OBSERVATIONS ON THE IRRADIATION BEHAVIOR OF U-Mo ALLOY DISPERISON FUEL

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ABSTRACT

Initial results from the postirradiation examination of high-density dispersion fuel test RERTR-3 are discussed. The U-Mo alloy fuels in this test were irradiated to 40% U-235 burnup at temperature ranging from 140ºC to 240ºC. Temperature has a significant effect on overall swelling of the test plates. The magnitude of the swelling appears acceptable and no unstable irradiation behavior is evident.

INTRODUCTION

Previous results from postirradiation examinations of microplates irradiated in RERTR-1 and RERTR-2 to screen a variety of candidate materials, performed in the Advanced Test Reactor (ATR), have shown several U-Mo alloys to be promising candidates for use as a high uranium density fuel [1-2]. U-Mo alloys with at least 6 wt% Mo showed low and stable swelling behavior comparable to currently used uranium silicide fuels. The RERTR-1 and –2 screening tests took these alloy fuels to high exposures (70 at % U^{235} burnup, $5 \bullet 10^{21}$ fiss./cm^3 fuel fission density), peak fuel temperatures of <100°C and fuel loading of ~4 g-U/cm^3. The peak fuel temperatures of <100°C and fuel loadings of ~4 g-U/cm^3 were below the values needed for use in high power research reactors. A third irradiation test, designated RERTR-3, containing fuel loadings of 8 g-Ucm^{-3} that reached temperatures >200° began irradiation in October 1999 [3]. The experiment included binary fuel alloys with 6-10 w% Mo, as well as 6 w% Mo – 1.7 w % Os and 6w% Mo – o.8 w% Ru ternary alloys. After reaching a peak burnup of ~40%, RERTR-3 was removed from the ATR and is currently undergoing postirradiation examination at ANL-E.

Although the test contained both atomized and machined fuel powders, priority in examination was given to the former. In order to offer an initial assessment at this meeting of the effects of temperature and composition on the irradiation behavior, four test plates were examined in detail, viz U-10 Mo irradiated at low temperature and U-6 Mo, U-6 Mo – 1.7 Os and U-10 Mo irradiated at high temperature. In this paper, observations of the basic irradiation effects observed during the initial examination are discussed whereas a more quantitative analysis of the postirradiation data will be presented in a companion paper [4].

DISCUSSION

Because of the small size of the test plates, volumetric measurement with the customary immersion method would not yield swelling data with sufficient accuracy; therefore swelling was determined by plate thickness measurement. These measurements for binary, atomized, fuel plates are shown in Fig. 1, normalized to a meat fission density of 10^{21} cm^{-3} (~30% Bu), as a function of beginning of life (BOL) fuel centerline temperature. These BOL temperatures were calculated with a one dimensional heat transfer model. It is clear form Fig. 1 that the plate swelling is a strong function of temperature and that there appears to be no difference, within the measurement uncertainty, between the various compositions.

There are two irradiation effects that each, or together, may be responsible for the observed temperature dependence of the swelling. One is the behavior of fission gas in the U-Mo alloy (the swelling due to solid fission products can be considered athermal), the other is the net density decrease associated with the formation of U-Mo/Al interaction phases resulting from irradiation enhanced interdiffusion. A glance at the optical micrographs shown in Fig. 2 suggests that the latter is the major contributor to the temperature dependence of the swelling. In addition to the large increase in interaction going from a fuel temperature of 175°C to 217°C in U-10 Mo, there is also more interaction in the U-6 Mo as well as the U-6 Mo –1.70s samples. There are indications of the presence of small gas bubbles at the grain boundaries in the unreacted fuel. This is better illustrated in the SEM fractographs shown in Fig. 3.

Small gas bubbles have begun to form in the 175°C sample and are more numerous and larger at 217°C. However, the 175°C sample reached only 30% burnup compared to 40% for the 217°C sample. Comparison of the bubble morphology in this latter sample with that of the same fuel irradiated previously in RERTR-1 to the same burnup, but at a much lower temperature of ~65°C, leads us to conclude that there is no effect of temperature on the fission gas behavior over the temperature and burnup range tested thus far, and that the temperature dependence of the measured plate swelling is the result of fuel-aluminum interdiffusion. Fission gas bubbles begin to form on grain boundaries somewhere below 30% burnup, between 30 and 40 % burn up fission induced grain refinement starts, providing additional grain boundaries for fission gas precipitation. This process, which has been described in detail previously [1], will progress with further burnup, eventually covering the entire fuel particles.

The evolution of the fuel microstructure does appear to be a function of composition as shown by a comparison of U-10Mo and U-6 Mo in Fig. 5. Grain refinement has evidently started at a lower burnup in the U-6 Mo sample and has already covered significant fractions of the grains. All fuel compositions are expected to develop microstructures as shown in Fig. 5c (a 70% burnup sample from RERTR-2) but the lower Mo compositions will complete the restructuring at lower burnup and should therefore have a somewhat higher fuel swelling rate.

