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PROGRESS IN DEVELOPMENT OF LOW-ENRICHED U-MO DISPERSION FUELS

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ABSTRACT

Results from postirradiation examinations and analyses of U-Mo/Al dispersion miniplates are presented. Irradiation test RERTR-5 contained mini-fuel plates with fuel loadings of 6 and 8 gU cm^{-3}. The fuel material consisted of 6, 7 and 10 wt.% Mo-uranium-alloy powders in atomized and machined form. The swelling behavior of the various fuel types is analyzed, indicating athermal swelling of the U-Mo alloy and temperature-dependent swelling owing to U-Mo/Al interdiffusion.

1. Introduction

For the past several years the focus of the fuels area of the U.S. Reduced Enrichment for Research and Test Reactors (RERTR) program has been the development of aluminum-based dispersion fuels that will accommodate uranium densities in the fuel meat of 8 to 9 gU cm^{-3} [1]. Our primary focus has been on determining the irradiation behavior of candidate fuels. Thus far, data are available from irradiation tests of very small fuel plates—RERTR-1, -2, and -3—and most recently for larger "miniplates"—RERTR-5. The first two tests resulted in the identification of U-Mo alloys with Mo contents of at least 6 wt.% as very promising candidates [2]. The third test was focused on the behavior of the U-Mo fuels under high-temperature (up to ~250°C) irradiation conditions [3].

The miniplates irradiated in RERTR-5 contained either atomized or machined fuel particles ranging in composition from, nominally, 6 wt.% Mo to 10 wt.% Mo. The fuel plates in the RERTR-5 test 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 gU cm^{-3} of fuel meat. The experiment was irradiated in the Advanced Test Reactor (ATR) for 116 EFPD (effective full power days).

2. Postirradiation data

The volume of the miniplates was measured with the customary immersion method after removal of the boehmite surface layers. The meat swelling was calculated by subtracting the known cladding volumes. The resulting data plotted against beginning-of-life (BOL) peak meat temperatures are shown in Fig. 1. A linear normalization to 50% burnup (Bu) was applied to the swelling values to aid comparison. The BOL temperatures plotted in Fig. 1 only serve to illustrate the trend in swelling with increasing temperature. The actual fuel temperature is a complex function of irradiation time and position in the fuel meat. Not only are there temperature gradients in the fuel meat, but the temperature changes during the irradiation as a result of the competing effects of the decreasing thermal conductivity and U-235 burnup. This issue was treated in detail previously [4] and remains the subject of continuing study. Apart from the obvious temperature dependence of the meat swelling, there is a clear difference in swelling magnitude between atomized and machined fuel. The lower swelling of the plates containing machined fuel powder can be attributed to the higher as-fabricated porosity, 4 to 8%, in the meat of the plates, compared to 0 to 2% in plates containing atomized fuel powder. This effect of fuel swelling accommodation by as-fabricated porosity is also clearly evident for the three U_3Si_2 miniplates which measured ~0% meat swelling and contained ~7% as-fabricated porosity. The apparent differences in swelling in plates with 6 g cm^{-3} and 8 g cm^{-3} uranium loading, or plates containing either 6 or 10 wt.% Mo fuel primarily results from differences in the amount of fuel-aluminum interaction phase formed during irradiation. The amount of interaction phase formed depends on the ratio of fuel to aluminum matrix, fuel composition, and the temperature history of each particular plate. Detailed characterization of the dependencies requires extensive metallographic examinations and modeling, which is an ongoing effort. However, to date we have completed the examination of a set of U-10Mo plates covering both machined and atomized fuel, 6 and 8 g cm^{-3} uranium loading, and high and low irradiation temperatures. The results of this examination are discussed below.

