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PRODUCTION AND CHARACTERIZATION OF ATOMIZED U-Mo POWDER BY THE ROTATING ELECTRODE PROCESS*

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ABSTRACT

In order to produce feedstock fuel powder for irradiation testing, the Idaho National Laboratory has produced a rotating electrode type atomizer to fabricate uranium-molybdenum alloy fuel. Operating with the appropriate parameters, this laboratory-scale atomizer produces fuel in the desired size range for the RERTR dispersion experiments. Analysis of the powder shows a homogenous, rapidly solidified microstructure with fine equiaxed grains. This powder has been used to produce irradiation experiments to further test adjusted matrix U-Mo dispersion fuel.

1. 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. 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. Greater loadings (in excess of 8 g/cm^3), however, are required to make U-

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† All compositions are given in weight percent unless otherwise noted

Mo dispersion a viable fuel candidate for a core conversion.

The dispersion fuel development effort in the United States has, to date, focused on miniplate (and smaller) irradiation testing with over one hundred U-Mo fuel plates in eleven experiments. These tests have continued with the smaller geometry being ideally suited to quick turnaround fuel scoping and material compatibility tests.

2. Fuel Atomization

Fuel powder for the RERTR high density metallic fuel dispersion irradiation tests has been produced by three methods, machining, grinding and atomization. Atomized powder that is spherical in shape has been shown to produce dispersion fuel plates with lower void porosity than powder made by mechanical means. This lower porosity results in inherently higher fuel loadings. Atomized powder used for the first six RERTR U-Mo tests was produced at the Korea Atomic Energy Research Institute (KAERI) [1]. This powder was produced in a range of compositions but was limited to less than 20% ^{235}U enrichment.

In order to more rigorously test fuel for the RERTR program, the fuel surface heat flux was to be raised beyond what could be obtained from an LEU powder. Since supplying fuel powder with a target enrichment of nearly 60% ^{235}U necessitated finding a powder source other than KAERI, the decision was made to build a small scale atomizer at the Idaho National Laboratory (INL) [2].

The INL atomizer produces fuel powder via the rotating electrode process (REP). This atomizer is based on a commercial design but differs in orientation and electrode feed [3]. Commercial REP units typically have a horizontal axis of electrode rotation and the consumable alloy is fed into the anode. The unit in operation at INL uses a vertical rotational axis and the anode is fed down onto the diminishing alloy cathode (figure 1). This arrangement allows for easier operation inside a glovebox and the non-translating cathode reduces alloy waste and provides greater stability for the rotating U-Mo rod.

3. Fuel Characterization

—*Size & Shape* Since particulate size is of importance in fuel fabrication [4] and the powder previously produced by the INL REP atomizer was trending too high, the process was redesigned to reduce the size fraction of the powder. Empirical calculations for powder size [5] show that the diameter for a given alloy is inversely proportional to the rotational velocity and related to the anode diameter and to the melt rate. To raise the rotational velocity, the current supply to the drive motor was increased from 120 to 140 Volts using a variable transformer unit. Angular velocity (measured by an adjustable strobe light) was shown in increase with greater applied voltage but the increase was trending to a plateau of slightly over 45,000 rpm at 140 V, the peak voltage of the transformer. This voltage rise (and subsequently higher rotational velocity) resulted in a finer powder size distribution (figure 2). The useable powder yield (for the RERTR-9 experiment the upper powder size threshold was set at 106 µm) for the two voltages is 29 and 60 percent for 120 and 140 volts, respectively. This yield can be further raised by re-melting the oversized powder and performing an additional atomization run—an added re-melting and atomization raised the yield of the 140 V process to over 75%.

