PROGRESS IN IRRADIATION PERFORMANCE OF EXPERIMENTAL URANIUM–MOLYBDENUM DISPERSION FUEL

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ABSTRACT

High-density dispersion fuel experiment, RERTR-4, was removed from the Advanced Test Reactor (ATR) after reaching a peak U-235 burnup of ~80% and is presently undergoing postirradiation examination at the ANL alpha-gamma hot cells. This test consists of 32 mini fuel plates of which 27 were fabricated with nominally 6 and 8 g cm^{-3} atomized and machined uranium alloy powders containing 7 wt% and 10 wt% molybdenum. In addition, two miniplates containing solid U-10 wt% Mo foils and three containing 6 g cm^{-3} U_3Si_2 are part of the test. The results of the postirradiation examination and analysis of RERTR-4 in conjunction with data from previous tests performed to lower burnup will be presented.

INTRODUCTION

At the time of the last RERTR meeting in October 2000 in Las Vegas, the results from three tests, RERTR-1, 2, and 3, were presented [1,2]. Tests RERTR-1 and 2 were low-temperature scoping tests that contained a wide range of uranium alloy compositions. The main conclusion after postirradiation analysis of these first two tests was that uranium alloys with a molybdenum content between 6% and 10% by weight showed excellent irradiation behavior [3]. Test RERTR-3 was designed to explore the higher-temperature behavior of these U-Mo/Al dispersion fuels.

Postirradiation examination allowed the characterization of U-Mo/Al interaction. The formation of the aluminide interaction phase appeared to be the only aspect of fuel behavior that is significantly affected by temperature. The irradiation behavior of the U-Mo fuel alloy itself was deemed athermal over the temperature range tested. Extensive metallographic analysis of these first three tests was used to develop a fuel behavior model [4]. Based on the positive results of the first three tests, two further tests, RERTR-4 and 5, were designed with larger, so-called miniplates that are more prototypic of full-size test reactor fuel plates and also allow for more accurate postirradiation measurements.

The miniplates irradiated in RERTR-4 and 5 contained either atomized fuel particles, supplied by KAERI, or machined fuel particles, supplied by AECL, ranging in composition from, nominally, 6 wt.% Mo to 10 wt.% Mo. The fuel plates in these tests measured 100 mm x 25 mm x 1.40 mm; the meat was in a rectangular zone nominally 0.64 mm thick and contained, nominally, 6 and 8 g U cm^{-3} in the fuel meat. The experiments were irradiated in the Advanced Test Reactor (ATR) for 116 EFPD (effective full power days) and 204 EFPD. In addition to 30 dispersion fuel miniplates, test RERTR-4 also included two miniplates with solid U-Mo alloy cores.

The RERTR-5 was removed from the reactor at a peak burnup of ~50% U-235, whereas RERTR-4 terminated at ~80% burnup. Postirradiation examinations up to the 50% burnup level have been largely completed, and a preliminary assessment was presented at the 2002 RRFM meeting in Ghent, Belgium. Examination of the miniplates from RERTR-4 has progressed through plate thickness measurements. In this paper the data obtained on RERTR-4 and -5 to date will be assessed.

2.1 Postirradiation Data

Thickness measurements were made at six positions along the longitudinal centerline of each plate of both RERTR-4 and 5. The average changes in plate thickness of U-10 Mo fuel are plotted as a function of burnup in Figure 1. As expected, fuel swelling results in an increase in plate thickness with burnup. The considerable scatter in the thickness values is due to the fabrication and operational variables present. Some general trends in the values plotted in Figure 1 may be traced to fabrication variables, such as the lower values for 6 g cm^{-3} plates that contain about ~40 volume % fuel compared to ~50% for the 8 g cm^{-3} plates, and somewhat lower values for plates containing machined fuel powder that have high as-fabricated porosity. The scatter in the data, apart from measurement errors, must be due to operational variables as they affect swelling.

2.2 Fission-Induced Swelling

Swelling of the fuel meat is composed of several components:

1. Swelling of the U-Mo alloy particles by accumulation of solid and gaseous fission products.
2. Volume increase as the interaction phase forms by interdiffusion of U-Mo and Al at the fuel particle surface.
3. Accumulation of fission products in this interaction phase.
4. Accommodation of the above swelling by as-fabricated porosity in the meat.

Swelling of the U-Mo alloy was determined from immersion volume data and quantitative metallography of several plates from RERTR-4 [5]. The results are shown in Figure 2. The data points in Figure 2 cover a fuel temperature range of ~100-280ºC and show no measurable temperature dependence. This is supported by scanning electron microscopy (SEM) of the fuel microstructure shown in Figure 3.

