MONOLITHIC FUEL PLATE DEVELOPMENT AT ARGONNE NATIONAL LABORATORY

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ABSTRACT

Initial fabrication results of U-Mo dispersion fuel have shown a practical uranium loading limit below that required to successfully convert some of the targeted research reactors to low-enriched uranium. In addition, irradiation tests have shown that U-Mo dispersion fuel suffers from interaction between the fuel and the aluminum matrix at higher temperatures. A potential solution to mitigate these phenomena is to use a fuel alloy foil in place of the fuel-aluminum dispersion. This monolithic fuel provides a lower fuel-matrix interfacial surface area and a much higher uranium density than dispersion type fuel. Lacking the compliant matrix of dispersion fuel types, monolithic fuel production requires the development of new fabrication methods. Research efforts at Argonne National Laboratory have resulted in the demonstration of a viable monolithic fuel plate production method, which is described in this paper.

1. Introduction

The Reduced Enrichment for Research and Test Reactors (RERTR) advanced fuel development program was reinitiated in the mid 1990's with the goal of developing a fuel type 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. In addition, uncertainties in the ability to reprocess existing U_3Si_2 fuel have led to demand for a new fuel type [1].

Uranium-molybdenum alloy fuel dispersed in an aluminum matrix was identified as a potential fuel type. Early fuel performance tests have been promising, but achievable uranium fuel densities have been lower than needed to convert several research reactors to LEU. In addition, irradiation tests have shown that fuel operating at higher temperatures and powers cause the U-Mo fuel powder in the meat to react with and almost completely deplete the aluminum in the matrix [2].

It is anticipated that replacing dispersion fuel with a monolithic fuel type can solve both the U-Al interaction and fuel density loading problems. In monolithic fuel the entirety of the fuel meat is comprised of a single foil of the fuel alloy. This fuel type represents the optimum in fuel meat density. The greatly reduced fuel surface area per unit mass and the fact that fuel-aluminum interfaces are in the cooler region of the plates should minimize the fuel-aluminum reaction.

2. Past Results

Previous irradiation experiments have been performed using a U-Mo foil in place of the fuel-matrix dispersion. As part of the RERTR-4 irradiation test, two U-10Mo monolithic plates were irradiated to greater than 70% U^{235} burnup in positions A-6 and 4-C [3].

U-Mo foil for the monolithic test plates was produced by hot rolling a U-10Mo ingot. This ingot was sealed in a steel can to isolate the uranium from the atmosphere during processing. The ingot pack was repeatedly heated to 650°C and rolled at temperature to reduce the thickness of the fuel alloy. During processing the steel can was rewelded or replaced as needed to maintain cladding integrity. Using hot rolling the thickness of the fuel alloy was reduced from 6.2 mm to 0.64 mm. Final thickness reduction to 0.36 mm was achieved by cold rolling with the foil sandwiched in an unwelded steel envelope.

The actual test plates were fabricated by application of the existing roll bonding method used for dispersion fuel plate production. The foil was placed in a thin "picture frame," sandwiched between two aluminum cover plates and hot rolled at 494ºC a total of 85% reduction. This reduction resulted in the tearing of the fuel foil inside of the plate (Figure 1-left). Bonding was marginal with only two of the five plates produced by this method passing the bonding standards for irradiation testing [4].

