Development of a Monolithic Research Reactor Fuel Type at Argonne National Laboratory

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Abstract
The Reduced Enrichment for Research and Test Reactors (RERTR) program has been tasked with the conversion of research reactors from highly enriched to low-enriched uranium (LEU). To convert several high power reactors, monolithic fuel, a new fuel type, is being developed. This fuel type replaces the standard fuel dispersion with a fuel alloy foil, which allows for fuel densities far in excess of that found in dispersion fuel. The single-piece fuel foil also contains a significantly lower interface area between the fuel and the aluminum in the plate than the standard fuel type, limiting the amount of detrimental fuel-aluminum interaction that can occur. Implementation of monolithic fuel is dependant on the development of a suitable fabrication method as traditional roll-bonding techniques are inadequate.

Introduction
In an effort to limit, or eliminate, international traffic of highly enriched uranium (HEU) used to fuel research reactors around the world, the RERTR program was started in 1978 by the US Department of Energy to develop the technology needed to allow conversion of these reactors to low enriched uranium (LEU). LEU is defined as uranium with a $^{235}\text{U}$ isotope content of less than 20%.

In order to convert a reactor using HEU to one using uranium of lower enrichment, the amount of $^{235}\text{U}$ must be the same after the conversion. This necessitates a converting fuel uranium density that must be substantially higher to successfully convert a reactor. Any reactor fuel conversions must be done without significant penalty in reactor performance or safety and involve, at most, minimal geometry changes in the fuel element or core design\textsuperscript{1}.

The two ways to achieve a higher fuel uranium density are to a) increase the loading of the fuel in the fuel plate and b) change the composition of the fuel to one which contains a higher concentration of uranium.

The initial RERTR effort focused on raising the loading density of the existing dispersion fuel types. This early work resulted in fuel meat uranium densities of up to 3.7 g-U/cm$^3$. More recently a new fuel type, $\text{U}_3\text{Si}_2$, has been qualified with a uranium loading of 4.8 g-U/cm$^3$. With even higher loadings required to convert some reactors, work has centered on producing an increased uranium density fuel composition.
Uranium alloyed with molybdenum (the addition of Mo acts as a γ phase stabilizer) has been shown to be a promising fuel type for research reactors. Used as a dispersion fuel, it has been fabricated in test elements to loadings greater than 7 g-U/cm$^3$. While U-Mo dispersion fuel would represent a major advance in the ability to convert reactors to LEU, it does not allow all of the outstanding reactors to convert. To facilitate these conversions, a uranium density of greater than 8 g-U/cm$^3$ is needed. A new fuel form, monolithic fuel, would radically alter the possible fuel loading, giving a limit of above 16 g-U/cm$^3$.

Dispersion fuel, the industry standard, consists of the fuel in a powder form dispersed in an aluminum matrix that is clad between two cover plates. Monolithic fuel consists of a single fuel foil clad in aluminum (see figure 1). Absent the aluminum matrix found in dispersion fuel, monolithic fuel represents the ultimate in fuel loading being limited only in the density of the fuel alloy.

![Figure 1. End view of a dispersion fuel plate (left) showing the fuel particles (black) dispersed in an aluminum matrix, and a monolithic fuel plate (right) with a single-piece fuel foil.](image)

In addition to the higher fissile loading capability, monolithic fuel has an additional advantage. In dispersion fuel, the interface area between the fuel alloy and aluminum (in both the matrix and the cladding) is orders of magnitude higher than it is in monolithic fuel. During irradiation a reaction layer forms at this interface (figure 2). This acts as a thermal barrier which raises the temperature of the fuel. This elevated fuel temperature decreases the fuel performance and leads to more interaction layer formation, exacerbating the problem.