Although the swelling resulting from fuel-Al interaction is rapid and substantial at higher temperatures, the amount of it is limited by the fraction of Al present in the dispersion. This is depicted in Fig. 4 where the meat swelling calculated from plate thickness increases and the volume fractions of interaction phase and residual Al are plotted.

As would be expected, the volume fraction of the interaction phase (v^{int}/v^{m})-plotted on the abscissa inversely correlates with the volume fraction of matrix aluminum remaining in the meat; (v^{Al}/v^{m}). However, the fact that the total meat swelling ()v^{m}/v^{m}) also intercorrelates with these parameters indicates that fuel-aluminum interdiffussion is a major factor in the observed fuel swelling. The meat swelling and plate thickness increase is expected to continue with burnup, as shown in Fig. 6 for two temperatures. The swelling is initially dominated by temperature dependant interdiffusion, but when the Al in the meat has been consumed, the swelling is further controlled by temperature independent fuel swelling. Complete consumption of aluminum will occur at approximately 50% burnup at a temperature of 217°C.

Another consequence of the extensive fuel-Al interdiffusion is its effect on the fuel temperature. The interaction phase is most likely (U-Mo) Al_x [4]. This aluminide phase has a rather low thermal conductivity and as it replaces aluminum, the effect will be that the fuel temperature will rises from its BOL level before it decreases due to U-235 burnup. There will also be a substantial temperature gradient across the fuel meat thickness, particularly for high temperature cases, as is apparent in the micrographs shown in Fig. 7 and in the interdiffusion layer thickness measured at several temperatures as shown in Fig. 8.

Analysis of the thermal conductivity changes is in progress.

CONCLUSIONS

The following conclusions may be drawn from the initial results of the post irradiation examination of high-density dispersion fuel test RERTR-3.

The extent of fuel plate swelling is acceptable and stable.

Swelling is predominantly due to temperature dependant U-Mo/Al interdiffusion up to burnup where the matrix Al is consumed by this interdiffusion process.

The aluminide interaction product appears stable and contains no fission gas bubbles. It has however a low thermal conductivity which results in an increased fuel temperature.

The swelling behavior of the unreacted fuel appears to be athermal in the range of temperature and burnup tested.

Lowering of Mo content results in somewhat higher rates of interdiffusion and fission gas swelling.