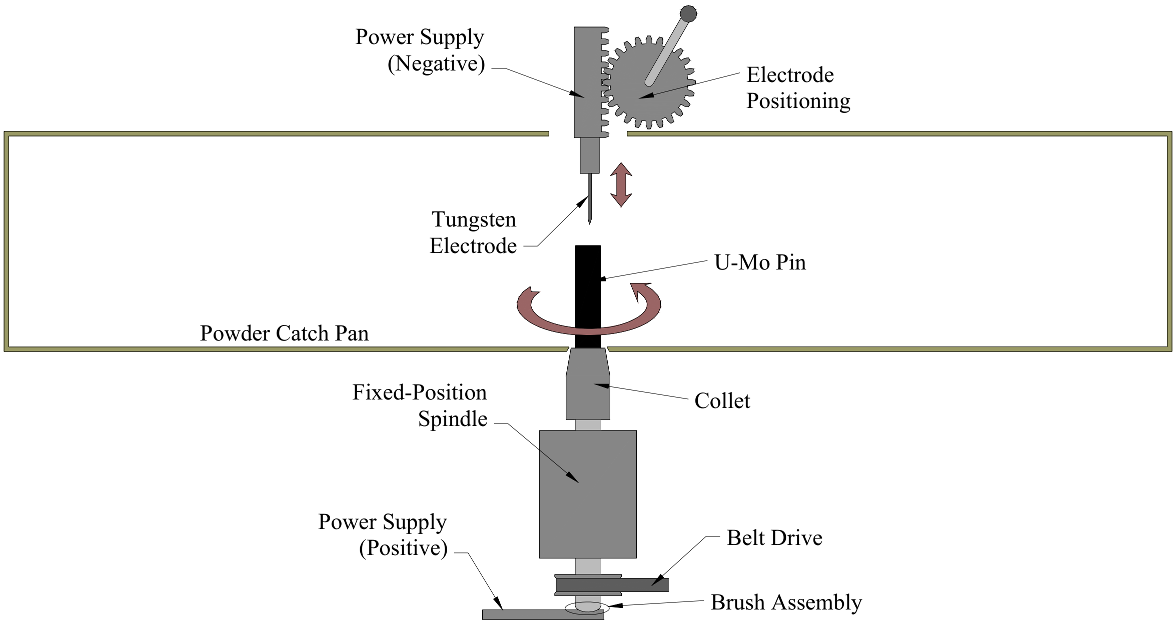

Figure 1. Schematic of the INL Powder Atomizer. The atomizer is a vertically oriented rotating electrode process machine capable of atomizing a ~45 g U-Mo pin. This unit is capable of higher rotational speeds and smaller particulate sizes than a standard REP type atomizer.