In a previous RERTR irradiation test, a total of two monolithic fuel plates were tested$^2$. These plates were fabricated by roll bonding$^3$. The thickness reduction required to fabricate the fuel tore the foils inside the fuel plates (see figure 3). Subsequent investigation has shown that roll bonding is not suitable for monolithic fuel plate fabrication as the foil will be stretched and torn with only light thickness reduction which is inadequate for the required plate bonding. Fabrication of monolithic fuel remains a major obstacle in the production of this fuel type.
Figure 2. Comparison of irradiated dispersion (left) and monolithic foil. Both plates were tested under similar irradiation conditions. The dispersion fuel has nearly consumed the aluminum matrix material and some voids have formed in the meat of the fuel. The monolithic fuel has a relatively clean interface with trivial amounts of oxide deposited during fabrication.

Figure 3. Radiograph of early monolithic fuel plate produced by roll bonding. The rolling required for bonding stretched and tore the foil which was originally a circular disk.

**Foil Fabrication**

The U-Mo fuel foil for the previous monolithic tests were produced by hot rolling in a process 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.

To aid in rolling the sample to the desired foil thickness, the U-Mo ingot is cast in a coupon 2.0 mm thick. This casting is performed in the arc-melting furnace using a gravity pour into a chill mold. Because the coupon is thin it can be easily cold rolled by means of a small, slightly modified jeweler’s rolling mill (see figure 4). Multiple passes are used to process the foil to a uniform desired thickness down to 0.13 mm.
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. 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 925°C. After annealing, the foil is rapidly cooled back to room temperature.
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. And, finally, should be a process that is suitable for commercial processing both in the ability to be scaled to full-sized plates and to be a commercially viable manufacturing method. Different bonding methods have been investigated for suitability in the monolithic fuel fabrication process. Transient liquid phase bonding, friction stir welding and hot isostatic pressing are all being investigated. The bonding methods were studied using Al-6061, an alloy that is prevalent in US research reactor fuel plates.

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\(^4\). Silicon and aluminum form a eutectic at 573°C, significantly lower than the melting point of aluminum (660°C).

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 590°C under load. The temperature and pressure are maintained for up to 30 minutes and the plate is removed.

Friction stir welding (FSW) is a process developed in the early 1990’s in Great Britain\(^5\). 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.

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.

Figure 7. 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 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 mechanical polishing.

FSW refines the grains to an extreme degree leaving behind different grain sizes. As reactor coolant is a corrosive environment the fuel plates are given a homogenizing heat treatment to remove potential regions of preferential corrosion attack.
Hot isostatic pressing (HIP) is a solid state process that was first developed in the 1950’s as a means to fabricate nuclear fuel by diffusion bonding. In this process, heat and pressure are applied to the process piece in an isostatic manner via a compressed gas, typically argon. The applied heat and pressure act to achieve diffusion and bonding across the interface.

For the samples tested in this study, the fuel plate assembly is encased in a steel can which is evacuated and seal-welded. Since this process has a non-dynamic interface, surface oxide removal prior to assembly is critical. The plate and foil surfaces are prepared and canned via a proprietary process and held at 583°C for 3 hours.

**Monolithic Bond Examination**

For fuel plates, two different interfaces are bonded. The cladding must be bonded with the opposing cladding and the cladding must also be bonded with the fuel foil. The fuel plate must be fully bonded around the edges to hermetically seal the fuel from the coolant water to avoid releasing fission products into the reactor. Three methods are used to examine the bond quality, bend testing, ultrasonic testing and microscopic examination of the bond.

Bend testing is performed by clamping a thin section of the non-fueled region of the welded plate into a fixture and bending it over a radius. The plate is bent three times for a total of 360°. If any delamination is noted the bond is considered unacceptable. As this test is destructive in nature, it is only performed on adjacent material removed in the sizing of the fuel plate. As such, the investigation is performed only to test the cladding to cladding bond (the bimetal bond between the fuel foil and the cladding is not robust enough to survive this treatment). All three bonding methods passed the bend test check of the cladding to cladding bonds.