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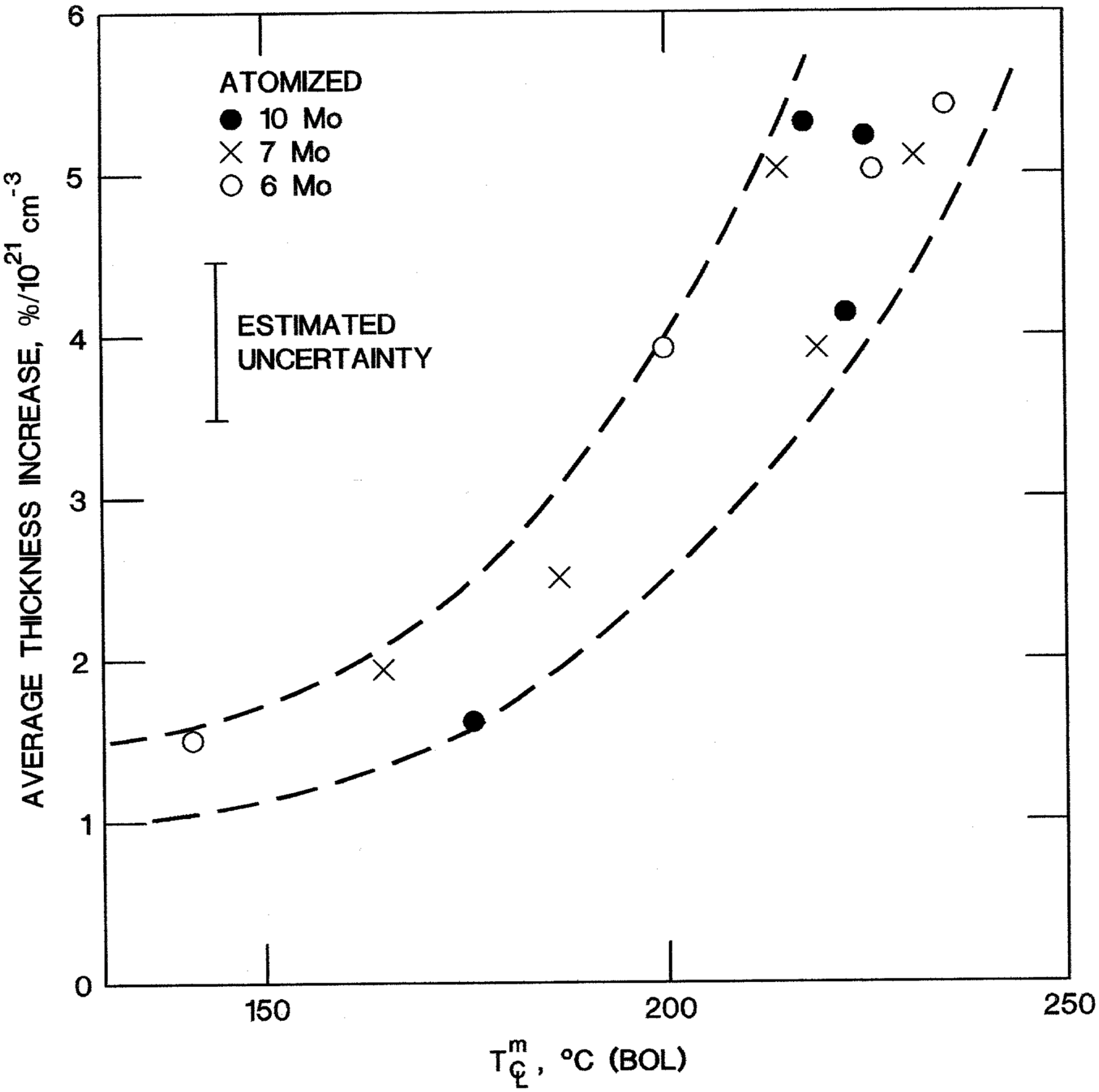
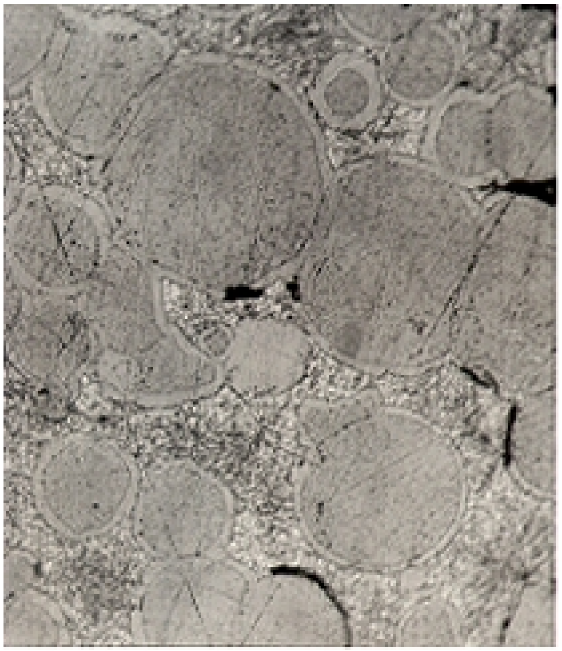
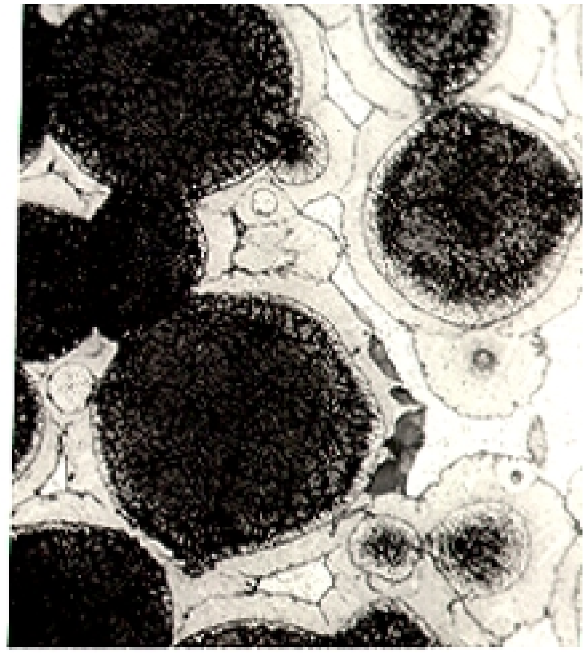
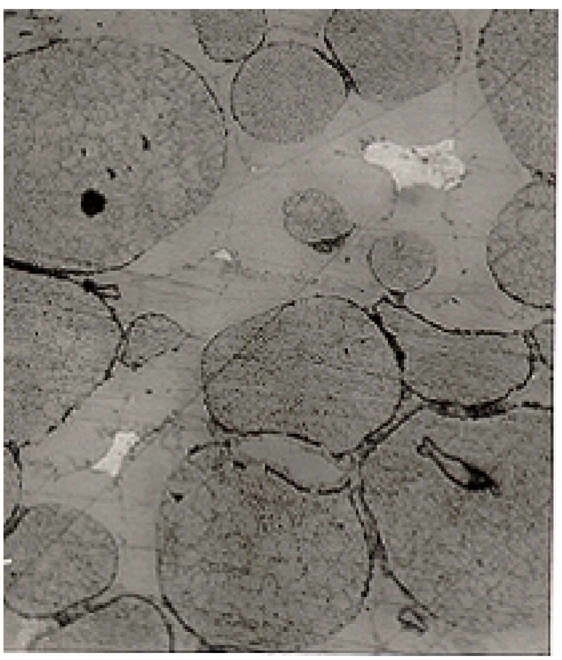


