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Abstract

Twelve different uranium alloys and compounds with uranium densities greater than 13.8 g/cc were fabricated into fuel plates. Sixty-four experimental fuel plates, referred to as microplates, with overall dimensions of 76.2 mm x 22.2 mm x 1.3 mm and elliptical fuel zone of nominal dimensions of 51 mm x 9.5 mm, began irradiation in the Advanced Test Reactor on August 23, 1997. The fuel test matrix consists of machined or comminuted (compositions are in weight %) U-10Mo, U-8Mo, U-6Mo, U-4Mo, U-9Nb-3Zr, U-6Nb-4Zr, U-5Nb-3Zr, U-6Mo-1Pt, U-6Mo-0.6 Ru, U-10Mo-0.05Sn, U_2Mo and U_3Si_2 (as a control). The low enriched (^{235}U<20%) fuel materials were cast, powdered, mixed with aluminum dispersant at a volume ratio of 1:3, compacted and hot rolled to form the microplates. Spherical atomized powders of two fuels, U-10Mo and U_3Si_2, were utilized to make microplates and included in the irradiation test as well. The experimental design and fabrication steps employed in the selection and production of the fueled microplates is discussed.

Introduction

The goal of our advanced fuel development effort is to produce a new low enriched uranium (LEU, ^{235}U<20%) fuel with a uranium density of 8 to 9 g/cc in the fuel meat [1]. Two approaches can be used to accomplish this goal, increasing the uranium content of the fuel and increasing the amount of fuel dispersasnt in the fuel meat. The latter approach is a matter of process optimization after a fuel material has been determined. Thus, the purpose of this irradiation experiment is to ascertain which fuel candidates are suitable for further fabrication development.

CERCA's experience with silicide fuel [2] indicates that the maximum fuel meat loading is around 55 volume per cent fuel dispersant. Consequently, viable candidate fuel materials must have a uranium density of about 14.5 g/cc. There are some uranium compounds which meet this requirement, but they either have demonstrated or are expected to undergo excessive swelling during irradiation [1]. Alternatively, uranium-rich alloys, which have higher uranium densities, could be employed. Alloy additions, which retain the body-centered gamma phase at low temperatures in uranium, appear to suppress fuel growth [3] under fast reactor conditions. Data from [4] indicate that U-Mo alloys not only retained the metastable gamma phase during irradiation but that the gamma phase forms preferentially during irradiation. Unfortunately the irradiation experiment in [4] was terminated at very low burnups, less than 0.1 atomic percent. Alloy additions, which promote the formation and stability of the gamma phase in

uranium, are Mo and Nb [5]. Ternary additions of Zr to U-Nb increase the compositional range of gamma stability more so than Nb alone [6] thus the major alloy families investigated are U-Mo and U-Nb-Zr. Further details on the physical metallurgical states of the actual alloys utilized in the irradiation in given by [7]. Using the two guidelines, (1) uranium density in the fuel alloy of at least 14.5 g/cc and (2) predicted gamma phase stability, the candidate fuel alloys presented in Table 1. U-9Nb-3Zr was added to gain a more complete understanding of the U-Nb-Zr system. The intermetallic U_2Mo was included because it is an equilibrium low temperature phase and a decomposition product of gamma in the U-Mo system. U_3Si_2 was included as a control.

Table 1. Densities of candidate fuel materials (as-cast), measured by the immersion density method.