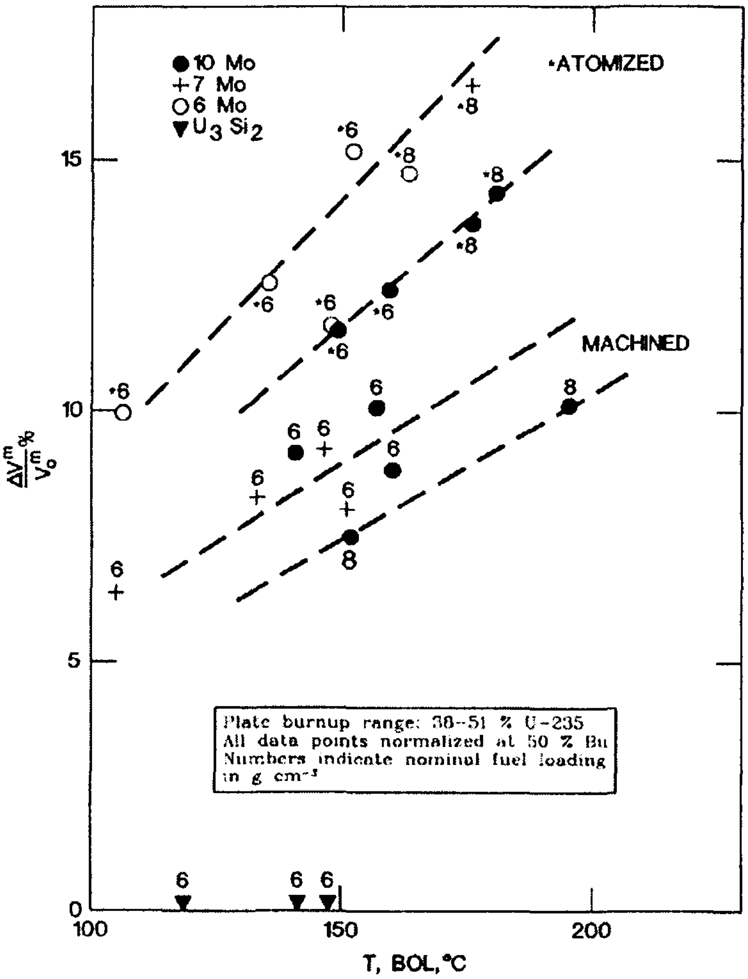


Fig. 1. Meat swelling of miniplates from experiment RERTR-5 as a function of Beginning-Of-Life peak meat temperature.

3. Metallographic Analysis

Metallographic samples taken from the center of the six U-10Mo miniplates listed in Table I were analyzed in order to characterize their swelling behavior. Examples of the meat microstructure of high- and low-temperature machined and atomized fuel are shown in Fig. 2. The effect of irradiation temperature on the extent of fuel-aluminum interdiffusion (represented by the light gray phase) and the diminution of aluminum matrix are the most prominent features.

