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## Research Reactor Fuel Management (RRFM)

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# UPDATE ON US HIGH DENSITY FUEL FABRICATION DEVELOPMENT

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## ABSTRACT

Second generation uranium molybdenum fuel has shown excellent in-reactor irradiation performance. This metallic fuel type is capable of being fabricated at much higher loadings than any presently used research reactor fuel. Due to the broad range of fuel types this alloy system encompasses—fuel powder to monolithic foil and binary fuel systems to multiple element additions—significant amounts of research and development have been conducted on the fabrication of these fuels. This paper presents an update of the US RERTR effort to develop fabrication techniques and the fabrication methods used for the RERTR-9A miniplate test.

## Introduction

The United States Reduced Enrichment for Research and Test Reactors (RERTR) Fuel development program is tasked with the development of a fuel type that will allow conversion to low enriched uranium (LEU) of the world's research reactors that are currently fueled by uranium enriched to more than 20% $U^{235}$ (HEU).

In order to accomplish this goal, metallic fuel (uranium-molybdenum alloy with 7-10 wt.% Mo*) clad in aluminum is being developed. This research is being focused in two areas: dispersion fuel with greater than 8 $gU/cm^3$ and monolithic fuel where the fuel loading can be greater than 16 $gU/cm^3$.

Fabrication development efforts on dispersion fuel have shown promise for U-7Mo fuel in a silicon doped aluminum matrix. The silicon addition to the matrix provides added stability in irradiation behavior that has been demonstrated at uranium loadings of 6 $g/cm^3$. Monolithic fuel development has focused on fabrication issues where two techniques have emerged, Friction stir welding and hot isostatic pressing.

## Fuel Fabrication

—*Dispersion Fuel* Unlike compound fuel types (silicides, aluminides, carbides, etc.) metallic fuel is difficult to fabricate into a powder form in the solid state [1]. Atomization has been used successfully to produce spherical fuel powder on a production scale at the Korean Atomic Energy Research Institute (KAERI) via a rotating disk atomization method. This powder has been used to fuel a number of past RERTR irradiation tests (RERTR-1 through RERTR-6). However, the need to quickly produce

* All compositions are given in weight percent unless otherwise noted

custom alloys and to raise enrichment levels to meet the requirements of increasingly more stringent irradiation conditions dictated the need to have a laboratory scale atomization setup at the US RERTR fuel development laboratory.

The need to fabricate laboratory-scale quantities of different alloy powders for testing, lead to the construction of the atomization system in place at the Idaho National Laboratory (INL). This rotating electrode process (REP) style atomizer is capable of rapidly producing quantities of powder in the 20 gram range. The REP operates by spinning a cylindrical rod of the desired alloy while an arc is used to melt the end of the rod. The centrifugal force on the molten portion of the rod causes the material to be flung off in small droplets which solidify into powder (figure 1).

$$D = \left(\frac{A}{\omega}\right)\sqrt{\frac{\gamma}{\rho_m R}}$$

Where:
$D$ = Expected Particle Size
$A$ = Process Constant
$\omega$ = Angular Velocity
$\gamma$ = Melt Surface Energy
$\rho_m$ = Melt Density
$R$ = Electrode Radius

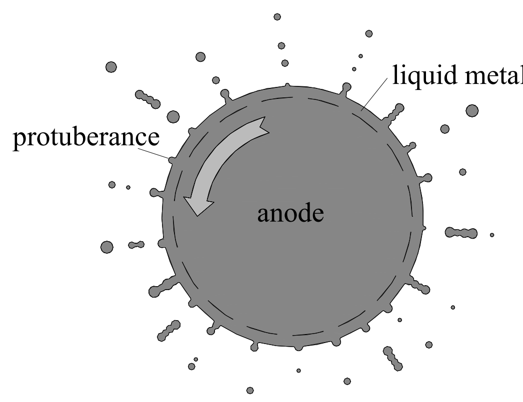


**Figure 1. Rotating electrode process atomization. Left, empirical equation expressing predicted powder size. Right schematic diagram of particle formation.**