Alloy (in weight %)	Measured Density (g/cc)	2σ	U Density (g/cc)
U-10Mo	16.900	0.008	15.2
U-10Mo-0.05Sn	16.775	0.010	15.1
U-8Mo	16.985	0.005	15.6
U-6Mo	17.604	0.002	16.5
U-6Mo-0.6Ru	17.476	0.004	16.3
U-6Mo-1.0Pt	17.530	0.002	16.3
U-4Mo	17.960	0.005	17.2
U-2Mo-1Nb-1Zr	17.822	0.006	17.1
U-9Nb-3Zr	15.904	0.025	14.0
U-6Nb-4Zr	16.158	0.008	14.5
U-5Nb-3Zr	16.588	0.011	15.3
U_2Mo	16.6*	---	13.81
U_3Si_2	12.2*	---	11.31

*From [8]

Design

The maximum fuel temperature during irradiation was set at 150°C (see [9] for further details). That criterion coupled with the desire to have a maximum burnup rate, resulted in the selection of a 38 mm diameter hole in Advanced Test Reactor (ATR) for the irradiation experiments. Miniplates, which were extensively used in the past, are 76 mm wide and thus too large for the selected experimental irradiation positions ATR. This necessitated a new fuel plate design. The redesigned fuel plate, referred to as a microplate, is illustrated in Figure 1. Analysis of the microplate confirms that the reduced geometry allows for fuel swelling under a hydrostatic stress state [10]. Other guidelines used in the design of the microplates were ease of fabrication and use of off the shelf tooling. Therefore, the fuel loading was kept low, limited to 25 volume percent and the initial compacts were round (9.5 mm OD) which results in the elliptical fuel zone in Figure 1. Another advantage of the microplate design is that the small fuel meat requires approximately 10 times less fuel by weight than a miniplate. This enabled us to fabricate this extensive test matrix in a very short time period. A maximum fuel zone was defined in order to ease the fabrication process. The microplates were fit into a "clam shell" type holder (Figure 2), welded and stacked along with two spacers into the irradiation vehicle [9].

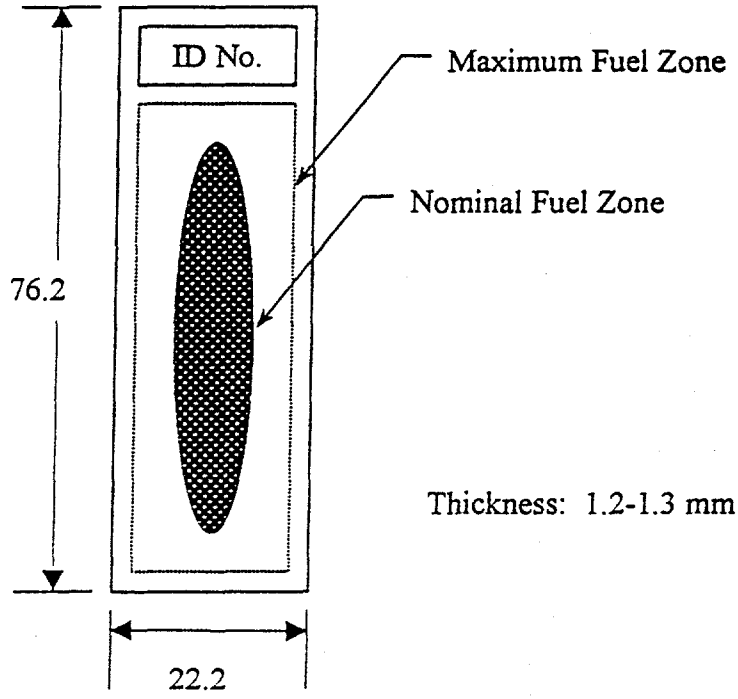

Figure 1. The redesigned fuel plate or microplate. All dimensions are in millimeters.