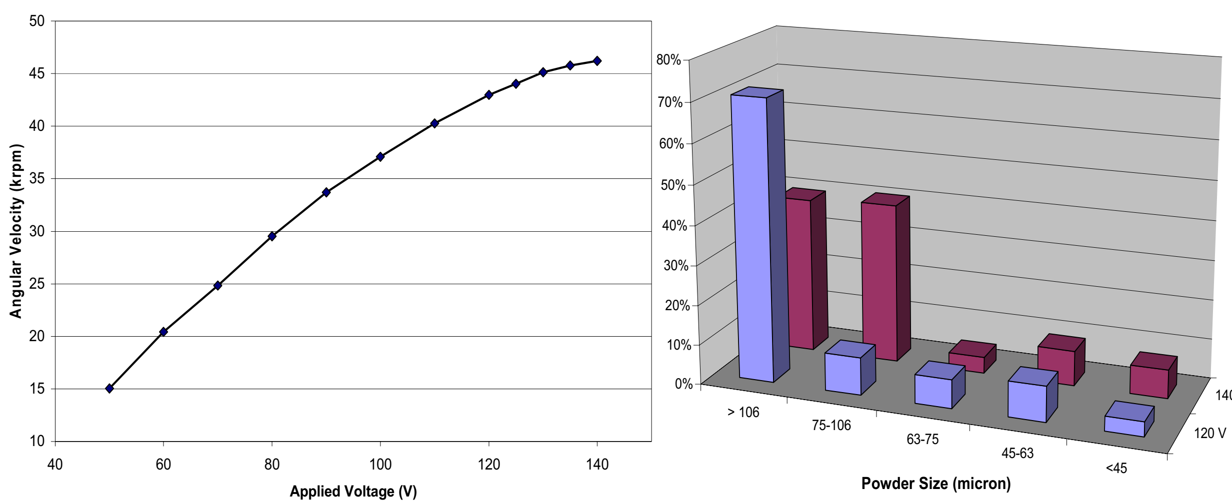

Figure 2. Plot of angular velocity (left) as a function of applied voltage (under load) for the REP atomizer. Powder size comparison (right) for U-7Mo atomized with rotational velocity driven by 120 and 140 volts.

The shape of REP type powder tends to be spherical so long as the material has enough time for the surface tension of the molten material to draw it into a sphere prior to solidification of the material. Material (melting point, surface tension and oxidation potential) and process (amount of superheat, type and purity of quench media, droplet size and cooling chamber diameter) characteristics factor into the final shape of the resulting powder. The fabricated powder was generally spherical with a small fraction of irregular clumping and a few flattened areas on some of the larger particles. The clumping is due to either insufficient separation of the molten alloy or alloy droplets that collide and coalesce prior to solidification. The flat faces on the larger powder are from impacting the collector wall before full solidification was achieved (figure 3).

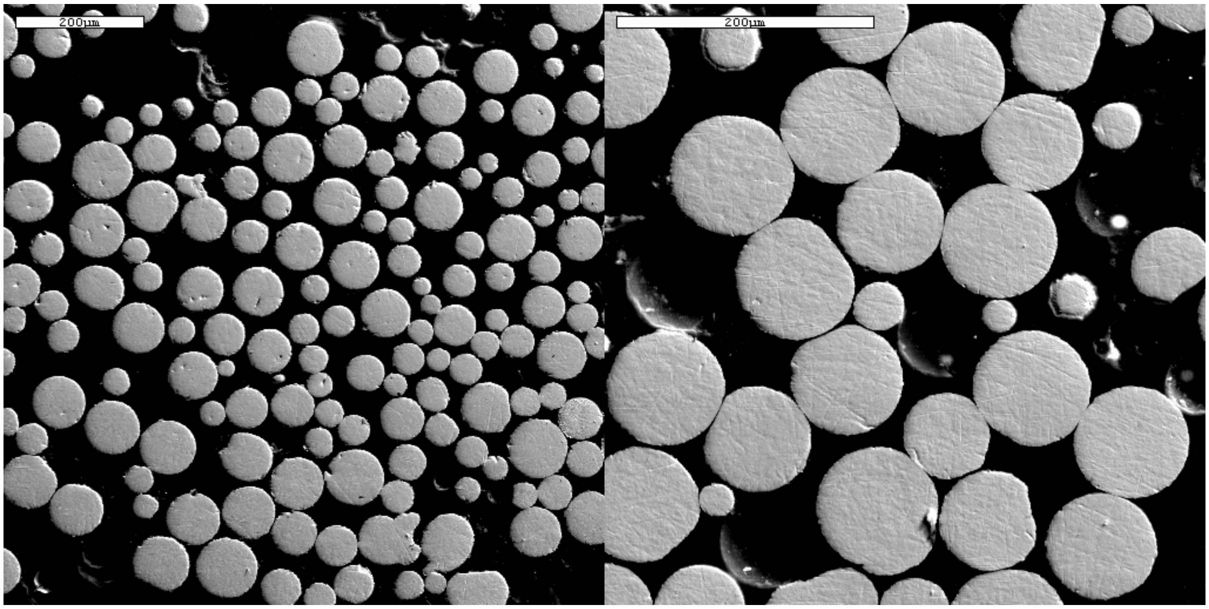

Figure 3. U-7Mo powder. SEM micrographs of sectioned and polished powder show shape and size. Note spherical appearance with few exceptions and a lack of large internal voids.

