High-Density, Low-Enriched Uranium Fuel for Nuclear Research Reactors

D.D. Keiser, Jr., S.L. Hayes, M.K. Meyer, and C.R. Clark

The objective of the Reduced Enrichment for Research and Test Reactor program is to enable the conversion of test reactors using fuel containing high-enriched uranium to fuel that employs low-enriched uranium. This paper describes the development of this low-enriched fuel, including fabrication, characterization, and testing of the fuel.

INTRODUCTION

Dispersion fuels are composite fuels consisting of a collection of fissile-bearing particles dispersed in a neutronically inert matrix that is typically aluminum due to its high thermal conductivity. High-energy fission events are thus isolated from the matrix, preventing fission-induced degradation of material properties. Dispersion fuels are very robust, allowing high burnup (i.e., the amount of heavy atoms that are fissioned) at high power densities and enabling the production of the high neutron flux desirable for research and test reactor operation.[1] Many of these reactors use uranium with U-235 enrichments greater than 20%, and it is desirable to convert these reactors to the use of uranium at lower enrichments.

The U.S. Reduced Enrichment for Research and Test Reactors (RERTR) program was created in 1978.[2] The mission of this program is to develop technologies to convert research and test reactors from the use of high-enriched uranium (HEU) (i.e., U-235 content greater than 20%) to low-enriched uranium (LEU) (i.e., U-235 content less than 20%), consistent with a U.S. policy to eventually eliminate the use of HEU in civilian nuclear programs worldwide. As uranium enrichment is decreased, uranium density in the fuel must increase to maintain the net fissile (U-235) atom density of the fuel. Over the last two decades, fuels with increasing uranium density have been developed to allow conversion of reactors with higher fissile density requirements.

The highest uranium density fuel currently licensed by the U.S. Nuclear Regulatory Commission is based on a dispersion of uranium-silicide (U_3Si_2) fissile particles at 4,800 kg U/m^3. A uranium density of 8,000–9,000 kg/m^3 in the dispersion is required to convert almost all remaining reactors. Since the conventional roll-bonding fabrication process for research reactor dispersion fuel plates is limited to fuel particle volume loadings of ~55% in the meat region, a uranium density above 14.5 g/cm^3 in the fuel particle is needed to achieve the meat-averaged uranium density target. Table I shows the candidate fuels that have been irradiated along with their uranium density. Only two types of fuel are suitable for these very high-density fuels: metallic uranium of low alloy content and the U_6Metal class of high-density intermetallics. Irradiation testing of U_6Fe and U_6Mn intermetallic fuels ruled out the use of these and other like high-density intermetallic fuels due to break-away

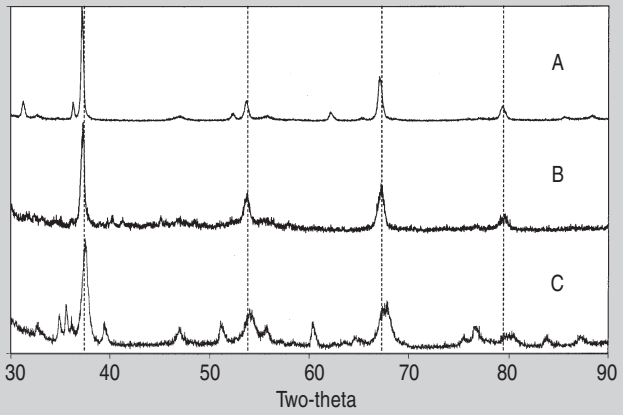

Figure 1. X-ray diffraction patterns of U-10Mo alloy fuel particles removed from as-fabricated fuel plates by dissolution in NaOH (aq.). Dashed lines show major γ-U peaks. (a) Fuel particles made by machining followed by γ-annealing are predominantly γ-phase. (b) Atomized powder. (c) As-machined particles showing extensive decomposition from the γ-phase.

Table I. Candidate Dispersion Fuel Particle Materials

Candidate Fuels	Uranium Density (g/cm^3)
UAl_3	5.1
UO_2	9.7
U_3Si_2*	11.3
U_2Mo*	13.8
U-9Nb-3Zr*	14.2
U_3Si	14.7
U-6Nb-4Zr*	14.8
U-5Nb-3Zr*	15.5
U-10Mo*	15.3
U-10Mo-0.05Sn*	15.3
U-8Mo*	16.0
U-7Mo*	16.4
U-6Mo*	16.7
U_6Fe	16.7
U-6Mo-0.6Ru*	16.5
U-6Mo-1Pt*	16.5
U-6Mo-1.7Os*	16.4
U-4Mo*	17.4
U	19.0

* Alloys tested for development of very-high-density fuels.