Ultrasonic testing (UT) is the only method being used that can non-destructively examine the bonding of the fuel plate itself. Tests have been performed on fuel plates to examine the bonding, both the cladding-cladding and the cladding-fuel bonds. The samples were inspected for debonds using standard ultrasonic through transmission techniques. The transducers are a pair of 15 MHz focused transducers with a 0.25” diameter lens, focused at 0.5 inches. The raster scan parameters were 100 mm by 30 mm, the raster index was 100µm, and the scan step was 100µm. The raster scan was performed with greater than a 50% beam overlap. Tests have been conducted on both the FSW and the TLPB methods (as HIP has only undergone surrogate testing it has not been tested by UT). UT shows both FSW and TLPB bonded over a substantial region of the plate. The TLPB results show, however two significant regions were there was not an acceptable bond (see figure 8).
Microscopic testing was done by sectioning the bonded elements, mounting, grinding and polishing through 1µ with diamond grit suspension. This method is used primarily to examine the bond quality between the cladding and the fuel foil. A scanning electron microscope is used to examine the bond. This allows examination of the interface to both determine porosity and reaction layers (figure 9).

To date, the HIP process has only been performed with stainless steel surrogate foil. The interface between the foil and the cladding clearly shows a reaction layer. This is troubling since the fuel alloy is much more reactive with the aluminum cladding material than the surrogate stainless steel. These interfaces are brittle and showed signs of delamination during cool down or removal of the protective over pack but it remains to be seen how this process will translate to the U-Mo foil.

In the SEM examination, the as-fabricated FSW bond shows no voids and no interaction layers. This bonding is also evident along the edges of the foil which are not along the two welding planes. Since heat treatment is a part of the fuel plate fabrication, a sample was annealed at 500°C for 30 minutes. A thin (>3 µm) reaction layer formed on the interface.

The TLPB process also shows an interface with a noticeable reaction layer. The thickness of this reaction layer seems dependant on the amount of silicon that is applied to the interface. As we have little control over the thickness of the applied silicon at this time, it is difficult to draw any real conclusions about the amount of reaction layer. One
troubling indication is the formation of spikes of a (U-Mo)Al$_7$ phase that has been implicated in detrimental fuel behavior in previous dispersion fuel irradiation tests.

Both the FSW and the TLPB processes have been used as crude diffusion couples to study the reaction layer kinetics of the fuel-cladding interaction zone. Heat treating the FSW sample at 500°C for 30 minutes incurs a minute (<3 µm) interaction zone. This layer appears stable with no regions of breakaway interaction or signs of debonding.

Questions about the amount of silicon deposited during the preparation of the TLPB samples makes these results suspect but the overall trends can be studied. The general
trend in the diffusion couple study show that the amount of reaction layer in the monolithic fuel is less than is typically found in the dispersion fuel.

There are some indications that the interaction layer suppression is achieved by different compositions of aluminum. The matrix dispersion material is unalloyed aluminum while the cladding material is aluminum 6061. The alloying constituents in the 6061 appear to inhibit the reaction layer growth. The effect of this on actual fuel performance will not be known until the irradiated samples are examined as irradiation typically enhances diffusion kinetics.

**Dispersion Fuel**

U-Mo dispersion fuel, the focus of the RERTR advanced fuel development for the past several years, has been showing some limitations under high temperature and burnup conditions. It is thought that in the right circumstances a detrimental phase forms at the interface between the pure aluminum matrix and the fuel powder.

Studies into this phenomenon have indicated that small silicon additions to the matrix material may inhibit formation of the problematic phase. The plate cladding material for the RERTR tests, Al-6061, contains some silicon and is markedly more resistant to reaction with the fuel than the pure aluminum found in the matrix.

It is anticipated that silicon additions to the matrix will inhibit excessive reaction layer formation and improve fuel irradiation performance under high-power operation. Several alloys are being considered as matrix replacements for pure aluminum. Four aluminum alloys will be tested, two Aluminum Association alloys (Al-6061 (0.4-0.8 wt.% Si) and Al-4043 (4.5-6.0 wt.% Si)) and two binary alloys (Al-2 wt.% Si and Al-5 wt.% Si).