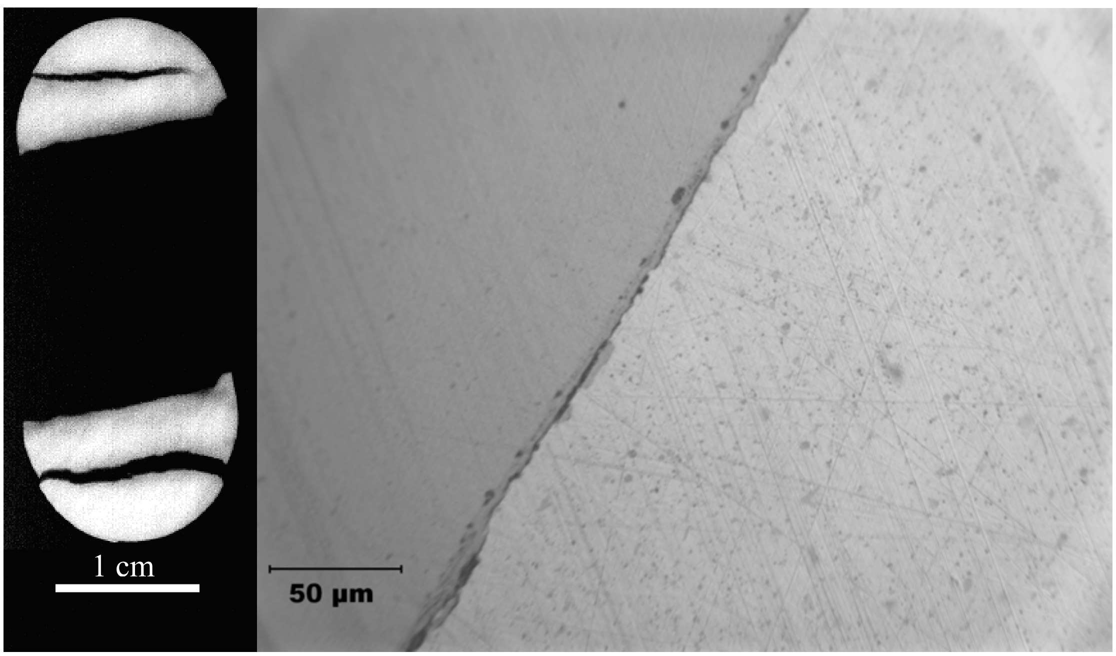

Figure 1. Monolithic fuel plate. Left—radiograph showing tearing of the as-fabricated foil. Right—micrograph of irradiated plate (foil on left) showing uniform irradiation damage and minimal fuel/cladding interaction.

Initial examination of the RERTR-4 test has given promising irradiation results for the monolithic fuel type. There is only minimal interaction with the cladding. The fission damage was uniform with only small gas bubbles formed in the bulk of the fuel (figure 1-right). Additional testing is required to better ascertain fuel irradiation performance and to demonstrate cladding adhesion to the fuel foil using more prototypic plates. The work outlined in this section has been used as a springboard for work at Argonne-West to improve fabrication methods geared toward production of experimental fuel plates for an upcoming irradiation test.

3. Foil Production

The U-Mo fuel foil for the previous monolithic tests were produced by hot rolling in a process (outlined above) that was both time consuming and expensive in terms of manpower and generated waste. Efforts have been undertaken to simplify the process using existing equipment (in the Argonne fuel development laboratory) or modifying existing equipment.

The material for each fuel plate foil is cast individually. The constituent uranium and molybdenum are massed and placed into an arc-melter. This charge is melted to consolidate the ingot then alternately turned over and arc-melted a number of times to homogenize the alloy.

To aid in rolling the sample to the desired foil thickness, it is cast in a coupon 2.0 mm (0.080 in) thick. This casting is performed in the arc-melting furnace using a gravity pour into a chill mold (Figure 2).