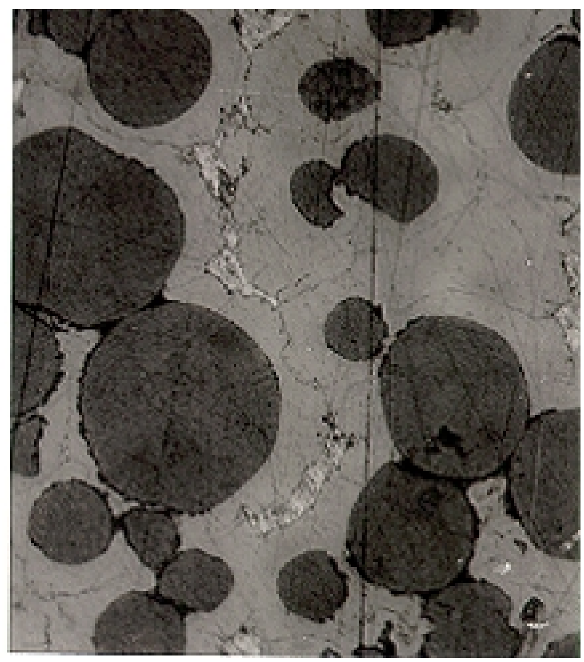
Figure 1. Thickness increase v.s. calculated beginning of life plate centerline temperature RERTR-3 with 6 to 10 weight percent Mo atomized fuel particles, normalized to $10^{21}cm^{-3}$ meat fission density.





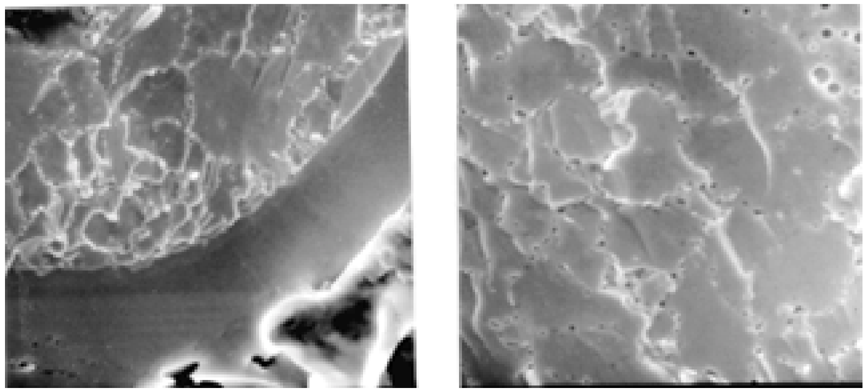
10Mo @ 175ºC 400 x 10Mo @ 217ºC



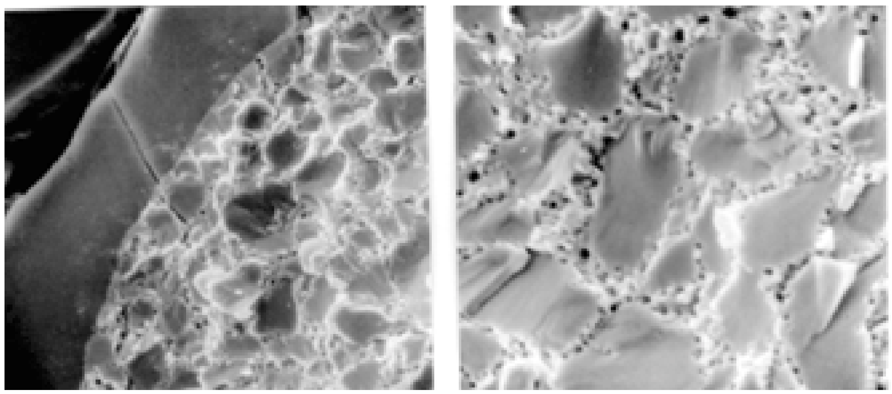


6Mo @ 223ºC 400 x 6Mo-Os @ 225ºC

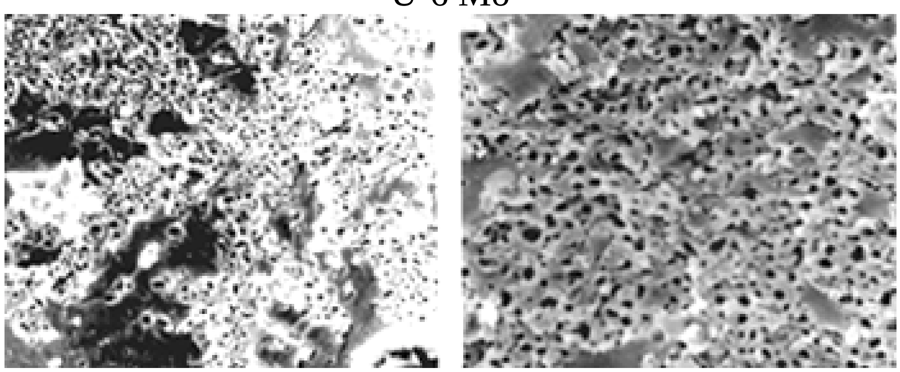
Figure 2. Microstructure at meat center of selected RERTR-3 samples showing effect of irradiation temperature.