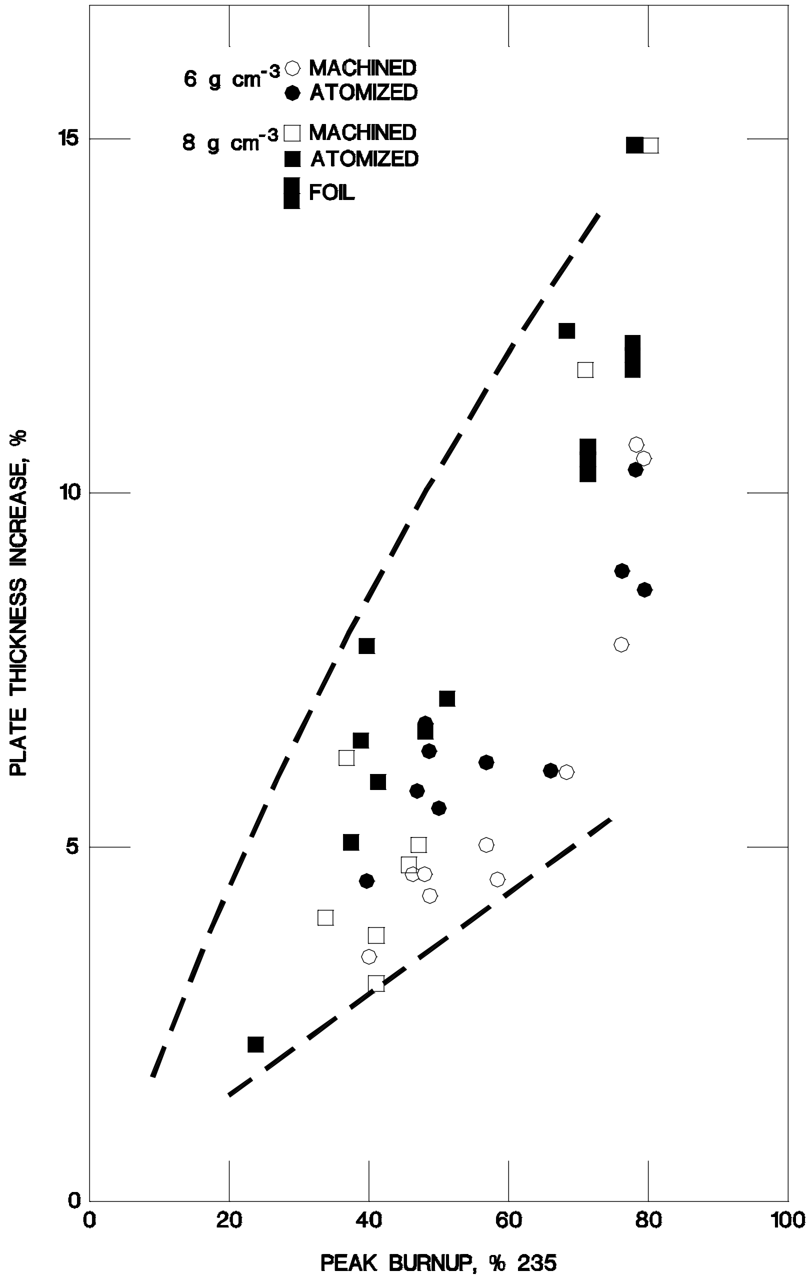


Figure 1. Thickness increases of U-10 Mo dispersion miniplates

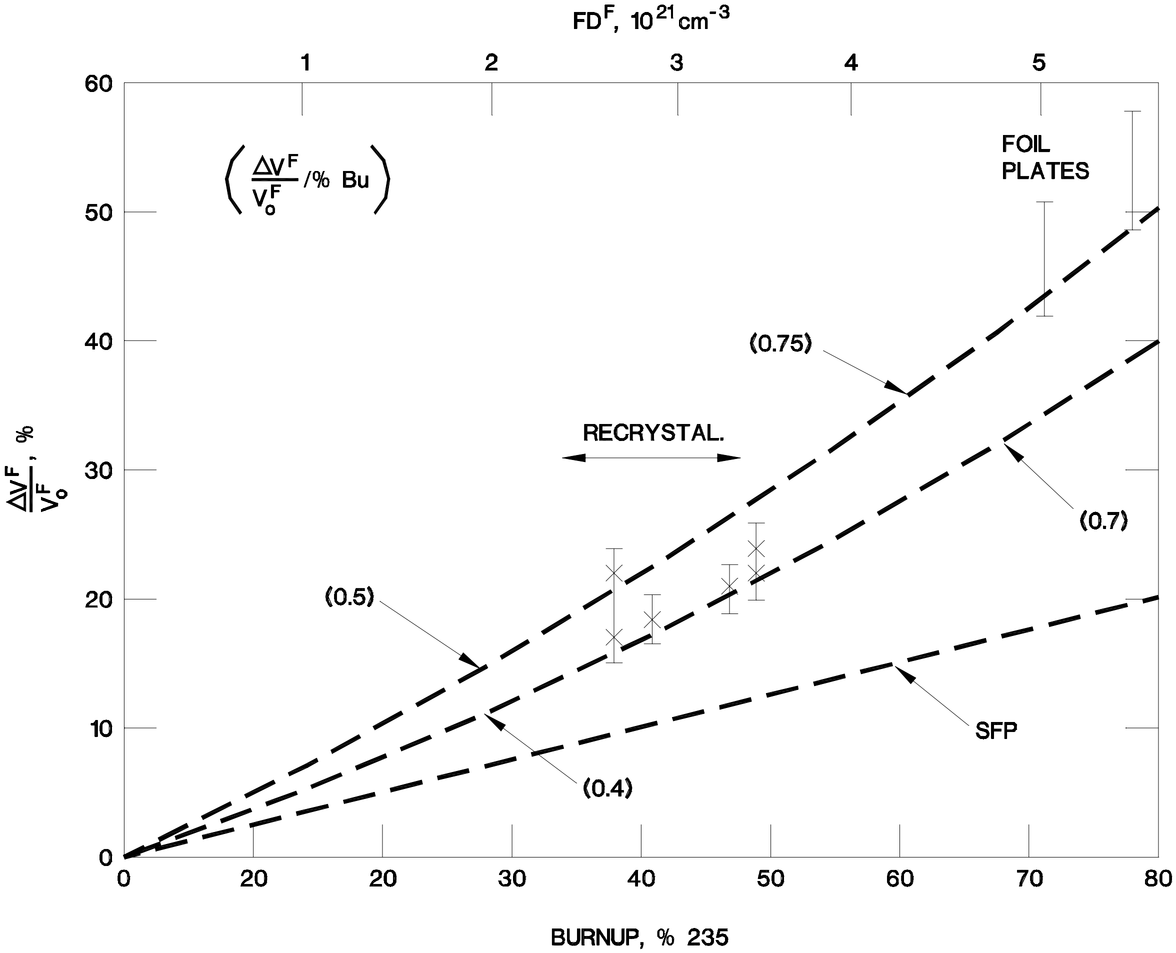


Figure 2. Swelling of U-10 Mo derived from immersion volume and quantitative metallography.

The microstructure at low and high temperature is very similar with small fission gas bubbles having formed at ~40% burnup at grain boundaries. By ~50% burnup transformation of the fuel into a very small recrystallized grain structure has started. This transformation increases the swelling rate as many more grain boundaries become available for