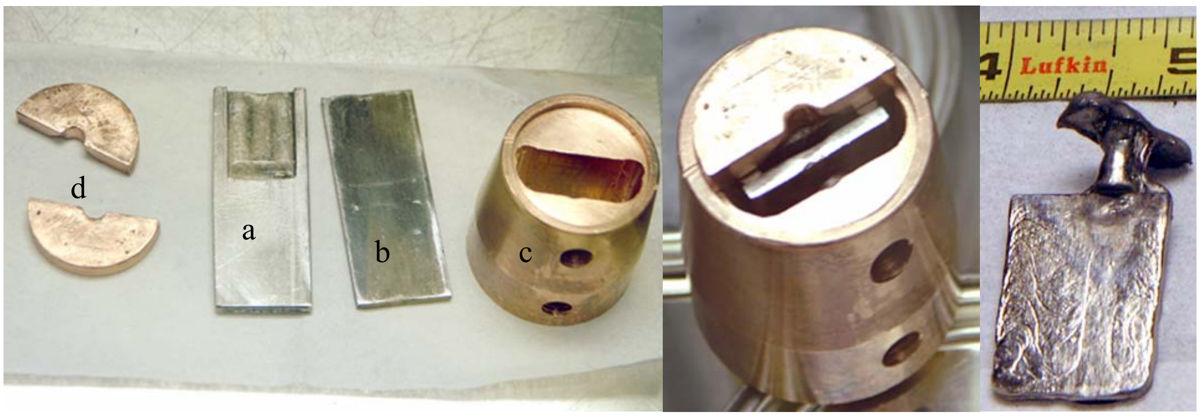

Figure 2. Arc-Casting Hearth. To cast U-Mo alloys into coupons for further processing the arc-casting hearth is used. The disassembled hearth is shown at left. The coupon mold (a) and cover plate (b) are held together by set screws in the copper hearth (c). The split copper melting platform (d) is positioned over the mold cavity. The assembled hearth (middle) is shown with one segment of the split copper melting platform removed to expose the mold cavity. The resulting coupon is shown at right. The sprue (at the top of the coupon) is removed prior to further processing.

Prior to rolling the sprue is removed from the casting and the coupon is annealed in an inert atmosphere to homogenize the microstructure. Because the coupon is thin it can be easily cold rolled by means of a small, slightly modified jeweler's rolling mill (see figure 3). Multiple passes are used to process the foil to a uniform desired thickness down to 0.13 mm (0.005 in).

Figure 3. Foil processing rolling mill. A small jeweler's mill is used to roll U-Mo coupons into thin foils.

If not subjected to a post-reduction anneal, the residual stresses in the as-rolled foil will cause it to crack in a matter of a few hours, rendering the foil useless (Figure 4). Annealing has been accomplished by an electric resistance apparatus (Figure 5). This apparatus consists of a standard welding power supply, which is used to drive a current through the foil that is to be annealed. The foil is contained in an inert atmosphere of flowing argon. The argon protects the heated foil from exposure to air during processing and quenches the foil after the annealing. The foil is heated in a matter of seconds to the target annealing temperature of 950°C. After annealing the foil is rapidly cooled back to room temperature.

Figure 4. Cracked U-6Mo foil. Residual stresses from the cold rolling process cause the foil to crack within hours of the rolling process if not annealed.