U-10 Mo



U-6 Mo



U-10 Mo 65ºC

Figure 5. Comparison of fuel microstructures of U-10 Mo and U-6 Mo at ~40% Bu irradiated at T>200ºC, and at 70% Bu irradiated at~65ºC.

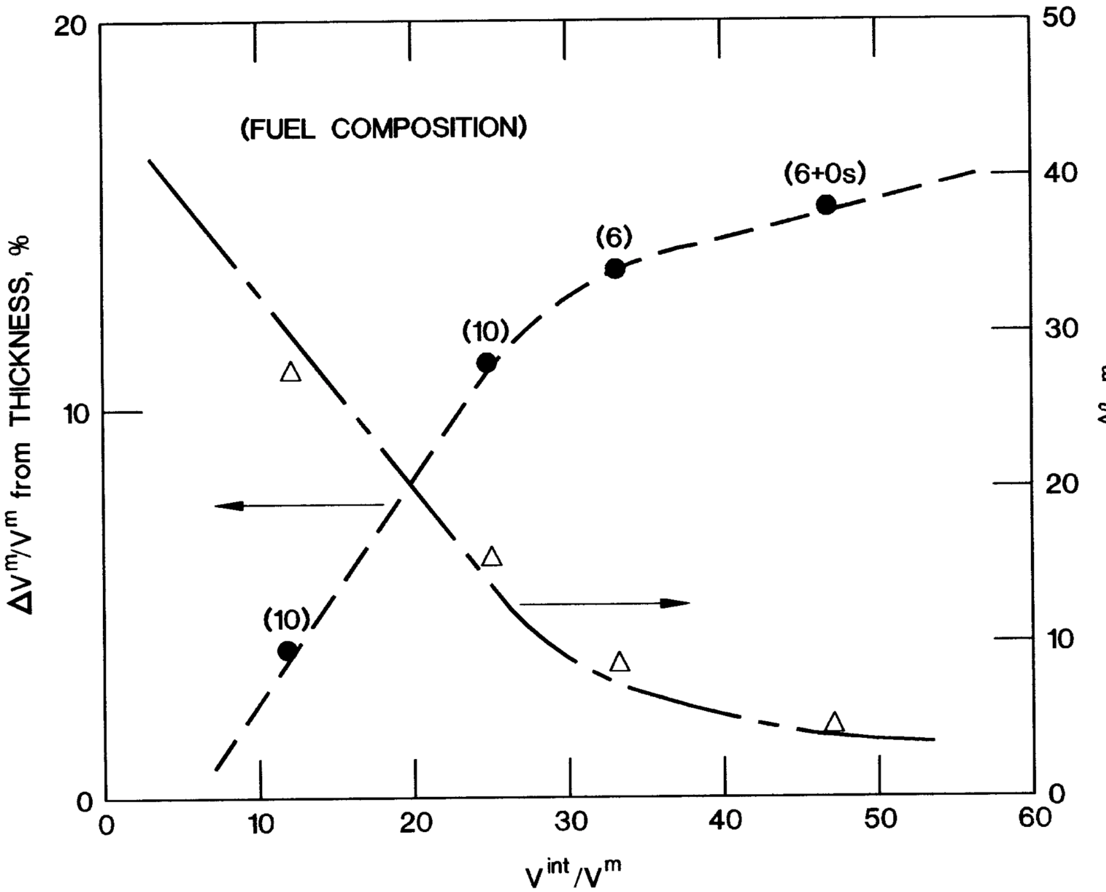
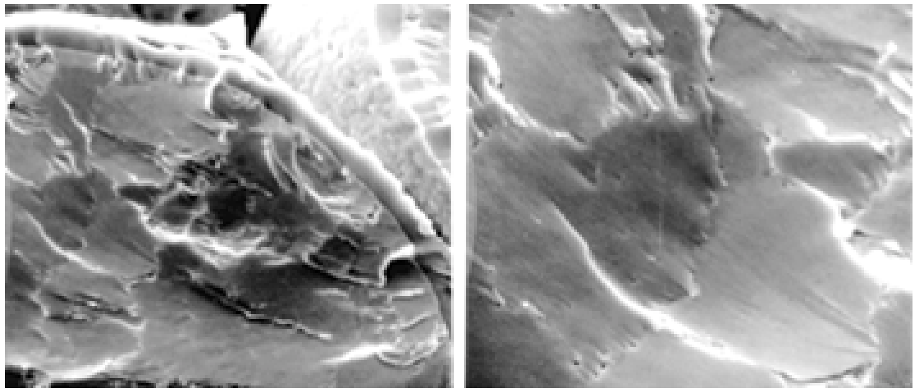
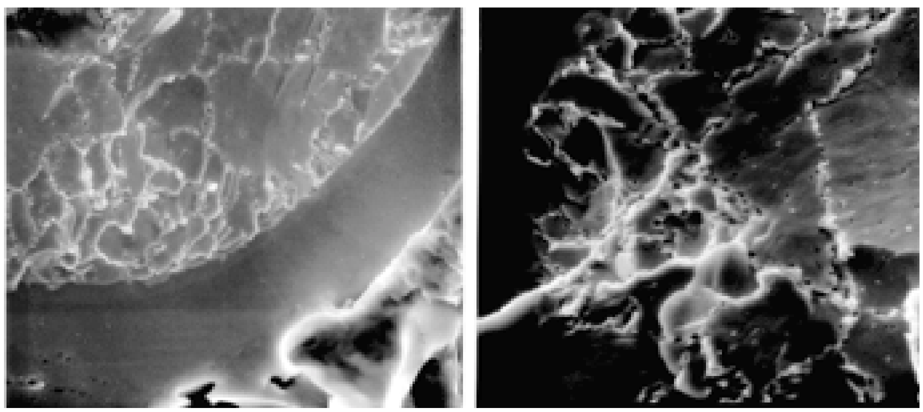