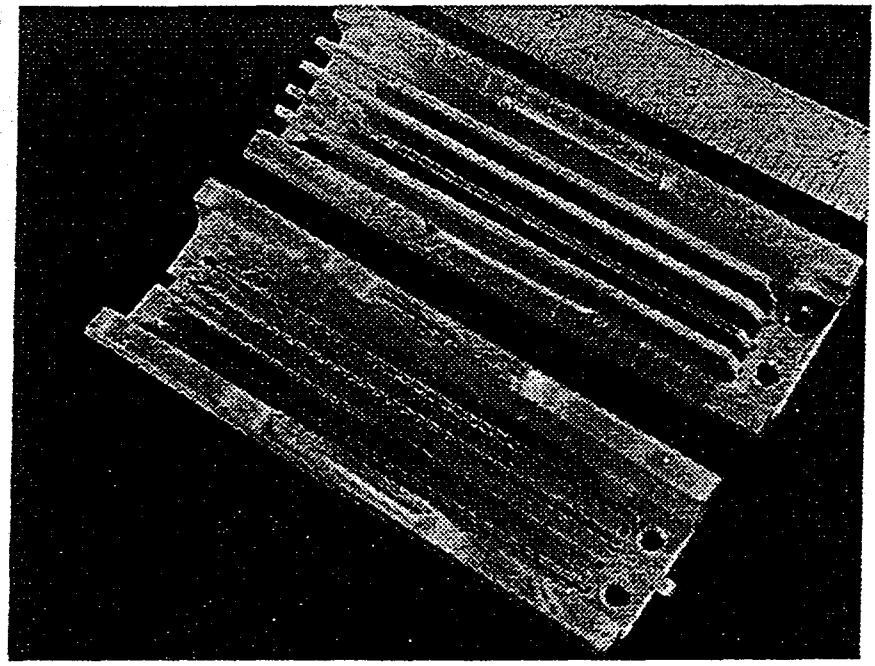

Figure 2. Microplates loaded in the capsule prior to final assembly and welding.

Fabrication

The overall fabrication scheme is shown in Figure 3. Elemental metals were combined and melted by arc melting (U_2Mo and U_3Si_2) or injection casting into quartz molds. The injection cast rods were heat treated in vacuum for 90+ hours at 900°C to ensure chemical homogeneity. All alloys formed gamma as the majority phase with the exception of U-2Mo-1-Nb-1Zr, which was alpha and gamma [7]. Sections of the heat treated rod were weighed in air and in water plus a surfactant (5 times) and a density was calculated (Table 1). X-ray diffraction (XRD) and scanning electron microscopy (SEM) and energy dispersive spectroscopy (EDS) was performed to access the microstructural state [7].

Powder making proceeded by comminution (U_2Mo and U_3Si_2) [11], machining or atomization [12] (U-10Mo and U_3Si_2). Initially, millimeter size chips of alloy were machined and used as feed for cryogenic milling. The coarse chips were milled under liquid argon in a hardened steel mill vial to reduce particle size. After each milling cycle, -212 μm powder was removed, the charge was adjusted with additional coarse chips, and another milling cycle was initiated. Milling efficiency ranged from 5% to 20% per cycle of -212 μm) powder. The powder produced by this method had a plate-like morphology (Figure 4b). The powder was found to have high iron content: about thirty-five weight percent of the uranium alloy powder could be extracted using magnetic separation. A close-up of the powder surface reveals dark spots on the powder surface (Figure 4d) which EDS determined to be iron–rich.

Due to low efficiency and high iron content, the cryogenic milling process was abandoned in favor of direct machining of powder to the proper size range using a high-speed lathe equipped with a rotary file. Fuel alloy rods were fed into a titanium nitride coated tungsten/tantalum carbide tool turning at approximately 2,500 rpm. The SEM images on the left side of Figure 4 illustrate powder fabricated by this machining process. Useable powder was produced at the rate of about 1-2 grams per hour. Powder yield ranged from 32%-63% of -212 μm powder, depending on the alloy. This process had the drawback of introducing carbide and nitride contamination due to wear of the rotary file. The presence of these type of contaminates are not expected to effect irradiation performance because they are chemically inert and thermally stable. Contamination levels ranged from 0.1–7.6 wt.% and were generally higher for uranium alloys with the highest alloy contents. In general the carbides appeared as separate individual particles. EDS identified few nitrides, an example of TiN is shown in Figure 4c. The resulting powders were flake-like (Figure 4a) and retain striation marks (Figure 4c) from the machining process. All powders except for U-2Mo-1Nb-1Zr displayed gamma as the major phase by XRD [7]. The machining and compacting operations took place in a glove box under an inert atmosphere.