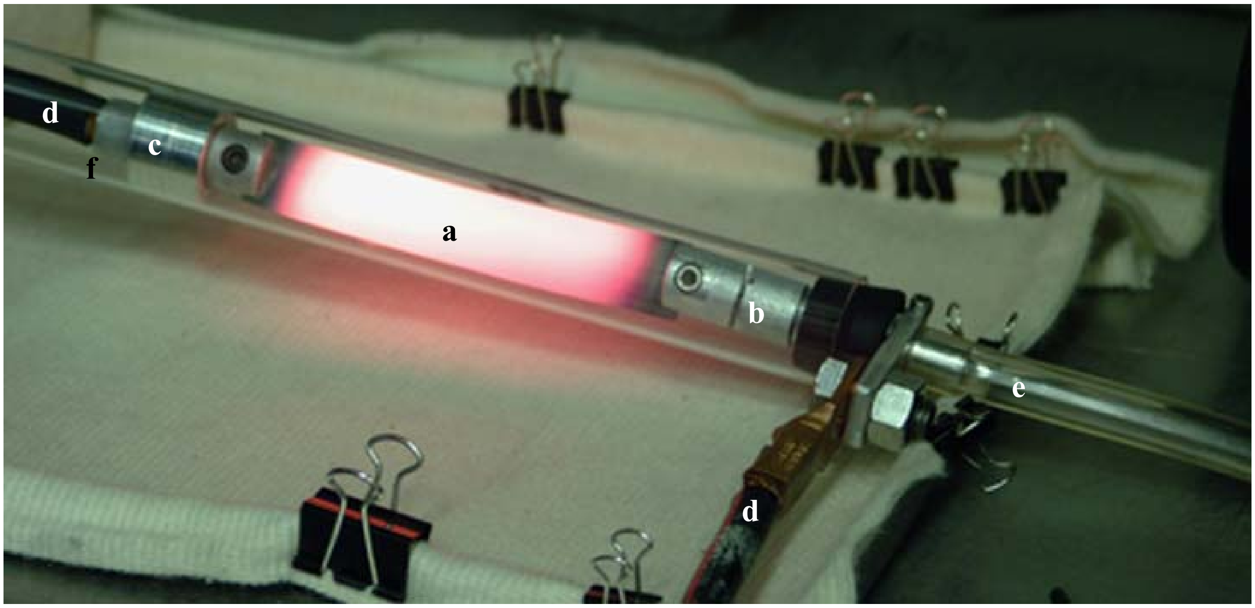

Figure 5. Resistance Annealing Apparatus. The foil (a) is held by two clamps (b) and (c). Electric current is supplied through the clamps via the leads (d). An inert atmosphere is maintained by flowing argon (e) through a clamp (b) and into a fused quartz process tube (f).

4. Bonding Methods

As stated in the previous section, fabrication of the monolithic fuel plate tested in the RERTR-4 experiment used a modification of the standard dispersion plate roll-bonding method. The intermittent bonding and the tearing of the fuel foil using this method make it unusable for future monolithic work. To overcome these difficulties, different bonding methods have been examined.

Ideally, the bonding method would impart little or no reduction into the plate assembly to avoid tearing of the fuel foil. It must achieve good adhesion both in the aluminum-aluminum bond and in the cladding-fuel bond. It would also be a process that can be implemented with as little complexity and capital outlay as possible. Different bonding methods have been investigated for suitability in the monolithic fuel fabrication process. High temperature rolling, transient liquid phase bonding, diffusion bonding, and friction stir welding were all examined. The bonding methods were studied using Al-6061, an alloy that is prevalent in US research reactor fuel plates.

High temperature rolling is similar to the dispersion bonding process with minor adjustments. To date dispersion fuel plate rolling temperatures have been limited to ~500ºC. Higher temperatures result in excessive reaction between the matrix and fuel powders. It was thought that the lower total surface of the foil would prevent problematic interaction and the higher temperature might enhance diffusion enough to get adequate bonding with a lower rolling reduction.

Several experiments were performed using total thickness reductions of 5 to 30% with temperatures up to 630°C. These preliminary tests were done using only aluminum with no foil present. Bonding in these experiments was sporadic at best. Oxidation of the cladding surfaces at the high temperatures appeared to be the limiting factor. In addition, the aluminum alloy (Al-6061 is an age-hardening alloy, temperatures above ~500°C will overage the material; ductility can be restored by proper heat treatments) was embrittled by overaging at the processing temperature. High temperature roll bonding was abandoned once other bonding methods were shown to be more promising.

Diffusion bonding occurs when two objects are heated under pressure and diffusion takes place across the interface. An interlayer of a different material is often placed in the interface to aid bonding. Due to the presence of a strong, easily formed oxide layer, aluminum-aluminum diffusion bonding is difficult to obtain without an interlayer [5]. Molybdenum and copper were investigated as interlayer materials; neither test proved successful. Relevant literature suggests that processing the material in either a vacuum or a reducing atmosphere is needed to obtain an acceptable bond with aluminum. Thus, diffusion bonding was also abandoned as a target bonding method.

Transient liquid phase bonding (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, 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 [6]. Silicon and aluminum form a eutectic at 573ºC, significantly lower than the melting point of aluminum (660ºC).

Silicon powder is blended with a mixture of ethanol and glycerin to facilitate application of the TLPB interlayer. A thin film of this Si "paint" is applied, allowed to air dry, and then the glycerin is "burned out" using a hot plate. The aluminum plates are assembled with the silicon powder in the interface. The assembly is loaded into a hot press and heated to 590ºC under load. The temperature and pressure are maintained for up to 30 minutes and the plate is removed.

Initial results of the TLPB process using a silicon interlayer are promising. More tests are needed to determine the exact process and parameters needed to make useable monolithic fuel. The processing temperatures required to obtain the Al-Si eutectic phase embrittle the age hardening aluminum 6061. So subsequent annealing steps are required to soften the material.