gas bubbles to nucleate and grow on. The swelling rates in terms of % fuel volume increase per % U-235 burnup $\left(\frac{\Delta V^F}{V_o^F} / \%Bu \right)$ are indicated in Figure 2.

The onset of recrystallization depends on the type of fuel; it occurs at lower burnup for machined powder because of its high initial dislocation density, and for lower Mo content because of its different mechanical properties. The burnup range where recrystallization has been observed to start is indicated in Figure 2.

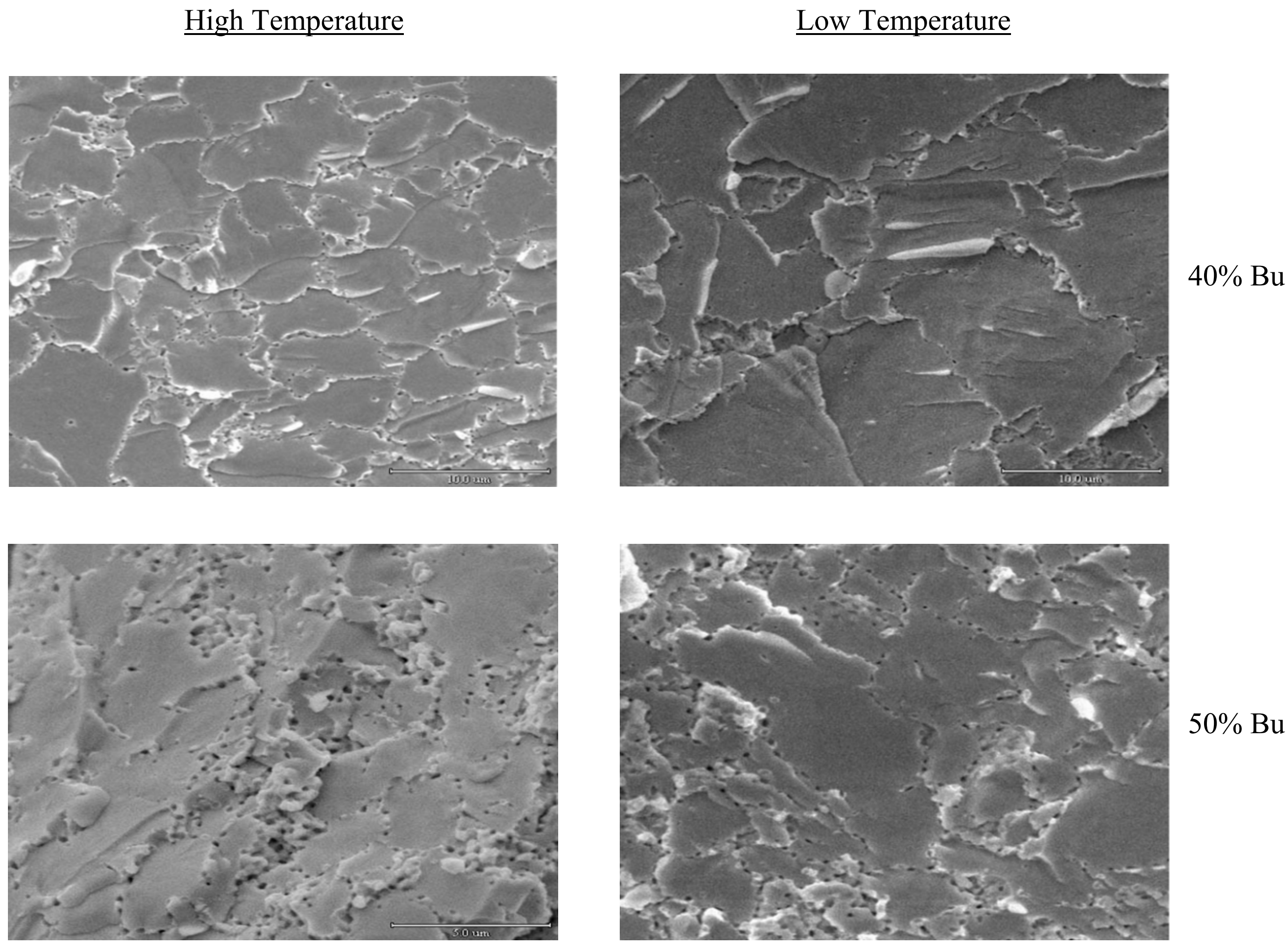
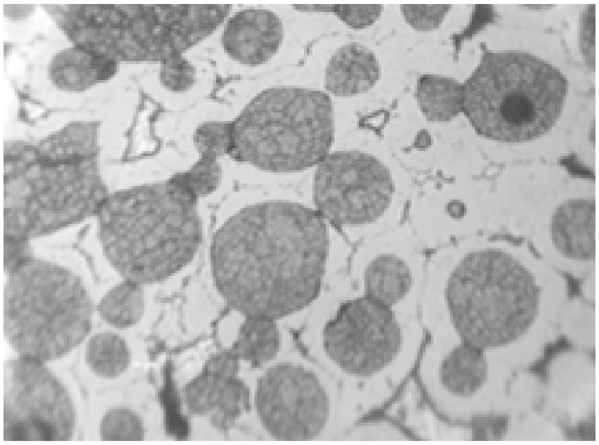