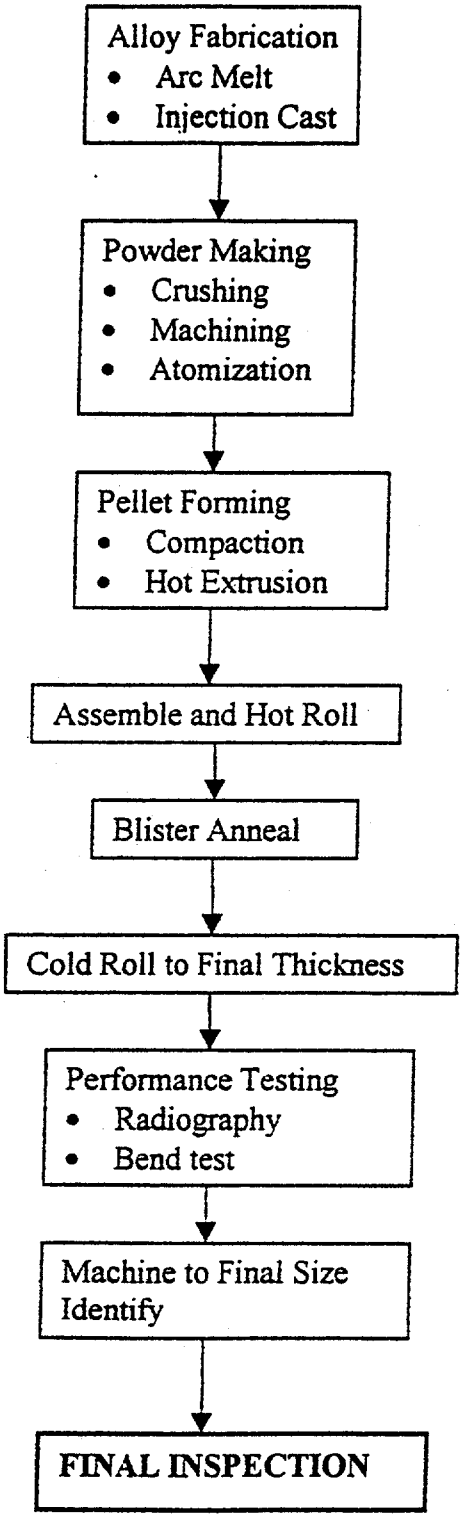

Figure 3. Process flow diagram of the fuel fabrication process

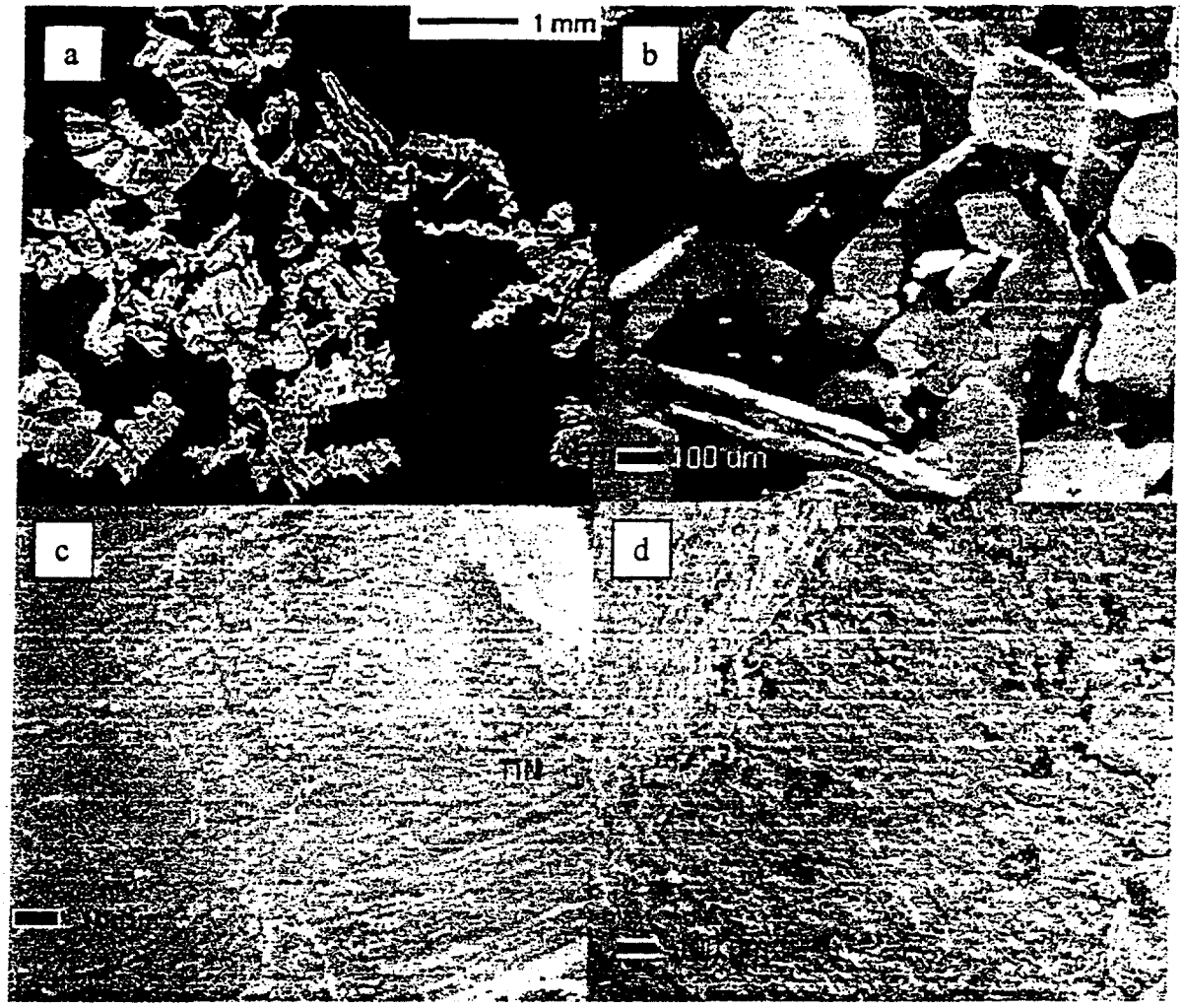

Figure 4. SEM images of as-produced powders; a) U-2Mo-1Nb-1Zr as-machined via high speed rotary file, b) U-10Mo as-cryogenically milled, c) surface of as-machined U-5Nb-3Zr, note TiN embedded in the surface and residual striation marks, and d) surface of cryogenically milled U-10Mo illustrating iron contamination.

Compacts were prepared by mixing appropriate masses of degassed aluminum powder (Alcan 101) with fuel powders less than 212 μm to form 25 vol.% compacts. Nominal compact dimensions were 9.5 mm diameter by 2.2 mm thick. Centrifugal atomized powers were produced [12] and extruded into rod by the Korea Atomic Energy Research Institute (KAERI). Fuel powders less than 125 μm mixed with aluminum powder to form a 25 vol.% compact. Compacts were extruded at 400°C and areal reduction ratio of 17.7:1 employing a containerless technique in an inert atmosphere [13]. The resulting 9.5 mm diameter rods were sliced to the same thickness as the powder compacts.