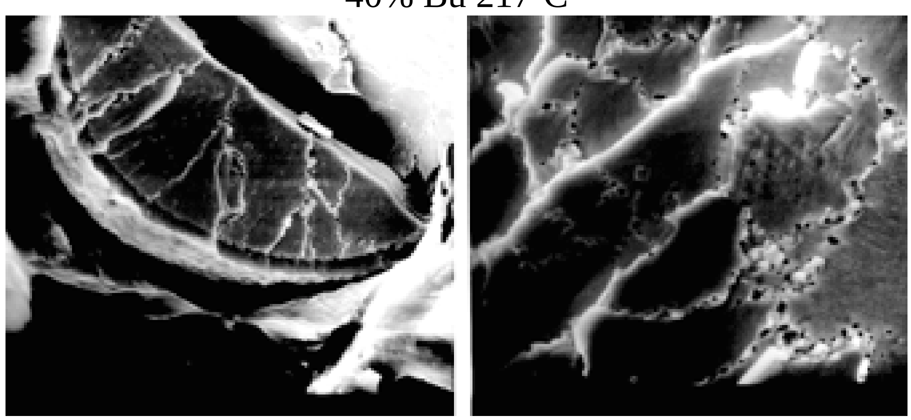
Figure 4. Correlation of meat volume increase, derived from thickness measurements, with volume fractions of U-Mo/Al reaction product formed and volume fraction of Al remaining with fuel meat.