—Microstructure

Previous studies of U-7Mo atomized powder have shown an equiaxed internal microstructure surrounded by columnar grains at the edge of the particles [6,7]. These studies were conducted on powder produced by a rotating disk centrifugal atomizer. In this production method a stream of molten alloy is poured onto a rapidly spinning disk which flings the metal off in fine droplets which solidify to form powder [3]. As the rotating disk atomization method relies on a certain degree of superheat (and added time needed to solidify from the superheated condition) a periphery of columnar grains is understandable in this type of powder.

Micrographs of the REP powder show a polycrystalline microstructure (figure 4) with grain sizes in the 5 μm range. This grain structure is consistent throughout the entire diameter of the fuel particle. No layer of columnar grain formation was observed, consistent with a rapidly solidified microstructure.

In REP atomization, the amount of superheated material is minimal; the highest temperature the molten alloy undergoes is only slightly over the melting point. Once the molten material is formed by the melting of the rod, centrifugal force generated by the spinning of the consumable electrode tends to fling it into the process chamber where solidification rapidly occurs.

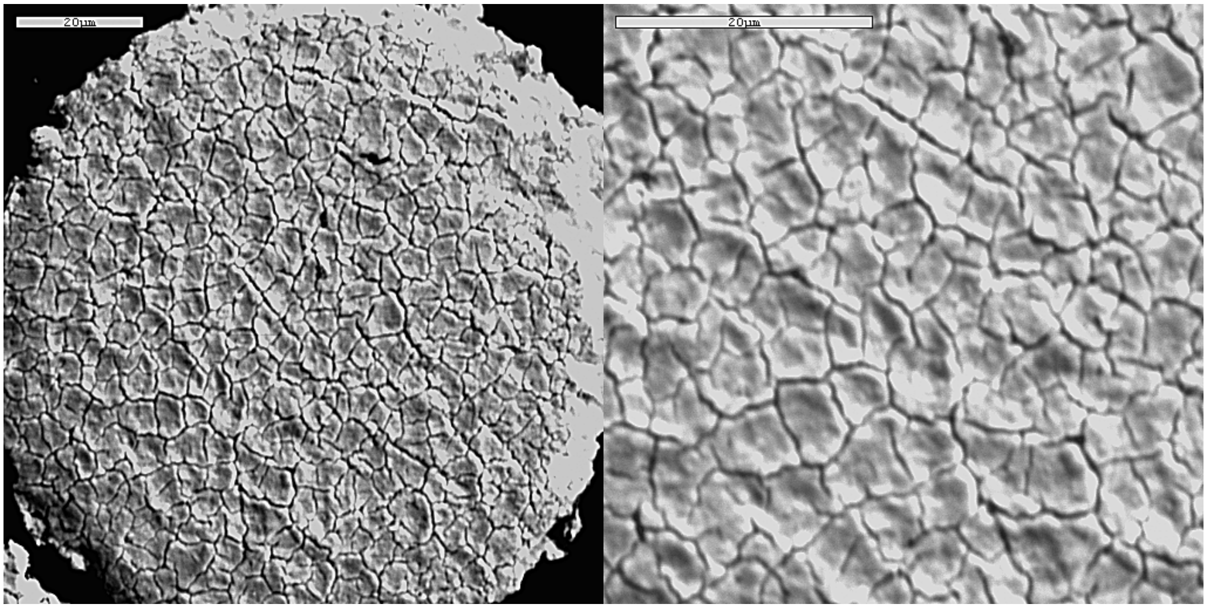

Figure 4. SEM image of an atomized powder microstructure. Images of an entire particle (left) and a magnified segment (right) show a consistent, equiaxed, polycrystalline microstructure. Material was chemically etched in a 76% H_2O, 23% HNO_3 and 1% HF solution for a few minutes.

Compositional segregation of the alloy powder was not seen in X-ray maps (figure 5). The X-ray map shows only random elemental distribution across a multi-grained particle surface. Some apparent elevated uranium concentrations around the periphery of the particles are due to edge effect—an artifact of the sample preparation. No discernable segregation was noted in SEM examinations of the grain boundary region.