Figure 3. Microstructure of unreacted U-10Mo fuel particles at low and high temperatures irradiated to 40 to 50% U-235 Bu (SEM) showing beginning of recrystallization at 50% Bu.

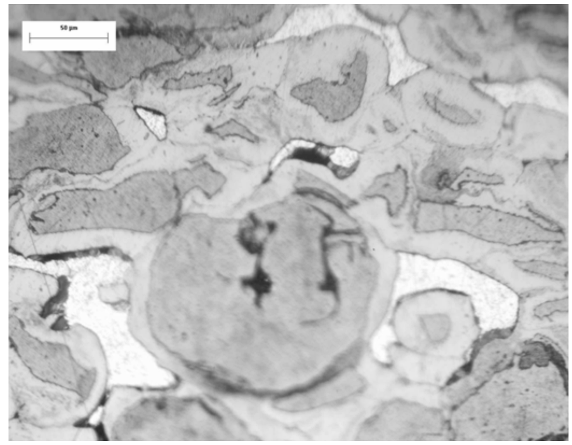
Because the formation of the interaction phase involves the consumption of both U-Mo alloy and Al, it imparts a pseudo temperature dependence on the net swelling of the U-Mo alloy, and because it has a very low thermal conductivity, its formation increases the meat temperature as it consumes highly conductive aluminum. The complex interplay of temperature dependencies and changing temperature of the swelling components require detailed modeling of each particular fuel plate. This subject is treated in a paper by S.L. Hayes in this conference [4]. Whenever temperatures are quoted in this paper, they refer to the beginning of the irradiation where calculations are least ambiguous; these temperatures are only used to indicate trends.

