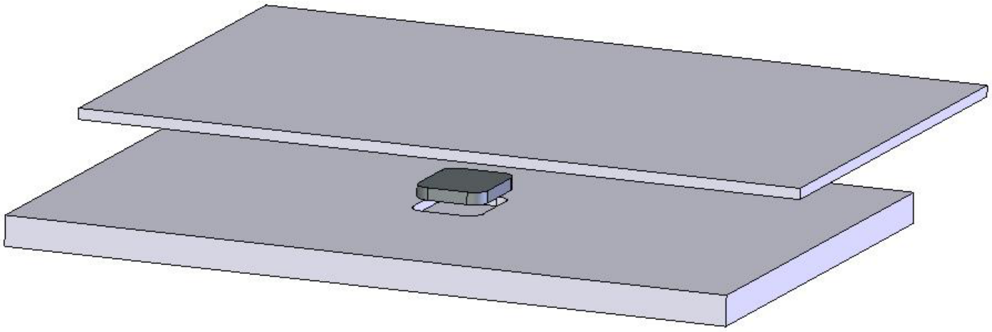

Figure 11. Mg-matrix dispersion fuel pellet assembly construction.

After the compacts are placed in the cladding assembly the compacts are sealed into the plates by FSW. The FSW process is of interest here because bonding is accomplished without any contribution from the rolling. Thus the rolling process, usually focused on obtaining bonding of the aluminum assembly, can instead be used to ensure the proper plate thickness and fuel meat homogeneity is obtained. The FSW processing takes place at temperatures below the Al-Mg reaction temperature (437ºC) Thus, the opportunity exists to produce the needed aluminum-aluminum clad-bonding without Mg-Al eutectic phase formation in the fuel plate assembly.

The FSW performed on the assembly was done by a computer numerical control (CNC) milling machine which allows a continuous lap-weld around the perimeter of Mg-matrix/W-dispersion pellet. This weld is performed with sufficient offset from the compact so as to not disrupt the fuel zone. Tool temperature measured during the operation was ~340-350ºC.

Radiography of the FSW assembly established that the encapsulated pellets remained undamaged during the welding process. Ultrasonic testing (UT) was also conducted in order to establish/confirm lap-weld integrity. Figure 12 r shows the recorded UT image. The UT analysis indicates near complete bonding along the FSW path. Discontinuities in the UT image are consistent with the location of surface flash features (yet still show an acceptable bond).

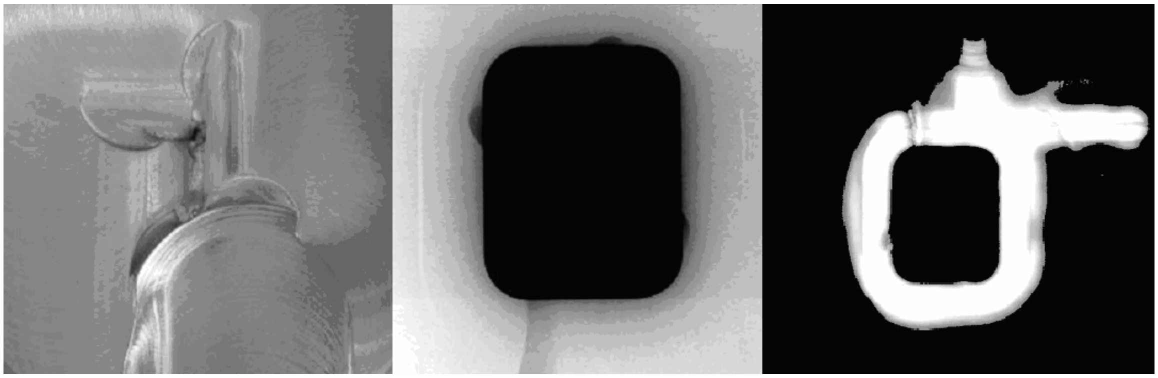

Figure 12. Friction stir welded Mg-matrix assembly (images are not to scale). l) optical image of the FSW surface. m) radiograph of fuel zone showing no disturbance of fuel meat. r) Ultrasonic scan of bonded assembly (darker regions in the weld correspond with FSW surface features).

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