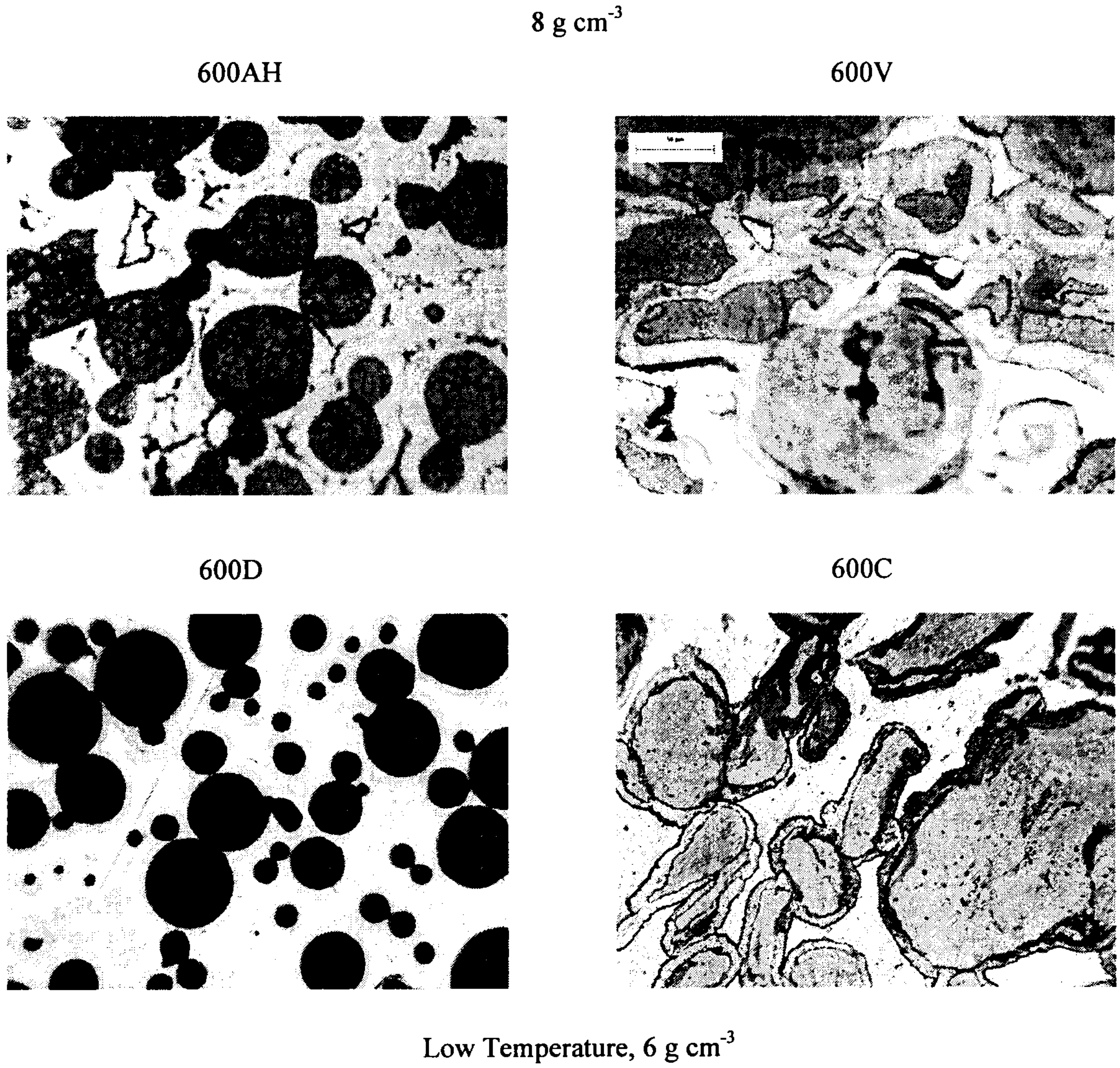


Fig. 2. Optical metallographic examples of machined and atomized U-10Mo fuel irradiated at ~100 to 200°C BOL temperature.

Quantitative image analysis was performed, yielding postirradiation volume fractions of the reaction product, unreacted U-Mo fuel, residual matrix aluminum, and porosity. These data were compared to preirradiation data from sibling miniplates to obtain the irradiation-induced changes in the meat.

3.1 Fuel –Aluminum Interactions

Characterization of the magnitude and kinetics of the interaction phase formation is an important item in the analysis of the irradiation behavior. Fuel-aluminum interaction is a major contributor to meat swelling and, as will be discussed in the next section, is the source of the temperature dependence of the meat swelling illustrated in Fig. 1. Moreover, the reaction product, which, based on available phase diagrams and out-of-reactor diffusion studies, appears to be a uranium aluminide compound (UAl_x), which has a very low thermal conductivity of ~6 W cm^{-1} K^{-1}. As it replaces matrix aluminum, with a conductivity of ~200 W cm^{-1} K^{-1}, it significantly increases the meat temperature during the course of the irradiation [4].

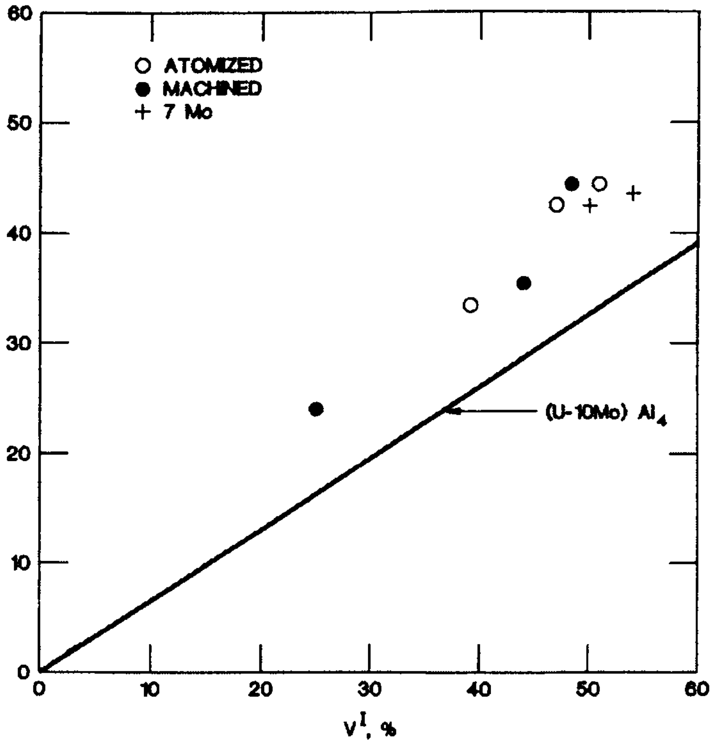


Fig. 3. Volume U-10 Mo/Al interaction phase versus volume of aluminum consumed in the formation of the interaction phase.