TLPB tests were also performed using germanium as an interlayer. With a lower eutectic temperature than silicon (420°C compared to 577°C for silicon) it was hoped that the embrittlement caused during silicon bonding would not occur. These tests were not successful. The higher density of germanium (5.32 g/cc compared to 2.33 g/cc with silicon), the eutectic composition (28.4% compared to 12.2 % with silicon) with aluminum and the lower temperature make TLPB inherently more difficult.

Friction stir welding (FSW) is a process developed in the early 1990's in Great Britain [7 and 8]. FSW employs a rotating tool (see Figure 6-left) 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 (see figures 6 and 7).

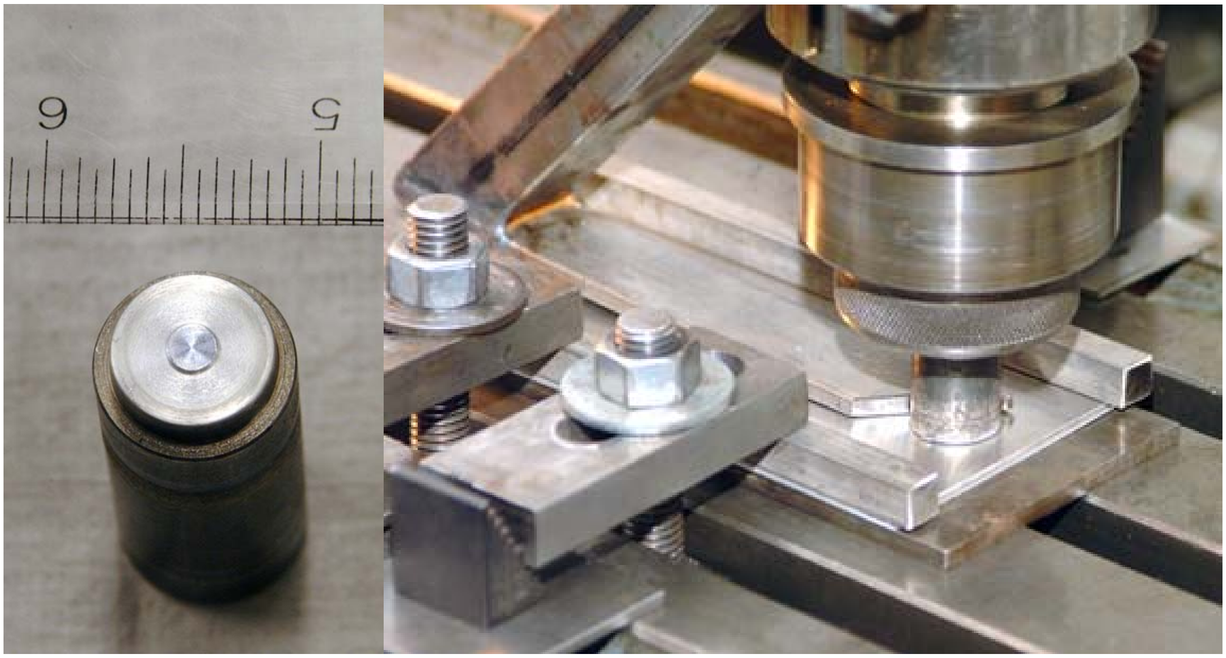

Figure 6. Friction Stir Welding. The view of the face of the tool (left) shows the pin in the center of the shoulder on the shaft. The pin is forced into the metal up to the shoulder which rides on the metal surface during processing. At right is shown the apparatus. The pin is rotated by a standard milling machine as the plate assembly is fed right to left. The sled (to the left of the tool) constrains the material in front of the tool aiding in a quality weld.