Initially, the powder size produced from this process was somewhat larger than typically used for dispersion fuel production [2]. This large powder was used in the RERTR-8 and -9 experiments with resulting 'dogbone' inhomogeneity, as fuel piled up at the ends of the fuel zone. To decrease the size of the powder produced by the REP atomizer for a given alloy, both the rotational speed and the radius of the consumable electrode can be increased to achieve the desired result as indicated in the equation shown in figure 1. For the RERTR-9 irradiation experiment, the size of the electrode was increased from a diameter of 6.3 mm to 9.5 mm. and the rotational speed was increased from 38,500 rpm to ~45,000 rpm. The resulting powder size distribution was comparable to that supplied by KAERI for earlier fuel testing (Table 1).

—*Monolithic Fuel* The monolithic fuel foil fabrication process at INL has been modified to accommodate a wider range of uranium impurity contents. Initially, a cast coupon was cold rolled to 90% reduction (the amount required for a 250µ foil—the typical desired thickness), annealed and sheared to final size [3]. This process introduced a large amount of cold work into the rolled sample and, while an annealing step would ameliorate this, foils would often develop cracks during rolling causing a higher-than-desirable rejection rate.

To improve the process, the coupon is now encapsulated in a carbon steel can (figure 2, left) and hot rolled at 650 ºC to ~85% reduction. The sample is then decanned and cold rolled the remaining 15% to achieve the target thickness (typically 250 μm). The process change has resulted in a success rate of > 90% for both the miniplate and full size foil geometries (19 mm x 82.5 mm and 40 mm x 510 mm, respectively).

| Powder Size (μm) | Powder Source (RERTR Experiment #) | | | |
|---|---|---|---|---|
| | KAERI (3) | AECL (4) | INL (7) | INL (9) |
| < 45 | 28.9% | 0.1% | 4.8% | 32.0% |
| 45-63 | 24.9% | 16.6% | 11.7% | 14.3% |
| 63-75 | 20.4% | 14.0% | 9.5% | 31.3% |
| 75-106 | 22.3% | 32.6% | 12.5% | 22.5% |
| 106-150 | 3.4% | 36.6% | 61.5% | — |

**Table 1. U-7Mo powder size comparison. A comparison of the four primary powder sources used for U-Mo irradiation testing. The sources were the Korean Atomic Energy Research Institute (KAERI) which provided atomized powder for the experiments 1-6 (size results are only for powder used for experiments 4-6); Atomic Energy Canada Limited (AECL) which provided mechanically milled powder for the RERTR-4 and 5 experiments, and the Idaho National Laboratory (INL). The two batches of the INL powder show the resulting refinement of powder size due to process alterations.**