30% Bu 175ºC

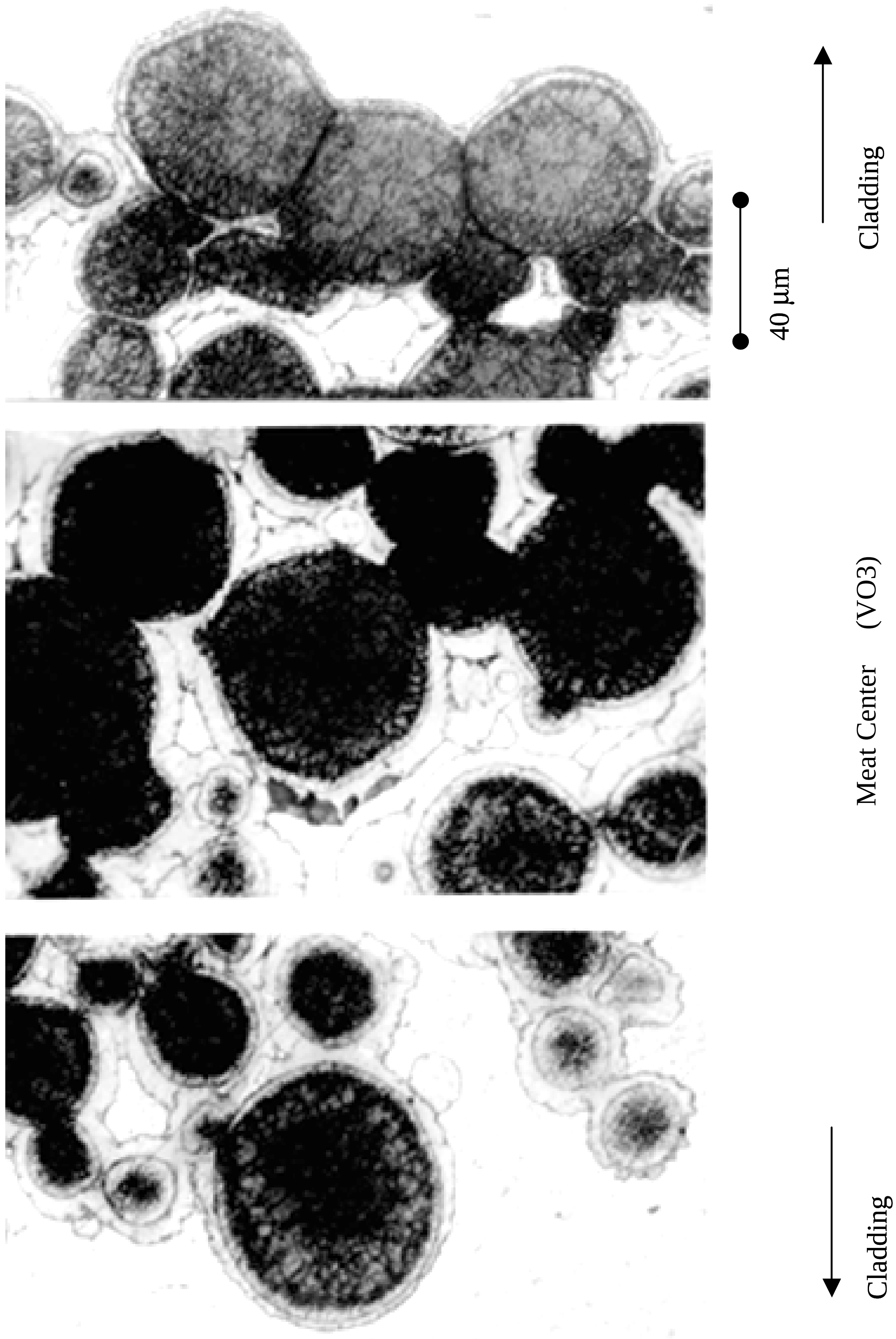


40% Bu 217ºC



40% Bu 65ºC

Figure 3. Fuel microstructure at low and high temperature U-10 Mo, showing apparent athermal fission gas behavior.