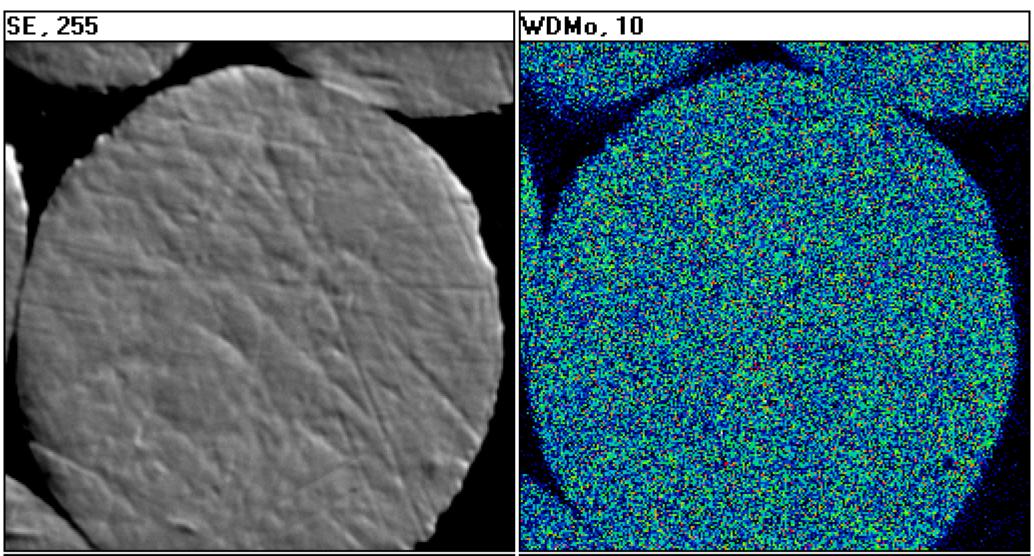

Figure 5. Secondary electron (left) and corresponding molybdenum X-ray map (right) of U-7Mo powder. Map of uranium intensity indicates no compositional segregation. Note 'fringe effects' around periphery of particles due to surface relief.

—Composition

X-ray diffraction (XRD) of a typical U-7Mo powder sample was performed using a Scintag X1 powder diffractometer (ThermoARL, Franklin, Ma) with a theta-theta goniometer configuration, and using Cu K-α radiation. The x-ray tube voltage was 45 kV with a current of 40 mA. The tube power was 1.8 kW. The scan range was from 30 to 110 2θ degrees with a scan rate of 0.5 degrees/minute. The powdered sample was mounted onto a zero-background slide as a thin film adhered with silicon grease.

The room temperature equilibrium state for the U-7Mo alloy is comprised of the α-U phase and a γ'-U_2Mo intermetallic compound [8]. The X-ray diffraction data for the U-7Mo powder produced at the INL (figure 6), shows the metastable γ-U (bcc) phase with no other phases detected. In U-7Mo at temperatures above ~560 ºC this phase is in equilibrium.