High temperature 8 g cm^{-3}

600AH

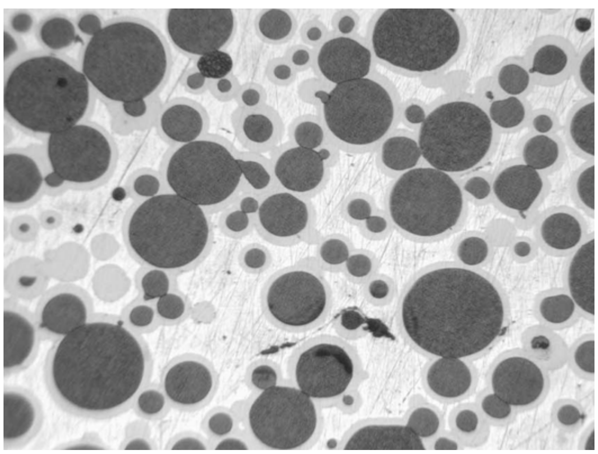


600V



Low temperature 8 g cm^{-3}

600D



600C

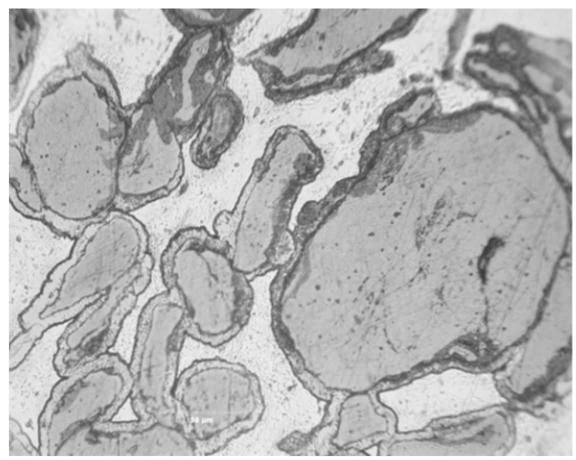


Figure 4. Optical metallographic examples of machined and atomized U-10 Mo fuel irradiated at ~100 to 200ºC BOL temperature, showing extent of fuel-Al interaction.

Finally, the porosity introduced in the fuel meat during plate fabrication to an extent absorbs the swelling of fuel and interaction and so reduces the net meat swelling and thickness increases. Irregular-shaped machined powder results in larger porosity in the meat compared to spherical (atomized) powders. However, because of the relatively larger specific surface of machined powder it has, all other factors being the same, a higher rate of interaction formation compared to spherical powder. Therefore, the mitigating effect of as-fabricated porosity on meat swelling may not manifest itself under certain conditions.

3. Discussion and Conclusions

The maximum plate thickness increases by ~80% burnup may seem relatively high at ~15%, however, this is inevitable for fuel loadings of 8 g U cm^{-3} because of the large volume of fission products formed. The important observations are: stable, apparently athermal swelling of the U-Mo alloy particles with the presence of small uniformly distributed fission gas bubbles. No evidence of unstable, break-away, swelling, characteristic of other high-density fuels, has been found.

The formation of a U-Mo/Al interaction phase will be significant, consuming practically all matrix aluminum at higher temperatures. The fission-induced swelling rate of this compound is, however, low and very stable. The interaction phase occupies a larger volume than its U-Mo and Al constituents and therefore, contributes to swelling. This contribution, however, is limited by the amount of Al matrix available. The main effect of the interaction product formation is the reduction of thermal conductivity of the meat, which should be carefully assessed for a particular fuel design.

That the relative complex interplay of the various irradiation effects requires detailed modeling is illustrated by the following example. A higher BOL temperature may not result in a larger amount of swelling. This is shown schematically in Figure 5, for identical fuel meats operating at different temperatures to 80% burnup. Because the interaction phase formation rate is strongly temperature dependent, it will deplete the matrix Al early during the irradiation for higher BOL temperature (140ºC in Figure 5) and the volume fraction of interaction product, (U-Mo)Al_x, will have reached its maximum value early. The amount of swelling associated with the interaction is proportional to this amount; however, in this higher temperature case, more U-Mo is consumed and since U-Mo has a higher irradiation swelling rate than (U-Mo)Al_x, this compensates for the interaction swelling. At lower temperature (100ºC in Figure 5) the U-Mo/Al interaction is only partially completed at 80% burnup, and its volume fraction and associated meat swelling contribution is consequently much smaller. However, because of the amount of U-Mo consumed is proportionally smaller as well, the original fuel volume fraction is only slightly diminished. The overall meat swelling at low temperature should be completely dominated by U-Mo swelling showing no measurable effect of (U-Mo)Al_x formation. At some intermediate temperature the interaction may be completed at the end of the irradiation (120ºC in Figure 5). Its contribution to swelling of the meat is maximal but the U-Mo volume fraction has remained higher during the preceding irradiation time and its high swelling rate has therefore contributed more to the overall meat swelling than is the case at 140ºC. The thickness data plotted in Figure 6 appear to show this behavior of maximum swelling at some intermediate temperature.

Finally, two unique miniplates were part of test RERTR-4. They contained each two thin discs of U-10 Mo of ~12-mm diameter and 0.3-mm thickness. The fuel meat density was 15.3 g U cm^{-3}. The meat was thinner than normal in these miniplates in order to remain within the U areal loading limits. These plates showed excellent irradiation behavior; their thickness increases are included in Figure 1 for comparison. The fuel swelling from these plates was derived from the thickness measurements and from a metallographic section shown in Figure 7. The results are plotted on the fuel swelling curve in Figure 2 and are consistent with the extrapolation of the data derived from RERTR-5 miniplates.