The measured quantities of interaction phase formed versus matrix aluminum consumed are plotted in Fig. 3, along with the calculated value for the highest aluminide compound known to exist in the U-Al system. The plot indicates that there is substantially more aluminum consumed than needed for the formation of $(U\text{-}10Mo)Al_4$. This observation was made previously during the analysis of RERTR-3 samples. The "missing" aluminum was accounted for by assuming that the aluminide compound was not stoichiometric, i.e., $x = 4.4$, and that a large amount of aluminum had diffused into the unreacted U-Mo fuel, residing there as solid solution [3]. The first assumption remains a possibility because UAl_4 is known to have a solubility range; it can, however, account for only a minor fraction of the missing aluminum. The second assumption requires postulation of a fission-induced solubility limit of aluminum in U-Mo that is several times higher than the maximum equilibrium value of 0.5 wt% for γ-U. Recent microprobe measurements indicate that the maximum aluminum content in the unreacted fuel is indeed only approximately 0.5 wt.%. This leaves the major fraction of the missing aluminum unaccounted for.

The explanation offered here is that, because of fuel and interaction product swelling, matrix aluminum is "squeezed" out to the periphery of the meat. This effect was indeed included in the DART code [5] to account for the observed irradiation behavior of U_3Si_2 dispersion fuel. The question can only be definitively settled when complete microprobe and planned neutron diffraction data become available.

4. Swelling Behavior

The swelling behavior was calculated in two ways: 1) using quantitative metallography data only and 2) using a combination of these data and the measured meat swelling data.

4.1 Reaction Product Swelling (U-10Mo)Al$_4$

The reaction 0.78U+0.22Mo+4Al results in a net volume increase of $\frac{V^I}{V^F + V^{Al}} = 1.16$ or 16%. Fission-induced swelling of (U-10Mo)Al$_4$ is assumed to be equal to that of UAl$_x$, and amounts to 0.05%ΔV/% U-235 Bu [6].

4.2 Fuel Swelling ΔV^F (U-10Mo)

1) $$\frac{\Delta V^F}{V_o^F} = \frac{\Delta V_m^F - \Delta V_o^F}{V_o^F}$$

where: V_m^F is the measured unreacted fuel volume, and

V_o^F is the as-fabricated fuel volume minus the fuel consumed in the formulation of interaction volume V^I. The results for the six U-10Mo miniplates are given in Table I.

2) $$\frac{\Delta V^F}{V_o^F} = \frac{\Delta V^m - \Delta V^I + \Delta V_e^p}{V_o^F}$$

where: ΔV^m is the measured meat swelling,

ΔV^I is the meat swelling due to interaction phase formation, and

ΔV_e^p is the difference between as-fabricated and measured residual porosity.

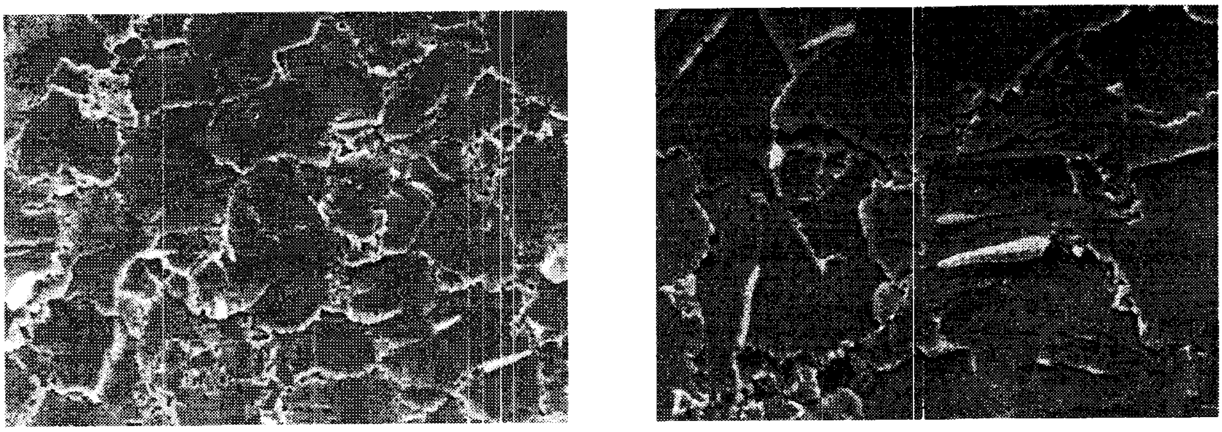
The data thus derived are also given in Table I.

Inspection of the resulting fuel swelling values indicates that fuel swelling is ~0.5%/%U-235 burnup and is independent of temperature. This conclusion is supported by the scanning electron micrographs shown in Fig. 4. The fission gas bubble morphology at various temperatures is very similar, with similar-size visible bubbles at the grain boundaries. It appears that the machined fuel has reached the initial stage of recrystallization; this was expected based on RERTR-1, -2, and -3 results.