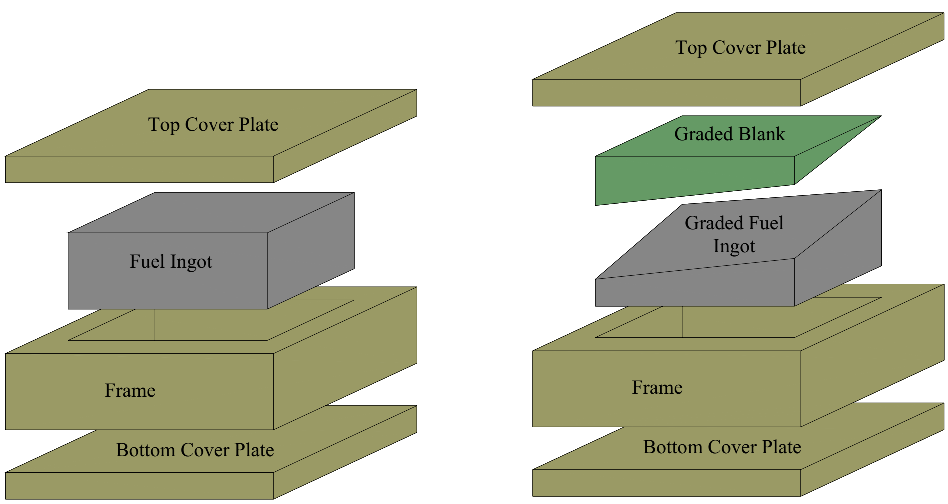


**Figure 2. Canned foil rolling. Left, standard foil production method: fuel ingot is enclosed in a welded steel can and hot rolled at 650 ºC. Right, graded fuel ingot and graded blank are canned in steel jacket and rolled at 650 ºC. Parallel rolling surfaces (the top of the blank and the bottom of the ingot) are needed to produce a graded foil by the rolling process.**

In addition to improved foil production of flat foils, the canning method also shows promise in the production of shaped foils. Reactors with complex fuel cross sections (such as HFIR) would, if monolithic fuel is to be used, need to have a method of manufacturing foils with a graded cross section. To obtain a shaped foil the outer can is used as before but the ingot is shaped to represent the desired final profile. A blank is then fabricated to fill the remaining space in the steel can. This blank provides the parallel rolling surfaces needed to process the material (figure 2, right). Initial testing on the graded foil concept have shown that shaped foil can be achieved by the graded blank method. A 400 mm x 23 mm U-10Mo foil was rolled to a wedge shape (with the thickness gradient across the minor dimension). The thin edge was rolled to an average of 68 μm (12% average variation), and the thick edge was rolled to and average of 363 μm (9% average variation). Figure 3 shows the rolled foil.

**Figure 3. Graded U-Mo foil. DU-10Mo foil produced by hot rolling.**

### Fuel Plate Bonding

For the RERTR-9A experiment three different bonding methods were employed: for dispersion fuel plates a rolling/hot isostatic pressing method was used, for monolithic plates friction stir welding and hot isostatic pressing were employed.

—*Dispersion Fuel* Plate specimens fabricated for previous RERTR irradiation tests with roll bonded dispersion fuel at high loadings (6 - 8 $gU/cm^3$) typically suffered from problems with homogeneity—both in pile-up at the ends of the fuel zone (dogboning) and through the body of the fuel zone. Three methods were employed to improve the homogeneity: reduction of fuel particle size, a dual compaction step, and decreasing the rolling reduction ratio.

Powder size for the RERTR-6 and 7 dispersion fuel plates was limited to a maximum of 150μm. The portion of the largest size fraction (106-150 μm) used was greater than 50%. For the RERTR-9A experiment, the maximum powder size was limited to 106 μm, and the portion of the largest size fraction (90-106 μm) was limited to 30%.

Typically U-Mo fuel powder is blended with the matrix powder and compacted to produce a fuel pellet. This method was altered to produce RERTR-9A dispersion fuel plates. To enhance the homogeneity and obtain better green strength a common powder metallurgy technique was employed: compaction followed by crushing and recompaction.

In previous experiments, the rolling reduction performed on dispersion plates was 6:1, an amount of reduction historically found to produce good bonding in dispersion fuel plates. For the RERTR-9A test this reduction was reduced to 3:1. This lower reduction dictated a change in geometry of the assembled frame. The compacts were pressed to half thickness and two compacts were included in each plate to retain the fuel zone volume (by laying the compacts end-to-end the decreased rolling reduction still resulted in a full length fuel zone). The plate hardware was reduced from three pieces (two cover plates and a 'picture frame') to two (a cover plate and a recessed bottom plate) to reduce the number of bonding interfaces from two to one.

The modifications to the dispersion fuel outlined above resulted in fuel plates with lessened dogboning. The bonding in the cladding region showed no negative indications in ultrasonic testing or in bend testing. The ultrasonic scans of the fuel region, however, showed reduced sound transmission. While reduced sound transmission is typically indicative of lower bonding quality it also probably due, in part, to signal attenuation by the added fuel loading.