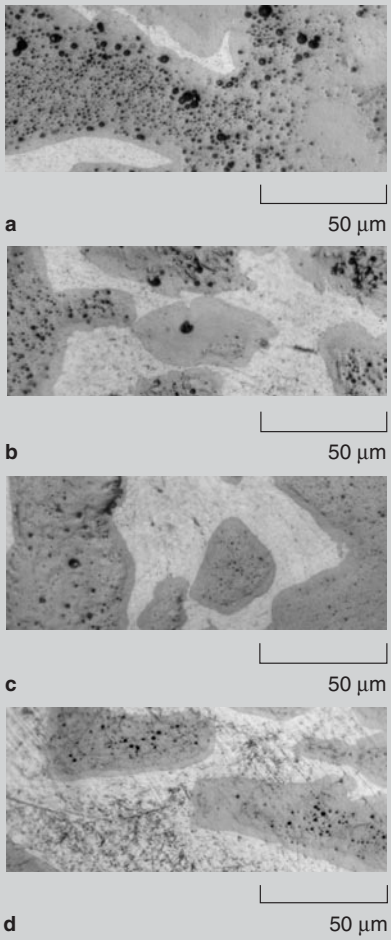

Figure 2. RERTR-2 fuels irradiated at low temperature to 70% burnup: (a) U-4Mo, (b) U-6Mo, (c) U-8Mo, and (d) U-10Mo.

swelling during irradiation, leaving alloys as the only solution.[3–5] Since it is known that γ-phase uranium alloys exhibit superior irradiation performance relative to α-phase uranium,[6] development was focused on this alloy class. Irradiation testing of various γ-stabilized alloys, including U-Mo, U-Mo-Pt, U-Mo-Ru, U-Mo-Sn, and U-Nb-Zr alloys showed that U-Mo-based alloys are the best alloy fuel candidates.[4,7] To adequately demonstrate the irradiation performance of the U-Mo alloys, a full demonstration effort is underway consisting of fabrication of the fuel, irradiation testing, characterization of the fuel before and after irradiation testing, and out-of-reactor testing.

IRRADIATION TESTING

Fourteen different fuel compositions, including 12 metallic alloys, have been irradiated as part of five experiments for alloy dispersion fuel development in the Idaho National Engineering & Environmental Laboratory's Advanced Test Reactor (ATR) up to burnups of 80% of U-235.[9–12]

Table I lists the fuel particle compositions contained in these tests, and Table II provides an overview of the five alloy dispersion irradiation experiments conducted during the very-high-density fuel development effort. The RERTR-1 and RERTR-2 experiments were identical in design, and the experimental fuel test matrices differed only for a few fuel plates; the principal difference in the two experiments was in test duration, with RERTR-1 being discharged at low burnup and RERTR-2 continuing irradiation to much higher burnup. The RERTR-4 and RERTR-5 experiments were similar but were irradiated to different burnup levels.

Table II. Overview of RERTR Very-High-Density Fuel Irradiation Testing Program

	Irradiation Experiment				
Test Parameter	**RERTR-1**	**RERTR-2**	**RERTR-3**	**RERTR-4**	**RERTR-5**
Reactor, position	ATR, I-22	ATR, I-23	ATR, B-7	ATR, B-12	ATR, B-11
Number of experimental plates	32	32	47	32	32
U-Mo powder fabrication†	Gr, At	Gr, At	At	Gr, At, Mn	Gr, At
Fuel meat loading (g-U/cm^3)	4.0	4.0	8.5	6.0, 8.0	6.0, 8.0
Enrichment (% ^{235}U)	19.5	19.5	19.5	19.5	19.5
Maximum heat flux (W/cm^2)	70	55	400	210*	320*
BOL cladding temperature (°C)	65	65	150	130*	175*
Peak fuel temperature (°C)	70	75	235	>200*	>200*
Coolant velocity (cm/s)	920	920	600	400	400
Status of experiment	Complete	Complete	Complete	In PIE	In PIE
Number of irradiation cycles	2	8	2	7	3
Irradiation duration (days)	94	232	48	230	116
Burnup at EOL (% ^{235}U)	40	70	40	80	50

† Metallic powder prepared by grinding (Gr) or atomization (At); (Mn) denotes monolithic fuel meat.
* Significant uncertainty in power at experimental position.