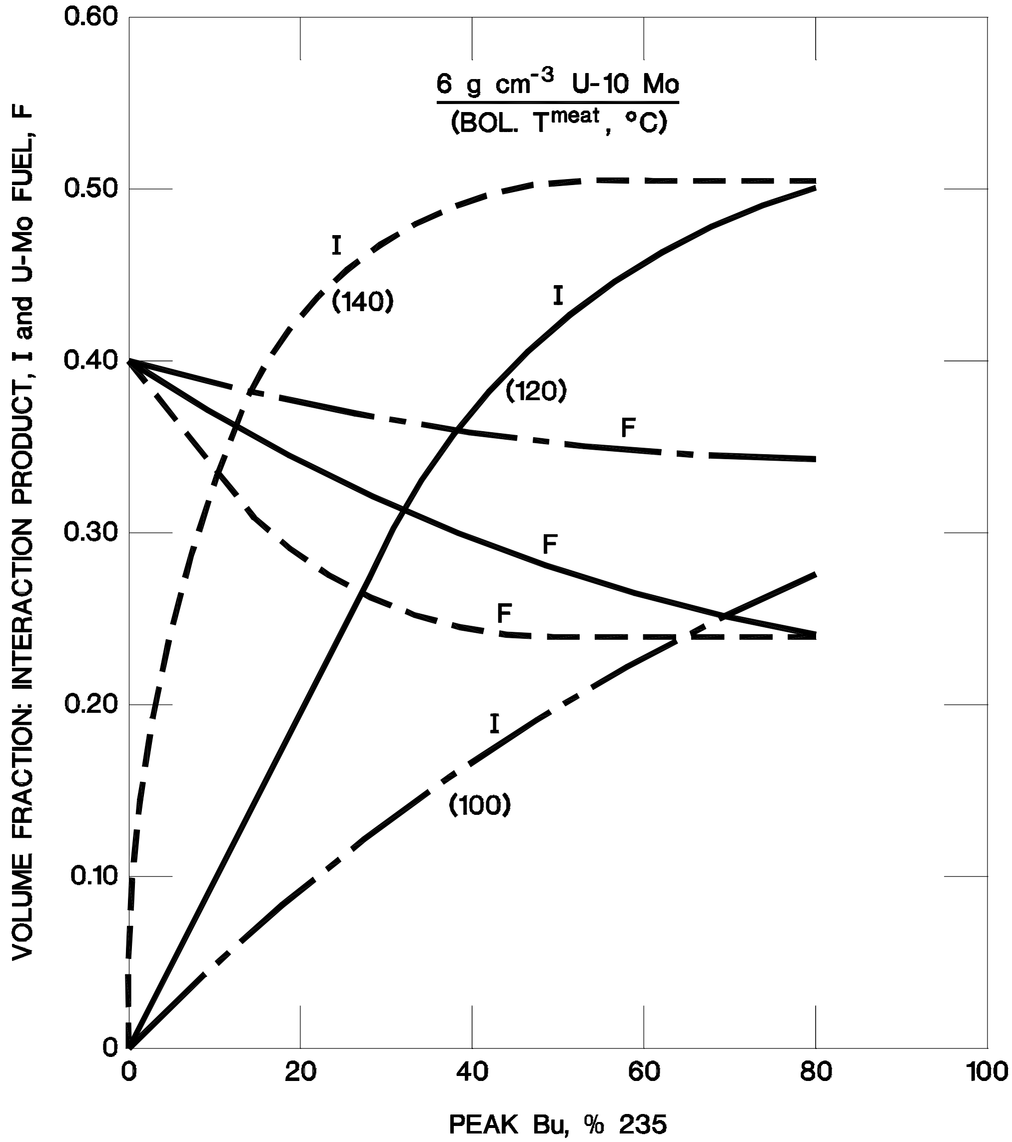


Figure 5. Changes in volume fraction of U-Mo/Al interaction and as-fabricated U-Mo particles as a function of BOL temperature and U-235 Bu.