To improve the fuel/clad bonding the rolled plates were subjected to subsequent processing in the HIP. The plates were heated with a ramp rate of 400 ºC/hr to 500 ºC and pressurized at 100 MPa and held for 30 minutes. As the plates had been hermetically sealed during rolling, no secondary encapsulation was required. Ultrasonic testing of the plates shows definite improvement in the UT scans (figure 4).

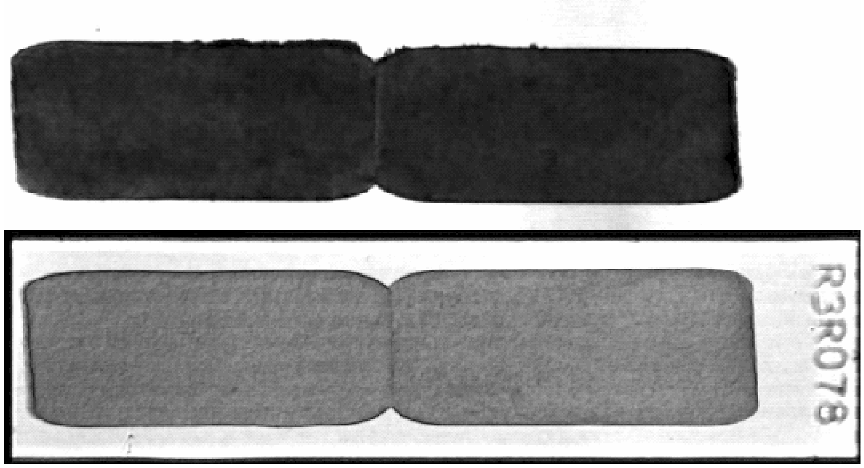


**Figure 4. Ultrasonic transmission scan of a roll bonded fuel plate before (top) and after HIP process. Note changes in the shape of the fuel zone caused by the HIP process.**

—*Monolithic Fuel* As noted elsewhere [4], the two monolithic bonding methods being pursued by the US RERTR program are hot isostatic pressing (HIP) and friction stir welding (FSW). These methods continue to undergo process enhancement to improve both the quality of the product and the ability to fabricate fuel, both on a small and large scale.

Monolithic fuel miniplates fabricated by HIP were first included in the RERTR-8 irradiation test. These plates were processed at 100 MPa and 580 ºC for 90 minutes. Since the HIP temperature and the aluminum 6061 alloy (the standard aluminum alloy used for US design research reactor fuel cladding) solidus temperature are in the same range*, actual production of fuel plates with this method may be problematic.

To avoid potential problems, as well as reduce the severity of the overall thermal history, the temperature of the HIP process was lowered 20º to 560 ºC—all other HIP parameters were left unchanged. U-10Mo plates processed with these parameters showed good bonding throughout the fuel plate. Comparison of the clad to clad bonding in the

* Due to compositional ranges in alloy composition the lower bound melting temperature is reported from between 580-592 ºC.

cladding region between plates processed at the two temperatures shows virtually identical bond interface microstructure (figure 5).