CONDITIONS

Fuel Fabrication

U-Mo dispersion fuels have been fabricated using atomized or mechanically ground U-Mo powders. These mechanically ground powders contained 4 wt.%, 6 wt.%, 8 wt.%, or 10 wt.% Mo. The ground U-Mo powders were produced by filing or machining U-Mo alloy rods. The atomized powders were made using a spinning disk melt atomization process.[8] To fabricate the U-Mo powders into dispersion fuels, commercially practiced methods are employed. The ground powders are blended with aluminum powder and pressed into pellets using a die, while atomized powders are blended with aluminum and either hot extruded or compacted in a die. The resulting rods are sectioned into pellets. In turn, these pellets are encased in aluminum-6061 frames, roll-bonded at ~500°C and reductions in cross-sectional area of 5–15, and sheared into final plate geometry.

Pre-Irradiation Characterization

Prior to fuel fabrication, U-Mo alloys are typically heat treated in the range of 800°C to 900°C to form the γ-(U,Mo) solid solution phase. The γ-(U,Mo) phase transforms eutectoidally at 565°C into an orthorhombic α-U phase and the ordered tetragonal γ′ phase, which has the nominal stoichiometry of U_2Mo. This transformation becomes very sluggish when molybdenum contents are greater than about 6 wt.%, and the alloy can be easily quenched into the γ-phase and remain in the γ-phase indefinitely at room temperature. During fuel plate fabrication at ~500°C, some alloys have been observed to react with the aluminum matrix more than others. This can be linked to two effects. First, alloy particle microstructures that contain α-U were found to react at an accelerated rate. After fuel fabrication, fuel particles produced by filing or machining were partially decomposed, thereby containing the α-U phase, while the atomized powders were not. Strain induced by grinding or machining apparently accelerates the eutectoid decomposition. Subjecting fuel particles to γ-annealing subsequent to powder formation and prior to plate fabrication minimizes this behavior. Second, the fuel/aluminum reaction rate is sensitive to alloy content. For fuel particles with 6 wt.% or less molybdenum content, out-of-reactor reaction occurs faster than in fuels with higher alloy content.

RERTR-1 and RERTR-2

These experiments were the first attempts by the RERTR program to irradiate high-density metallic alloy fuels dispersed in an aluminum matrix. The experiments were scoping in nature with the intent of investigating the feasibility of the various fuel system candidates under consideration, so the experimental fuel plates were fabricated with fuel particle loadings of only 25–30 vol.% in the meat, giving meat-averaged uranium densities of ~4 g/cm^3. The focus of these experiments was to observe the phenomena of fuel-matrix interaction and fuel particle swelling under irradiation. Fuel plate powers, and consequently temperatures, were maintained low.

Sixty-four miniature fuel plates were fabricated using a variety of uranium alloys based on the U-Nb-Zr and U-Mo systems and irradiated to U-235 burnup levels of 40% and 70%. Post-irradiation examinations (PIE) of these fuel plates revealed poor performance of the U-Nb-Zr fuel alloys; fuel plates fabricated with these alloys exhibited large fuel plate thickness increases caused by both extensive fuel-matrix reaction and incipient breakaway swelling of the fuel particles themselves.[5] Fuel plates fabricated with the U-4Mo alloy showed similar poor behavior. The U-Mo alloys fabricated with at least 6 wt.% Mo, however, performed quite well up to 70% burnup; as seen in Figures 2 and 3, fuel-matrix interaction was minimal (<5 μm), and stable fission gas bubble growth resulting in low fuel swelling was observed within the metallic fuel particles.[13] The U-Mo alloys with ternary additions (Ru, Pt, Os), known to enhance the stability of the cubic γ-U phase out-of-pile, showed no significant improvement over the binary alloys under irradiation.