All compacts and extruded fuel meats were loaded into aluminum (6061) picture frames 89 mm by 152 mm and 2.2 mm thick with eight holes in an asymmetric array. The frames and cover plates were cleaned, assembled and welded in the conventional

manner [11]. Rolling was done with the assemblies preheated to 500°C. Multiple rolling passes were performed hot. Height reductions were nominally 25% for the initial 2 passes and 20% thereafter. Blister annealing, to assure bonding between the fuel meat and aluminum clad has been achieved, was done after hot rolling for 50 ± 10 minutes at 485 ± 15°C. None of the microplates displayed any blistering after the blister anneals. However, some alloys showed signs of bulging over the entire fuel zone. Metallographic sectioning of these plates indicated that the fuel material had significant interaction with the aluminum [6,14]. This led to the elimination of U-2Mo-1Nb-1Zr from the irradiation experiment and limited the U-4Mo and the U-5Nb-3Zr to a short irradiation schedule. Rolling to final thickness was done cold and limited to 10% reduction. Typical fuel meat sections from fully processed microplates of the mechanically prepared and atomized powders are illustrated in Figure 5. Fuel meats prepared by mechanical methods (Figure 5a and 5b) appear remarkably similar in spite of the different powder production methods. The atomized powder in fuel meats in Figure 5c and 5d retain their spherical shape even after extensive rolling. Silicide fuel particles appear cracked and broken regardless of the processing method.