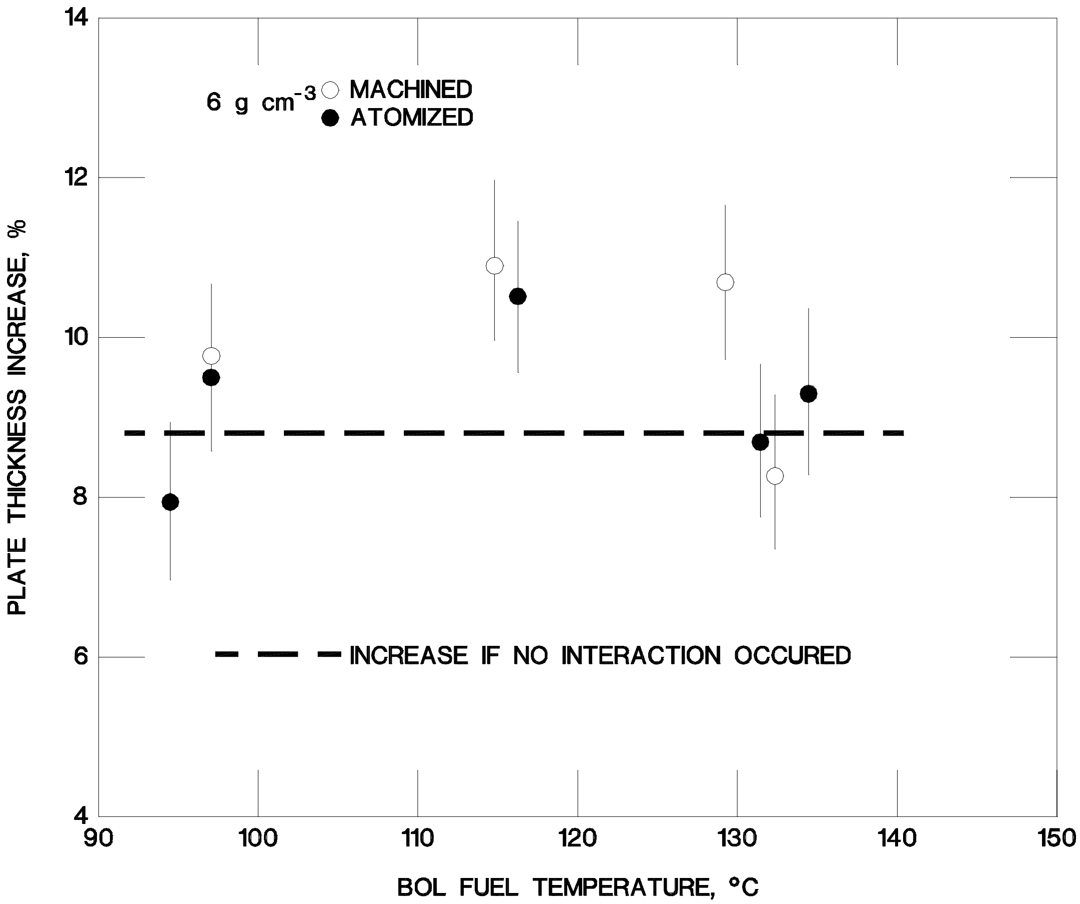


Figure 6. Plate thickness increase at 80% U-235 Bu showing effect BOL temperature on 6 g cm^{-3} fuel plate swelling

As shown in Figure 7 there is a slight interaction zone at the U-10 Mo/Al interface, consistent with the temperature at that location. The fission gas bubble morphology is uniform throughout the fuel, consisting of small, evenly distributed bubbles on recrystallized grain boundaries (see Figure 8). This morphology is very similar to that observed in low-temperature fuel particles at ~70% burnup in test RERTR-2.

In conclusion, the results from the five irradiation experiments have shown excellent irradiation behavior of U-Mo/Al dispersion fuel with uranium loadings of up to 8 g cm^{-3}. Swelling of the U-Mo alloys is athermal and stable up to ~300ºC. The interaction between U-Mo and Al affects overall meat swelling to various degrees depending on irradiation conditions, primarily temperature. The main effect of U-Mo/Al interaction is a decrease in meat thermal conductivity. The components of the irradiation behavior are largely understood and allow detailed modeling of the fuel plate swelling and temperatures. The initial tests with monolithic U-Mo fuel meat are very positive, opening the possibility for development of fuel with uranium densities well excess of 8 g cm^{-3}.