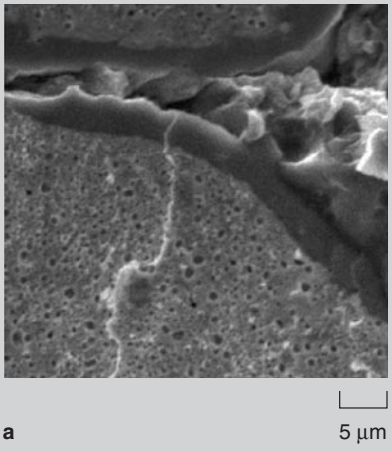

Figure 3. Scanning electron micrographs of irradiated U-10Mo fuel from RERTR-2 at 69% burnup.

RERTR-3

The RERTR-3 experiment was designed to test experimental fuel plates under irradiation conditions considered aggressive for research reactor fuels. Forty-seven miniature fuel plates were fabricated and irradiated to a U-235 burnup level of 40%. Based on the results of the RERTR-1 and -2 experiments, the RERTR-3 experiment focused principally on the U-Mo binary alloy fuels with 6 ≤ Mo ≤ 10 wt.%. In this experiment the test fuels were fabricated with fuel particle loadings of over 50 vol.% in the meat, giving meat-averaged uranium densities of up to 8.5 g/cm^3; metallographic cross sections of these highly-loaded dispersion fuel plates are shown in Figure 4.

Post-irradiation examinations of these fuel plates showed generally acceptable fuel performance. Fuel swelling was relatively low, with no tendency toward breakaway behavior apparent from microscopy. However, at the elevated fuel temperatures of this experiment, significant fuel-matrix interaction was observed, as seen in Figure 5. In fact, fuel-matrix interaction was so extensive that no matrix aluminum remained in the hot central portion of the fuel meat in some fuel plates (e.g., Figure 5c). Nonetheless, acceptable fuel plate performance was achieved even in cases where all of the matrix aluminum phase was consumed.

RERTR-4 and RERTR-5

The RERTR-4 and -5 experiments were designed to test larger fuel plates under prototypic research reactor conditions. Each experiment contained 32 fuel plates irradiated to U-235 burnup levels of 50% and 80%. These experiments

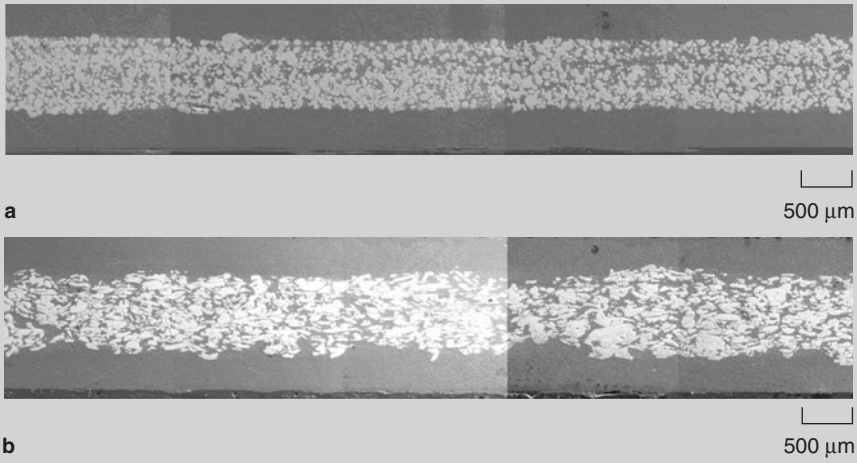

Figure 4. Metallographic cross sections of as-fabricated U-10Mo/Al fuel plates loaded to 8.5 g-U/cm^3 in the fuel meat; (a) atomized fuel powder and (b) machined fuel powder.

continued to focus on the U-Mo binary alloy fuels with $6 \leq Mo \leq 10$ wt.%. However, the RERTR-4 experiment included two fuel plates of an innovative U-10Mo monolithic fuel design. The monolithic fuel design is not a dispersion fuel, but rather an aluminum-alloy-clad U-10Mo foil. This design is attractive since meat-averaged uranium densities much higher than 8 g/cm^3 are possible, and fuel-matrix interaction is mitigated by the small aluminum-to-fuel contact area of the foil.

The irradiation of these experiments was completed recently, and PIE is still in progress. However, preliminary results indicate all fuel plates performed in an acceptable manner. The fuel plate of the U-10Mo monolithic design showed promising performance.