**Irradiation Testing**

New fuel types will be tested in the Advanced Test Reactor (ATR) in Idaho. The ATR has a number of high flux positions that have been used for previous RERTR irradiation tests. Current planning calls for two irradiation tests, RERTR-6 and RERTR-7. RERTR-6 will focus more on the monolithic fuel type while the RERTR-7 test will be focused more on alternative matrix dispersion fuel. It is anticipated that both experiments will use the same hardware design employed for the RERTR-4 and -5 irradiation tests (see figure 10 l.). This hardware consists of four capsules containing eight miniplates (measuring 2.54 cm x 10.16 cm x 1.4 mm) each. The capsules are open on each end to the primary coolant water, which will flow in direct contact with the plates. The capsules are housed in an outer basket that is inserted into a large B-hole near the ATR driver fuel (figure 10 r.). Table 1 gives the anticipated parameters for the upcoming irradiation tests in comparison to those of past tests.
The current anticipated RERTR-6 test matrix is comprised of 21 monolithic fuel plates and 11 dispersion plates. The monolithic plates will consist of a foil sized 8.3 cm x 1.9 cm with corner radiiuses of 0.32 cm. The thickness will be varied as an experimental parameter. The plates will be fabricated by the FSW and the TLPB processes. Composition of the foil will be 7 and 10 wt.% Mo. The standard thickness for the foil will be 250µm but will be expanded for a limited number of plates to 500µm.

Table 1. RERTR Experiment Comparison

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* All data for RERTR tests 6 & 7 is anticipated
† G = Ground Powder; A = Atomized Powder; M = Monolithic
‡ Uncertainty in power of experimental position
Dispersion fuel plates will also be tested in the RERTR-6 experiment. As in previous tests, $U_3Si_2$ dispersion fuel will be included as a known control. Tests will also be run looking at various matrix alternatives to pure aluminum. Current plans include using the alloys (ASTM alloy designation) Al-6061 (commonly used as the cladding material in the RERTR tests), Al-4043 (containing 4-6 wt.% Si) and two binary alloys: Al-2 wt.% Si and Al-5 wt.% Si. In addition, plates will be included to test the hybrid fabrication method outlined above.

The RERTR-7 test will be irradiated under conditions in which excessive fuel matrix interaction and void formation has been found in previous in-pile tests. It is anticipated that this test will be comprised primarily of dispersion fuel plates with some monolithic plates added to fill out the test matrix. This test will be run to an estimated burnup of $\sim 75\%$.

It is anticipated that the RERTR-7 test will be comprised of many of the fuel and matrix compositions being included in the RERTR-6 test but the makeup of the matrix is awaiting more detailed results from postirradiation examinations of previous tests and for a number of diffusion couple experiments to be performed on the various alloys being considered.

Irradiation tests of full-sized plates are planned in addition to the miniplate tests in ATR. These tests could start at about the same time as the miniplate tests, or they could start up to six or more months later, depending on fabrication schedules and on which reactor is used for the tests. If the plate tests are successful, full-sized element tests and review of the data by the U.S. Nuclear Regulatory Commission are anticipated to complete the fuel qualification process.

**Summary**
- Fundamental loading limits of dispersion fuel will not allow conversion of all high power research reactors to low enriched uranium.
- Monolithic fuel has a substantially higher loading than dispersion fuel.
- Monolithic fuel requires a new fabrication method.
- Three fabrication methods are being pursued: hot isostatic pressing, transient liquid phase bonding and friction stir welding.
- Dispersion fuel matrix material is being adjusted in hopes of limiting detrimental reaction phase formation.
- Two irradiation tests are being fabricated to test the new fuels, these will be inserted in the Advance Test Reactor in 2005.

**References**
Reduced Enrichment for Research and Test Reactors, Las Vegas, NV, October 1–6, 2000.