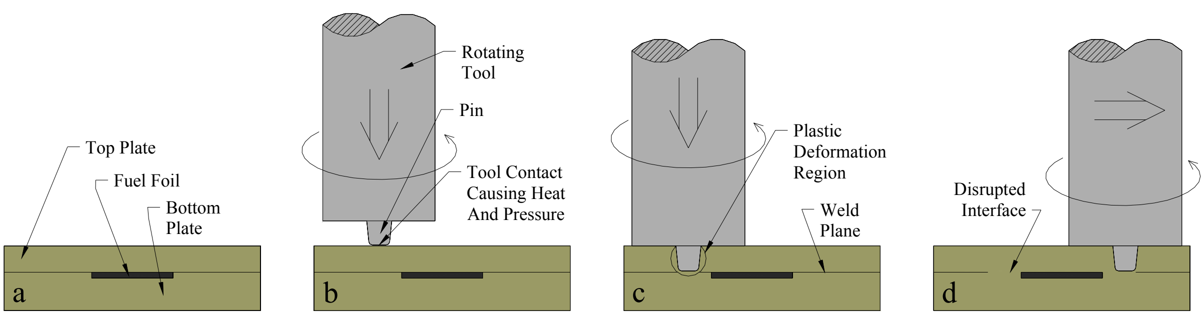

Figure 7. Friction Stir Welding Schematic. a) Shows the plate assembly for FSW of monolithic plates. In b) the rotating pin contacts the surface of the plate causing heat and pressure. In c) the tool has been plunged into the surface of the material up to the shoulder of the tool. The rotating tool is 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.

In conventional FSW the pin is forced through the interface of the materials to be joined. In order to fabricate monolithic fuel the pin is kept slightly above the interface to avoid disrupting the fuel foil and stirring it into the cladding. Aluminum 6061 cladding-to-cladding bonds fabricated using this process pass a standard bend test where a strip of the bonded aluminum can be bent 90° over a small radius and straightened without delaminating. This processing method also produces a good cladding to foil interface (see figure 8).

Figure 8. Friction Stir Weld Interface. The interface between aluminum 6061 (top) and a (surrogate) stainless steel foil is shown. The FSW processing plastically deforms the aluminum and smears it onto the foil creating an interface visually free of voids.

The action of FSW leaves a scalloped finish on the surface of the plate making it unsuitable for reactor use without further processing. This finish is smoothed and the plate is reduced to final thickness by cold rolling at low reduction (typically less than 5%), by machining or by mechanical polishing.

5. Bonding Discussion

In order to bond two aluminum surfaces together there must be material diffusion across the interface. Aluminum, a highly reactive metal, forms a stable oxide layer that protects the surface from additional oxidation and serves to effectively inhibit diffusion. This layer can be easily cleaned off through chemical or mechanical means but, unless done in the absence of oxygen, a thin layer reforms in a matter of seconds. To be successful, a bonding method must interrupt the oxide layer and allow diffusion across the interface.

Roll bonding disrupts the oxide layer that is stretched thin by the rolling process. The heat and pressure of the rolling process aid in promoting diffusion for bonding. High reductions are needed to sufficiently thin the layer and allow bonding. High temperature roll bonding was an attempt to increase the diffusion rate. It was hoped that the added diffusion would overcome the still significant oxide layer on the interface. The higher temperature, however led to additional oxide formation that hindered bonding.

Diffusion bonding in aluminum is difficult to achieve without taking measures to prevent oxide layer formation such as processing in either a vacuum or a reducing atmosphere or by eliminating the oxide in-process by using an alkali flux to clean the surfaces. The presence of an uninterrupted oxide layer hindered diffusion and bonding by this method.

In TLPB the eutectic phase formed with the silicon interlayer and the aluminum cladding acts as a flux to penetrate and erode away the oxide layer bridging the interface. As the liquid eutectic absorbs more of the aluminum it solidifies into the cladding forming a bond between the two plates. Diffusion in the plates strengthens the bond and disperses the silicon into the bulk of the aluminum.

Friction stir welding acts to mechanically disrupt the interface. This mechanical action of the rotating tool stirs the oxide layer into the bulk of the cladding. This mechanically aided diffusion successfully bonds the aluminum cladding.

Development of nondestructive bonding test methods will be an important part of further development and implementation of this fuel type. Development in this area is at an early stage.

6. Summary

- Gravity casting, cold rolling and resistance annealing have been used to produce U-Mo alloy foil on the miniplate scale.
- Both transient liquid phase bonding and friction stir welding show promise in bonding monolithic bearing fuel plates.
- Work has begun on production of depleted uranium surrogate monolithic fuel. This work will focus on ensuring the processes outlined above translate to the processing of U-Mo alloy fuel.

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