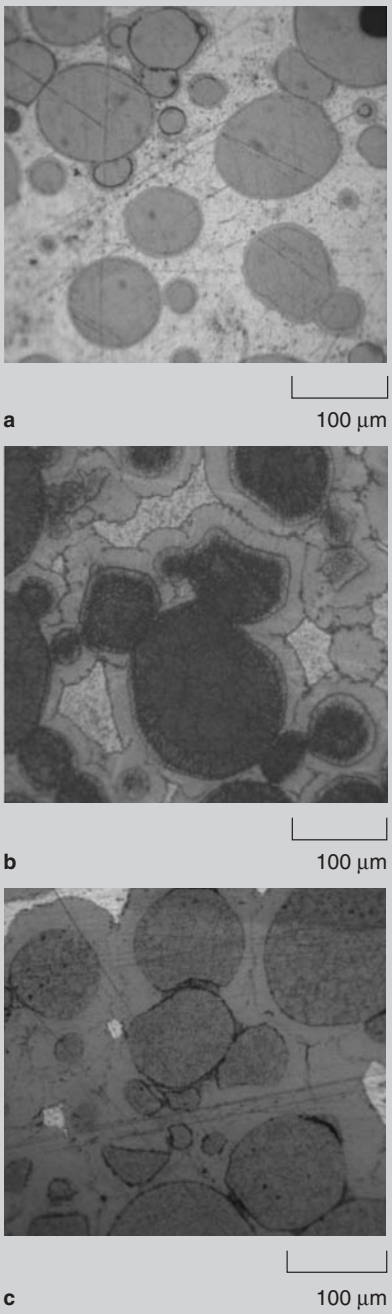

Figure 5. Increasing U-Mo/Al fuel-matrix interaction with temperature seen in metallography of RERTR-3 fuels; (a) U-10Mo at 139°C and 30% burnup, (b) U-10Mo at 203°C and 38% burnup, and (c) U-6Mo at 224°C and 40% burnup.

EX-REACTOR TESTING

As described, interdiffusion between the U-Mo alloy fuel particles and the aluminum matrix that occurs during irradiation produces uranium aluminides. These aluminides have lower thermal conductivities than the original aluminum resulting in higher fuel operating temperatures. Understanding what types of aluminides develop as a result of the interaction between the U-Mo alloy and aluminum is useful for modeling the behavior of the fuel with respect to aluminum depletion and its effect on fuel temperature.

Diffusion couples have been constructed between U-7 wt.% Mo alloy and aluminum and annealed at various temperatures to investigate the interdiffusion behavior of the elemental components.[14] The results from the diffusion experiments show that $(U,Mo)Al_3$ is the major intermetallic phase that forms out-of-reactor due to the interdiffusion of uranium, molybdenum, and aluminum. Changes occur in the types of intermetallics that form, in the amount of interdiffusion that is observed, and in the morphology of the diffusion zones when the original γ-phase U-Mo alloy decomposes into α-U and U_2Mo phases during the annealing treatment.

CONCLUSIONS

Based on the test results described in this paper, U-Mo/Al dispersion fuel promises to be a viable candidate for powering many of the world's research reactors using low-enrichment uranium. The U.S. RERTR program is continuing work toward the qualification of this fuel for use.

ACKNOWLEDGEMENT

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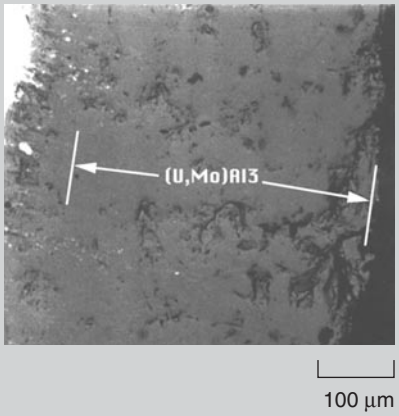

Figure 6. A backscattered electron image of the interdiffusion zone that develops when a U-7 wt.% Mo versus aluminum diffusion couple is annealed at 575°C for 50 h. The fuel is the bright-contrast area to the left, and the aluminum is the darkest area to the right. The bulk of the interdiffusion zone is comprised of the $(U,Mo)Al_3$ phase.

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