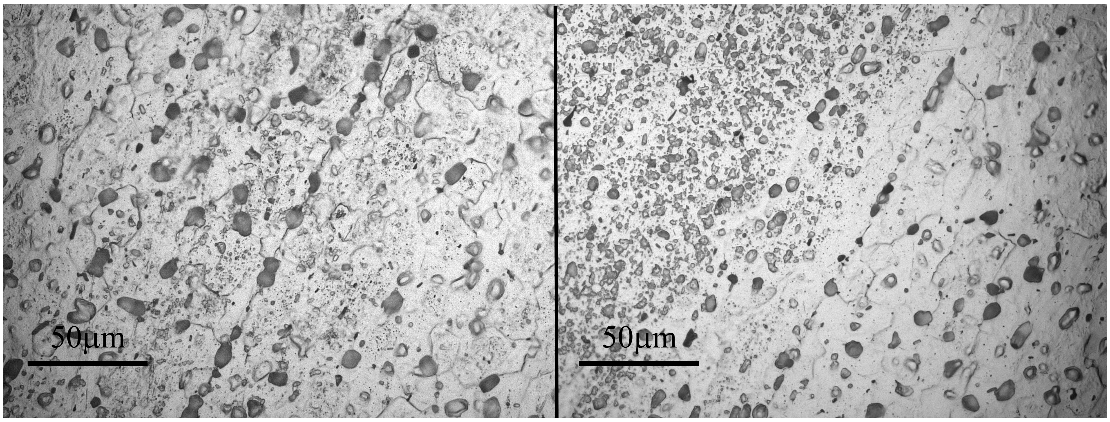


**Figure 5. Aluminum bonding after hot isostatic pressing. Aluminum 6061 clad/clad bonding in HIP 100 MPa, 15 min dwell time 580 ºC (left) 560 ºC (right). Bond line runs diagonally down (right-to-left) in both micrographs.**

The FSW process has been used to fabricate 26 miniplates in irradiation tests starting with RERTR-6. Since the process quality is greatly dependant on controlling the temperature of the aluminum during processing significant effort has been placed on the development of a cooling system to remove excess heat from the welding face.

A cooling system designed to provide coolant flow through the steel tool resulted in higher quality welding, but instrumentation showed that temperature rise at the tool face was still negatively affecting the quality of the bond [5]. To facilitate heat removal from the face of the tool, the material used for the tool face was changed to one with a substantially higher thermal conductivity. This new tool material has shown marked improvement in the weld quality and the ability to maintain consistency of the weld during processing.

## Experimental Testing

The RERTR-9 experiment is a miniplate test undergoing irradiation testing in the Advance Test Reactor (ATR) in Idaho. This overall test consists of 32 fuel plate positions in 4 flow-through capsules. The experiment plate and hardware geometry is identical to that used in previous RERTR miniplate tests in the ATR [6]. The experiment will be inserted into the reactor in two stages. RERTR-9A began irradiation in February of this year while RERTR-9B is scheduled for insertion in June. The RERTR-9A stage of the experiment is contained in the A and C experiment capsules. Capsules C and D are filled with blank plates until they are replaced with the fueled RERTR-9B portion of the experiment.

The RERTR-9A portion of the experiment contains 14 fuel plates (two blank plates were added to fill the capsules). The experimental matrix is comprised of 5 dispersion fuel plates and 9 monolithic fuel plates (Table 2). The dispersion fuel plates were fabricated with U-7Mo fuel powder at a nominal loading of 8 $gU/cm^3$ and were fabricated by the rolling process followed by a HIP bonding step described above. Three different types of matrix material were used for this experiment: aluminum 4043 alloy (nominal Si content 5%), binary aluminum 2% silicon alloy, and a binary powder mixture consisting of aluminum and 2% silicon.