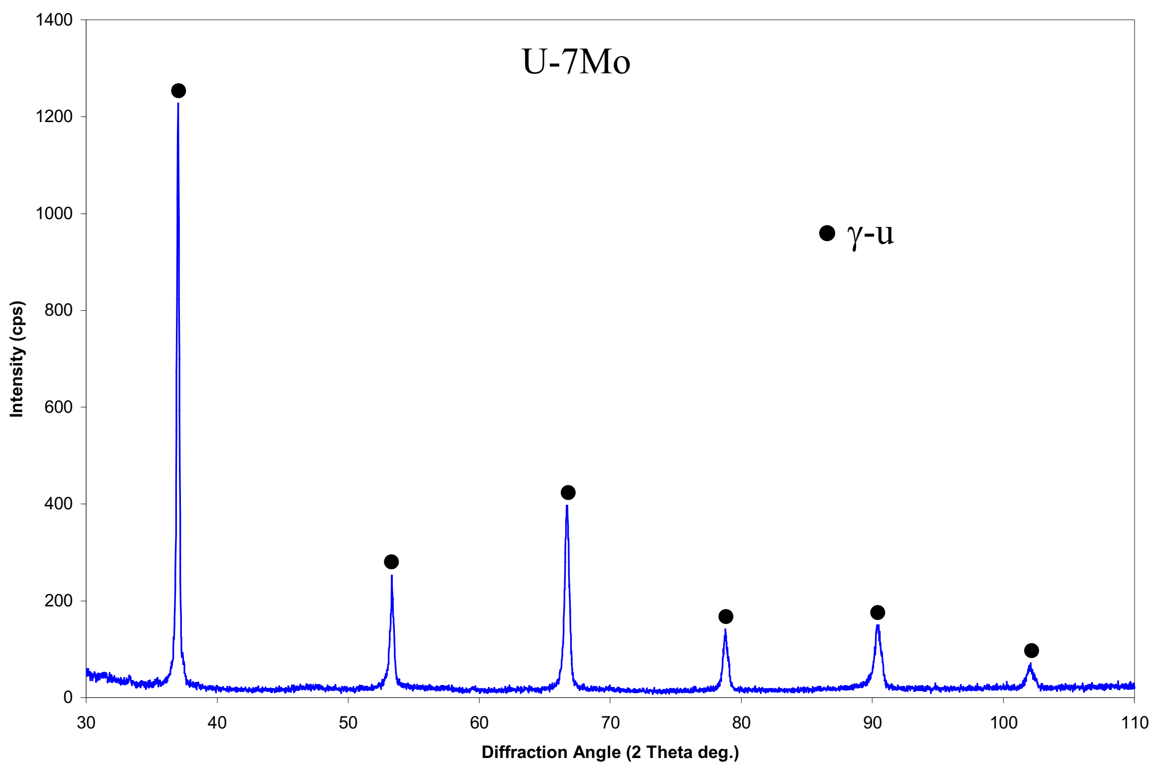

Figure 6. X-ray diffraction pattern of U-7Mo powder. The scan shows only peaks for γ-U.

4. Miniplate Fabrication and Testing

Previous RERTR fuel irradiation tests with U-Mo dispersion fuel typically suffered from problems with fuel load homogeneity. These difficulties arise in the 'dog bones'—regions of excessive fuel thickness at the ends of the fuel zone—and in the bulk homogeneity throughout the fuel plate.

Atomized powder used for the first 6 RERTR U-Mo tests was produced at the Korea Atomic Energy Research Institute (KAERI) and had a size distribution finer than powder produced at the Idaho National Laboratory (INL) for the RERTR 7 & 8 irradiation tests using the INL REP atomizer. The two RERTR-9 tests reduced the particle size threshold from 150 to 106 μm and reduced the percentage of the largest size fraction (90-106 µm) to 30 and 15% for the RERTR-9A and the RERTR-9B tests, respectively.

To reduce the dogbone pileup at the ends of the fuel zone that results in cladding thicknesses being less than required, an effort was undertaken to minimize the amount of rolling reduction that was required to fabricate a fuel plate. For the RERTR-9A experiment the reduction was cut in half from 6:1 to 3:1. This resulted in a fuel plate that was hermetically sealed but was not judged to be durable enough for reactor insertion. To improve the bonding, the plates were further processed in a hot isostatic press (HIP) at 100 MPa for 30 minutes, resulting in a dramatic improvement in the bond quality. As the plates processed in the HIP were already hermetically sealed, no process outer containment vessel was needed; the dispersion plates were merely placed in an open top steel cup to align them during the process.

To avoid the necessity of a dual process, a bonding study was implemented to examine the bonding of aluminum under various roll bonding reductions. Four experiments, each consisting of two thin plates of aluminum 6061 chemically cleaned using standard methods were welded together to form assemblies. These plates were hot rolled at reductions of 3:1, 4:1, 5:1, and 6:1. The samples were then bend tested and sectioned for metallographic analysis. The results are summarized in Table 1.