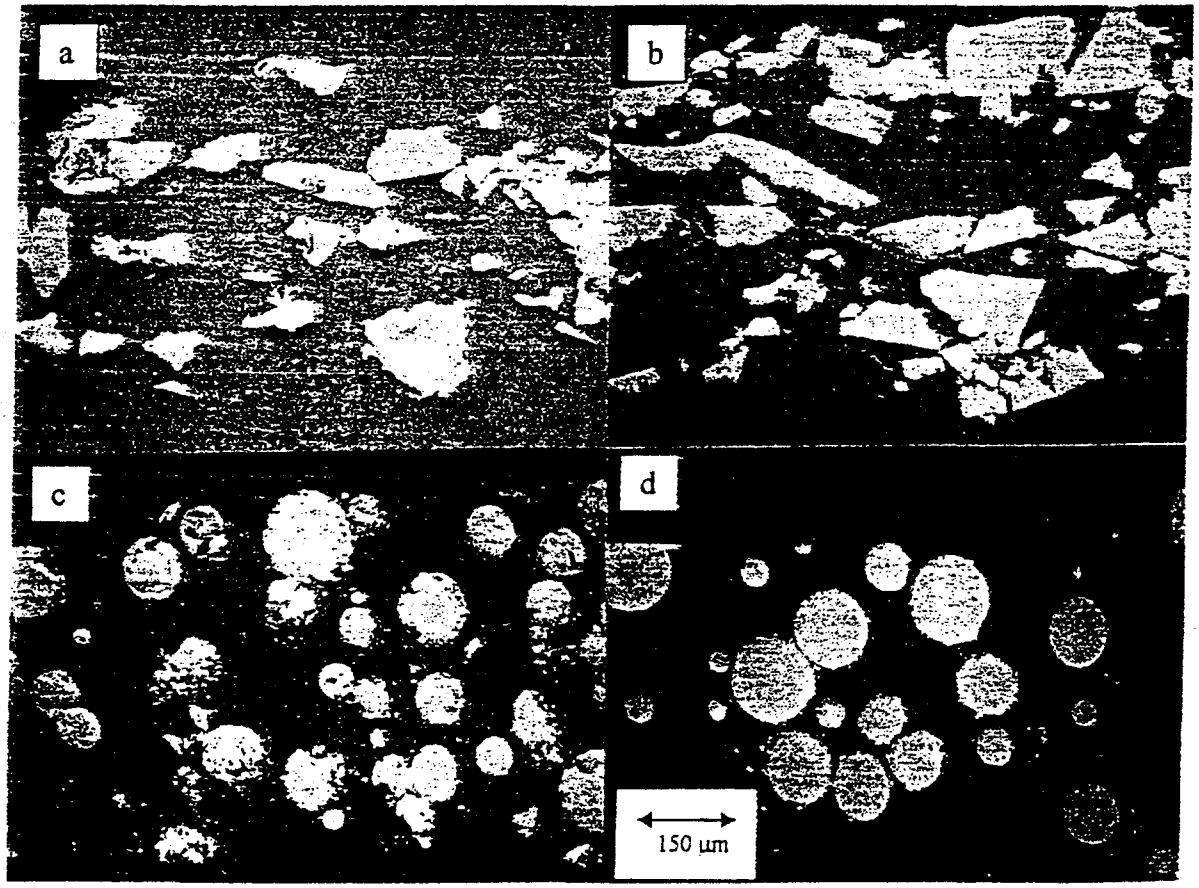

Figure 5. Comparison of fuel meats made from comminuted or machined versus atomized fuel powders; a) U_3Si_2 comminuted, b)U-10Mo machined, c) U_3Si_2 atomized, and d) U-10Mo atomized. All are SEM images at the same magnification.

Crucial microplate attributes are fuel particle distribution within the fuel meat and bond strength between the picture frame and coverplates. Thus, prior to the final machining steps and the extensive physical inspection, the plates are radiographed and subjected to the bend test. The radiography specification required that no 3 mm diameter spot would contain more than 10 g of U per cc. Approximately, 30 spots were measured on each microplate. All microplates met this criterion with average uranium densities ranging from 4.2-5.8 g-U/cc. The bend test subjects a portion of the rolled assembly to bending in two directions. A strip of the rolled assembly, completely outside the fuel zone, nominally 76 mm x 13 mm, is cut with the long dimension transverse to the rolling direction (i.e. the long dimension is perpendicular to the rolling direction). The strip is bent 90° in one direction, returned to 0° then bent 90° in the other direction and returned to 0°. The edges are visually examined for delamination. Three strips from each microplate rolling assembly were tested. Any strip which appeared to have separations between the layers are determined to be unbonded and the entire assembly of microplates would have been rejected. There were no bend test failures. Further details and validation of the bend test are presented in [15].

Future Activities

The machining method for producing alloy fuel powders is not suitable for large-scale fuel production. The contamination of the powders due to tool disintegration is undesirable. Thus, a new method of powder production is sought. Currently, the feasibility and practicality of a gas atomization process is underway. The high purity of the powder product and potential flexibility of process are attractive. However the equipment is complex and capital intensive, meriting thorough evaluation process prior to acquisition. The physical metallurgy of the uranium alloys used in this study has large unknowns. Specifically, there is a dearth of the time-temperature-transformation data. In addition, the transformation kinetics and mechanisms are apparently complex and not well studied. These attributes will play a major role in process optimization of fuel plates. Thus, detailed physical metallurgy studies will be undertaken on the alloys, which demonstrate good irradiation performance. In the realm of fabrication, fuel loadings must be increased to meet the overall program goal of 8-9 g/cc of uranium in the fuel meat. We will also attempt to lowering the rolling temperatures to simplify the process.

Summary and Conclusions

A further miniaturization of the dispersion fuel miniplate has been designed without any mechanical compromise, the microplate. Sixty-four microplates were successfully fabricated and are currently being irradiated in the ATR. The majority of the microplates were fabricated from high density uranium alloys with a retained gamma phase. Powders were prepared either by high speed machining of cast and heat treated metallic rods, communition of intermetallic compounds, or atomization of uranium melts. The fuel dispersants were mixed with aluminum powder and compacted. Atomized powders were preformed into fuel meats via extrusion. The as-extruded fuel meats were to fit in microplate picture frames. All fuel materials, compacts and extruded fuel meats were hot rolled and finished into microplates. Inspection criteria, for fuel distribution (radiography) and bond integrity (blister anneal and bend testing) were performed and all fuel materials successfully passed.

Acknowledgements

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