Cladding
40 µm
Meat Center (VO3)
Cladding

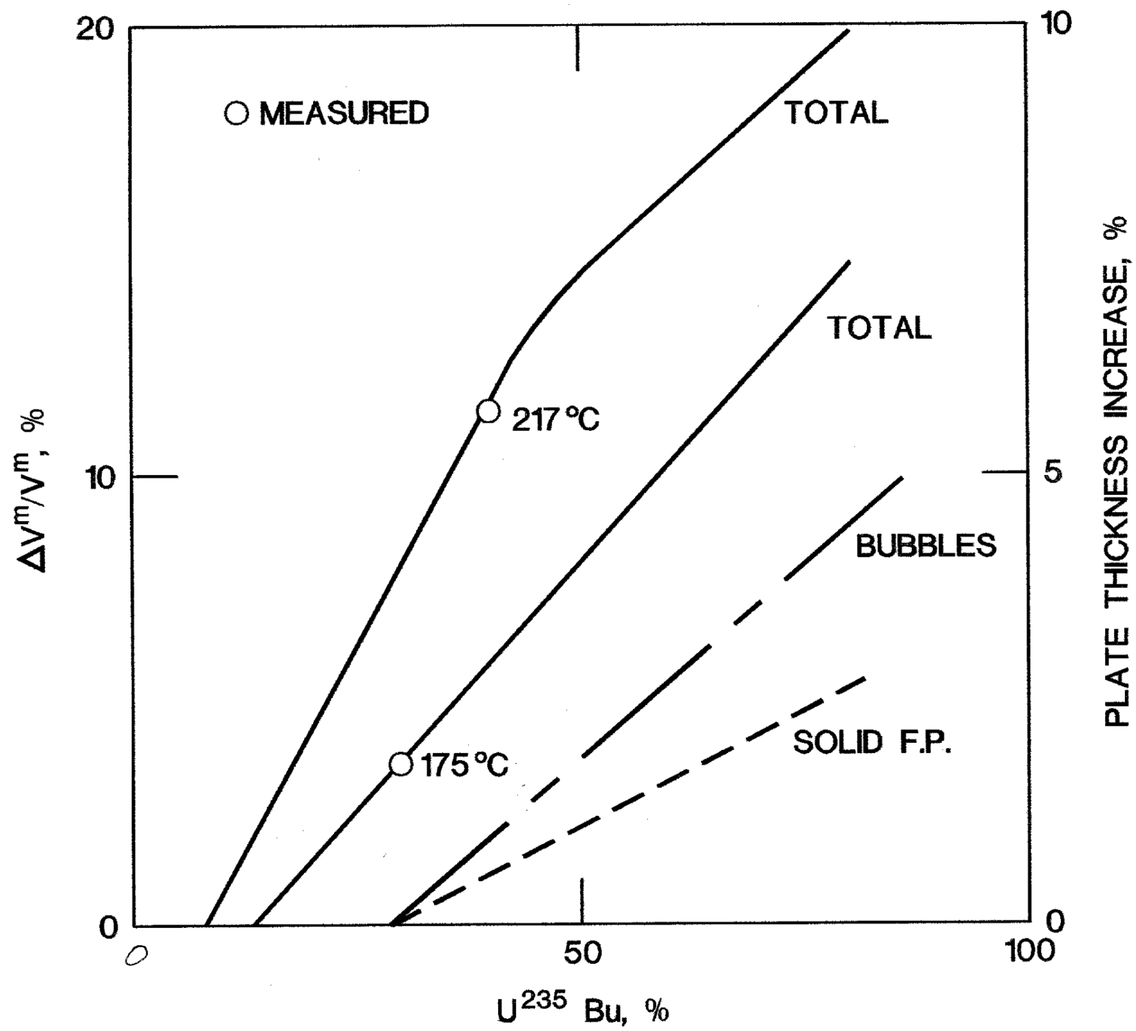


Figure 6. Projected fuel meat swelling and plate thickness increase for two fuel centerline BOL temperatures. (U-10Mo @ 8 g cm^{-3}, 0.060 inch thick plate with 0.030 inch thick meat).

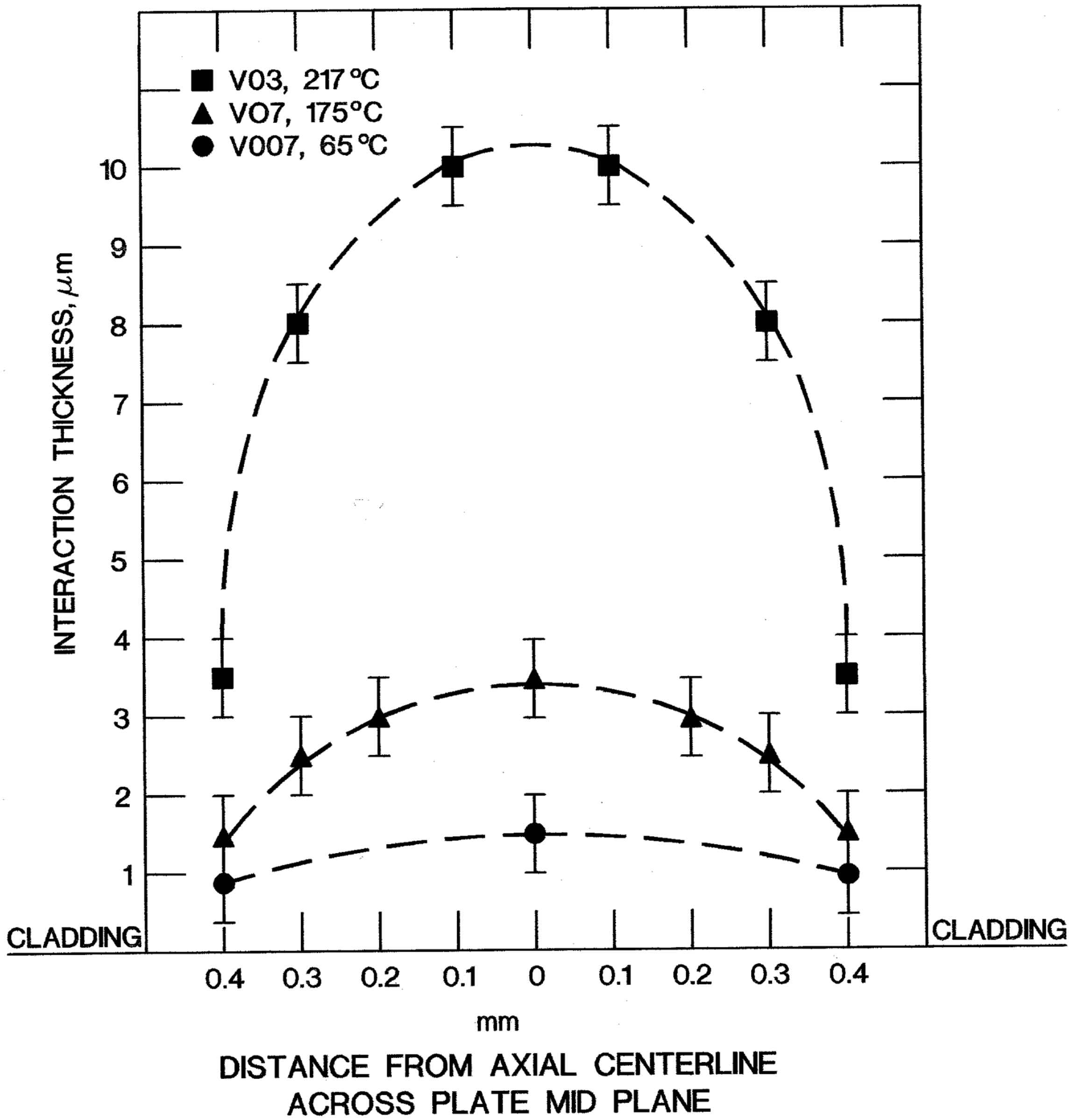


Figure 8. Thickness of U-10Mo/Al interdiffusion layer measured across the meat of samples irradiated at several temperatures.