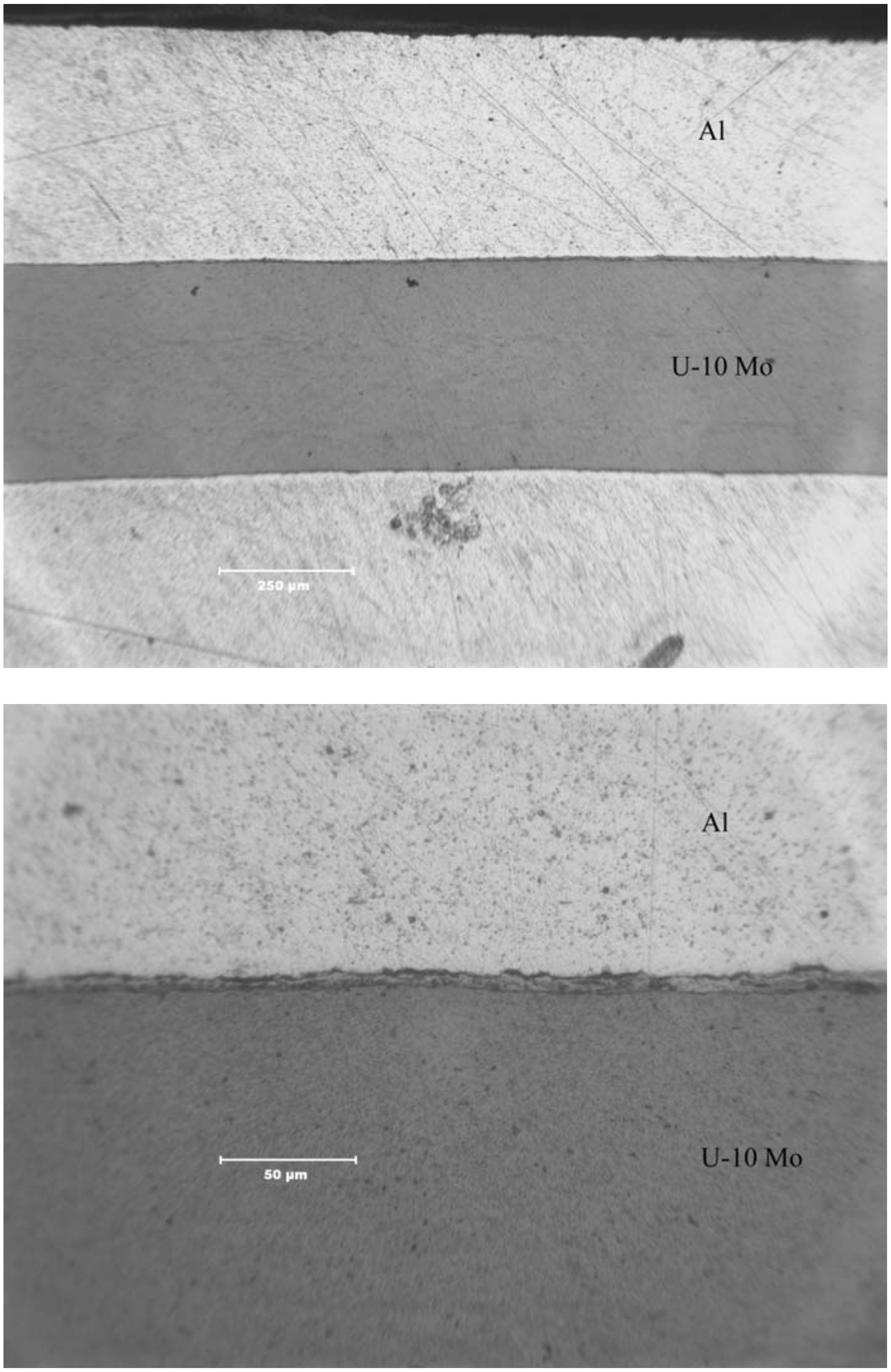


Figure 7. Metallographic connection of LEU U-10 Mo foil (monolithic) miniplate @ 80% U-235 Bu.

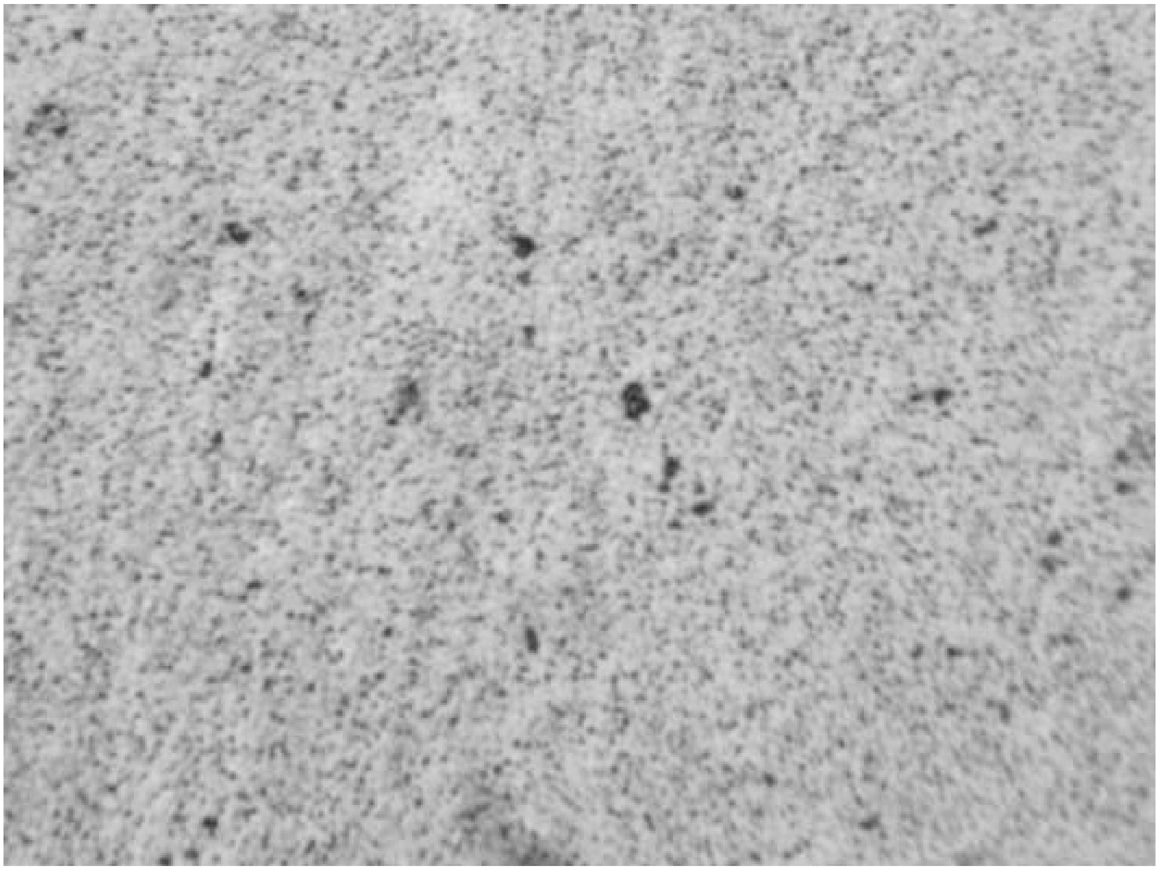


Figure 8. Bubble morphology in monolithic U-10 Mo fuel at ~80% U-235 Bu.

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