| As-Loaded RERTR-9A Experimental Matrix | | | | |
|---|---|---|---|---|
| **Capsule** | **Column 1** | **Column 2** | **Column 3** | **Column 4** |
| A-Top | A1<br>Blank | A2<br>U-7Mo 8 g-U/cc<br>Al-4043<br>**R3R078** | A3<br>Blank | A4<br>U-10Mo FSW<br>250 µm Foil<br>**L1F22C** |
| A-Bottom | A5<br>U-10Mo FSW<br>250 µm Foil<br>**L1F27C** | A6<br>U-10Mo HIP<br>Al-4043 layer<br>**L1P03A** | A7<br>U-7Mo 8 g-U/cc<br>Al + 2 Si<br>**R4R028** | A8<br>U-10Mo FSW<br>250 µm Foil<br>**L1F29C** |
| B-Capsule | Blank B-Capsule (Reserved for RERTR-9B) | | | |
| C-Top | C1<br>U-10Mo FSW<br>250 µm Foil<br>**L1F26C** | C2<br>U-7Mo 8 g-U/cc<br>Al-4043 Matrix<br>**R3R108** | C3<br>U-7Mo 8 g-U/cc<br>Al-2 Si Matrix<br>**R2R078** | C4<br>U-10Mo FSW<br>250 µm Foil<br>**L1F28C** |
| C-Bottom | C5<br>U-10Mo FSW<br>250 µm Foil<br>**L1F32C** | C6<br>U-10Mo HIP<br>Al-4043 layer<br>**L1P04A** | C7<br>U-7Mo 8 g-U/cc<br>Al + 2 Si Matrix<br>**R4R018** | C8<br>U-10Mo FSW<br>250 µm Foil<br>**L1F24C** |
| D-Capsule | Blank D-Capsule (Reserved for RERTR-9B) | | | |

**Table 2. RERTR-9A experimental matrix**

The monolithic fuel plates were fabricated by two different methods. HIP was used to fabricate two fuel plates containing U-10Mo with an aluminum 4043 alloy foil placed along a single face of the foil. The addition of the silicon-bearing aluminum alloy is designed to counteract the fission gas bubble formation noted in past irradiation experiments [7]. The remaining 7 plates were fabricated by FSW. These plates were fabricated using the two different tool face materials resulting in different fuel-to-cladding bond strength. The plates will serve as a test to determine the effect on of initial bond strength on the post irradiation bonding

The RERTR-9A experiment is designed to have irradiation conditions similar to other recent irradiations tests. The anticipated conditions and a comparison to previous tests is found in Table 3.

**Acknowledgment**

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| | RERTR Experiment Number | | | | | | | |
|---|---|---|---|---|---|---|---|---|
| **Variables** | **1 & 2** | **3** | **4** | **5** | **6** | **7*** | **8** | **9A** |
| ATR Reactor Position | I-22 / I-23 | B-7 | B-12 | B-11 | B-12 | B-11 | B-11 | B-11 |
| Experimental Plates | 32 each | 47 | 32 | 32 | 32 | 29 | 8 | 14 |
| U-Mo Fuel Fabrication† | G, A | G, A | G, A, M | G, A | M, A | A, M | A, M | A, M |
| Fuel Loading (g-U/cm$^3$) | 4 | 8.5 | 6, 8 | 6, 8 | 6, 16 | 6, 16 | 6, 16 | 8, 16 |
| Enrichment (% $^{235}$U) | 19.5 | 19.5 | 19.5 | 19.5 | 19.5 | 20 / 58 | 58 | 58 / 44 |
| Max Heat Flux (W/cm$^2$) | 70 | 400 | 210 | 320 | 250 | 320 | 300 | 320 |
| BOL Cladding Temp. (°C) | 70 | 150 | 130 | 175 | 130 | 100 | 100 | 100 |
| Peak Fuel Temperature (°C) | 70 | 235 | 140 | 189 | 150 | 130 | 130 | 130 |
| Coolant Velocity (cm/s) | 920 | 600 | 600 | 600 | 600 | 1400 | 1400 | 1400 |
| Number of Cycles | 2 & 8 | 2 | 7 | 3 | 3 | 2 | 2 | 2 |
| Total Duration (days) | 94 & 232 | 48 | 230 | 116 | 136 | 100 | 100 | 100 |
| Burnup at EOL (% $^{235}$U) | 40 & 70 | 40 | 80 | 50 | 50 | 85‡ | 70‡ | 75‡ |

* Includes experiments 7A and 7B
† G = Ground Powder; A = Atomized Powder; M = Monolithic
‡ LEU (20% $^{235}$U) Equivalent

**Table 3. Comparison of RERTR miniplate irradiation experimental conditions**

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