Table 1. Rolling Reduction Bonding and Minclad Studies

Reduction Ratio	Bend Test	Bonding*	Minclad†
3:1	Pass	5%	—
4:1	Pass	65%	230 µm
5:1	Pass	80%	180 µm
6:1	Pass	90%	—

* Amount of interface with intruding grain growth (50% threshold for 'bond')
† Minimum allowable minclad distance is 150 µm.

Following the aluminum bonding study a test was run to examine the effect of rolling reduction on fuel cladding thickness. Two compacts of DU-7Mo were made and welded into aluminum frames assemblies sized so that rolling reductions of 4:1 and 5:1 would result in a final thickness plate. After rolling at standard temperature and percentage reduction, the plates were examined by ultrasonic testing to obtain minimum cladding values. The resulting distance from the cladding to the fuel for the 4:1 and 5:1 reductions was 230 and 180 µm, respectively; both under the allowable minimum cladding of 150 µm.

Based on the rolling study results the reduction chosen for the RERTR-9B experiment was 4:1. To keep the total plate fuel mass unchanged while reducing the rolling reduction the fuel compacts must be thinner and larger in area. The 3:1 reduction used in the RERTR-9A was achieved by placing two compacts end-to-end in an aluminum frame. For the RERTR-9B experiment a new punch and die set was procured with the proper geometry for a 4:1 reduction (figure 7).

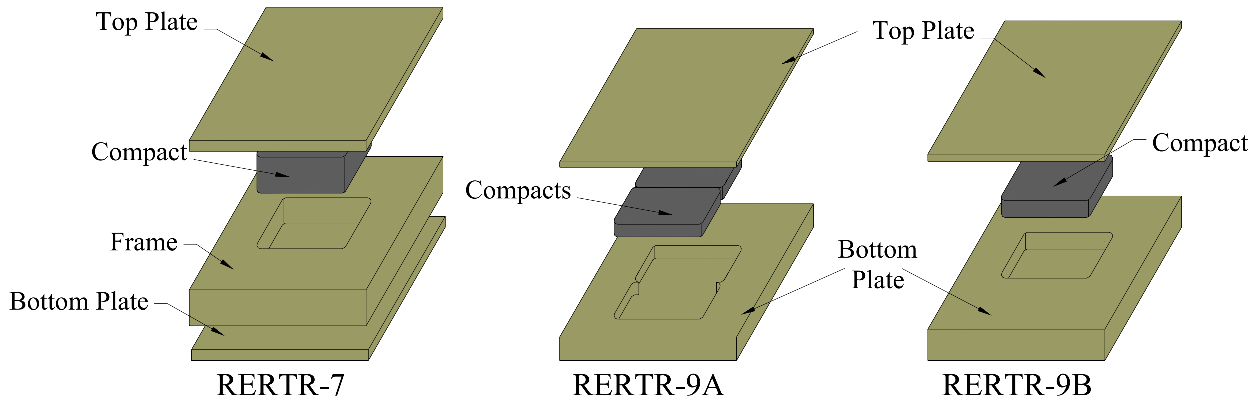

Figure 7. Exploded view of dispersion hardware assemblies used for various rolling reductions used in RERTR miniplate irradiations.

Another change implemented in the design of the RERTR-9B experiment was the reduction of the nominal fuel meat thickness from 635 to 505 µm. The thickness was reduced to allow the use of uniform uranium enrichment throughout the experiment (the monolithic fuel plates use 58 % ^{235}U for both iterations of the RERTR-9 irradiation tests) and to allow for a thicker cladding surface. This thickness reduction, along with the lower rolling reduction, served to increase the distance from the fuel meat to the surface of the cladding. As this fuel thickness reduction lowers the areal density the enrichment of the dispersion fuel plates was increased from 44 to 58 % ^{235}U to maintain a nominally identical surface heat flux (table 2).

Table 2. Comparison of Fabrication Parameters in the RERTR-9 Experiment

Parameter	**RERTR-9A**	**RERTR-9B**
Fuel Alloy	U-7Mo	U-7Mo
Large Fuel Powder Fraction (>90 µm)	30%	15%
Matrix Alloys	Al-2Si Alloy Al+2Si Blend* 4043 Alloy	Al-2Si Alloy Al-3.5Si Alloy
Nominal Fuel Loading (gU/cm^3)	8.0	8.5
Reduction Ratio	3:1	4:1
Bonding Method	Roll + HIP	Roll
Fuel Meat Thickness (µm)	635	505
Enrichment	44% U^{235}	60% U^{235}

* The blended matrix was comprised of pure Al and Si powders mixed together

A total of ten dispersion fuel plates are included in the two phases of the RERTR-9 fuel plate irradiation test. The specific experimental matrix loading for the RERTR-9 Tests is shown in Table 3. The RERTR 9 irradiation experiment was designed to further test advanced concepts in dispersion and monolithic fuel. In dispersion fuel the silicon-doped aluminum matrix material was to be tested with higher fuel loadings at higher burnup and heat flux. The target values for these tests were 8 to 8.5 gU/cm^3 loading, 320 W/cm^3 surface heat flux, and an end-of-life burnup (% ^{235}U) of 75%*

* LEU (20% ^{235}U) equivalent

Table 3. RERTR-9 Irradiation Test Matrix—Dispersion Plates Highlighted

A1*	A2	A3	A4
Blank	U-7Mo 8 g-U/cc Al-4043 Roll + HIP	Blank	U-10Mo Monolithic Plain Foil Friction Bond
A5	A6	A7	A8
U-10Mo Monolithic Plain Foil Friction Bonded	U-10Mo Monolithic Al-4043 Interlayer HIP	U-7Mo 8 g-U/cc Al + 2 Si Roll + HIP	U-10Mo Monolithic Plain Foil Friction Bond
B1	B2	B3	B4
U-10Mo Monolithic Al-Si 125 µ TS† Friction Bond	U-10Mo Monolithic Plain Foil Friction Bond	U-10Mo Monolithic Al-4043 Interlayer HIP	U-10Mo Monolithic Al-Si 250 µ TS Friction Bond
B5	B6	B7	B8
U-10Mo Monolithic Zr Interlayer HIP	U-7Mo 8.5 g-U/cc Al-2Si Roll	U-7Mo 8.5 g-U/cc Al-3.5Si Roll	U-10Mo Monolithic Al-Si 125 µ TS HIP
C1	C2	C3	C4
U-10Mo Monolithic Plain Foil Friction Bonded	U-7Mo 8 g-U/cc Al-4043 Matrix Roll + HIP	U-7Mo 8 g-U/cc Al-2 Si Matrix Roll + HIP	U-10Mo Monolithic Plain Foil Friction Bond
C5	C6	C7	C8
U-10Mo Monolithic Plain Foil Friction Bonded	U-10Mo Monolithic Al-4043 Interlayer HIP	U-7Mo 8 g-U/cc Al + 2 Si Matrix Roll + HIP	U-10Mo Monolithic Plain Foil Friction Bond
D1	D2	D3	D4
U-10Mo Monolithic Al-Si 125 µ TS Friction Bond	U-7Mo 8.5 g-U/cc Al-3.5Si Roll	U-10Mo Monolithic Al-4043 Interlayer HIP	U-10Mo Monolithic Al-Si 250 µ TS Friction Bond
D5	D6	D7	D8
U-10Mo Monolithic Zr Interlayer HIP	U-7Mo 8.5 g-U/cc Al-2Si Roll	U-7Mo 8.5 g-U/cc Al-3.5Si Roll	U-10Mo Monolithic Al-Si 125 µ TS HIP

* Capsules A & C comprise the 9A experiment, B & D the 9B experiment
† Thermal Sprayed Al-Si alloy

5. Acknowledgment

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