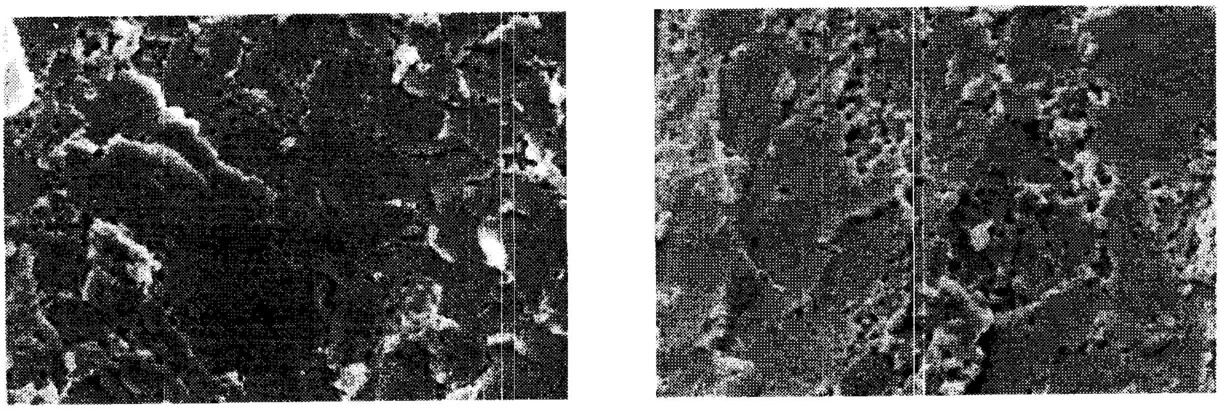
5. Miscellaneous

5.1 Cladding Failure

During the postirradiation examination of the RERTR-5 experiment, a small lateral crack was observed in the fuel zone near the trailing edge of plate Q8003I. Q8003I is one of a series of plates fabricated with a uranium loading of 8 g cm^{-3} using U-7Mo ground powder. A slight increase in coolant and stack activity was noted by ATR Operations on August 29, two days after irradiation start. Activity decreased to normal levels after approximately two weeks. Irradiation of RERTR-4 and RERTR-5 continued for 116 days, through the remainder of cycle 123B and cycles 123C and 124A with no further activity increase. Total irradiation time for RERTR-5 was 116 EFPD. It appears from the metallographic examinations that the failure of Q8003I was due to a thin area in the cladding that was introduced during fuel plate fabrication. The cladding measures 0.002-0.003 inches (50-75 μm) thick in the region of the failure. Fuel pile-up in this region increases stresses from thermal expansion and fuel meat swelling (owing to reaction and fission products) beyond the ultimate strength of the



Atomized



Machined

Fig. 4 Microstructure of unreacted U-10Mo fuel particles at low and high temperatures irradiated to 40 to 50% U-235 Bu (SEM).

thin area in the cladding. This is exacerbated by the additional local heat loading from the high-density 'dogbone' region of the fuel plate. More information is presented in a paper by A. Languille [7].

5.2 Comparison with U_3Si_2 Fuel

Three U_3Si_2 miniplates were included in test RERTR-5 to provide comparison with our previous experiments on this fuel type. As shown in Fig. 1, the swelling of the U_3Si_2 miniplates was measured as 0%. Metallographic examinations showed that the original as-fabricated porosity of ~7% was reduced to ~1%. This behavior is consistent with our previous experience. The fuel swelling rate of the U_3Si_2 is approximately 0.25%/%U-235 burnup. This is, considering the lower fission density at equivalent burnup when compared with U-10Mo, only fractionally lower than the latter. This is probably due to the absence of grain boundaries in U_3Si_2 and, therefore, the absence of grain boundary bubbles (see Fig. 5). The main difference is in the meat swelling component represented by fuel-aluminum interaction. As is clear from Fig. 5, the extent of interaction is much greater in U-10Mo. Moreover, the (U-Mo)Al_4 compound contains much more aluminum than the $U(Al_{0.75}, Si_{0.25})_3$ compound formed in U_3Si_2 dispersions while the volume increase associated with the latter is only

Table I. Results from fuel swelling analysis of RERTR-5 U-10Mo miniplates

Plate Number/ Position	**BOL T, °C**	**Bu, %**	**Interaction Depth, y, μm**	$\frac{\Delta V^F}{V_0^F}$,* %	$\frac{\Delta V^m}{V_0^m}$, %	$\frac{\Delta V^F}{V_0^F}$,** %	$\frac{\Delta V^F}{Bu}$
A8002L B00V C-4	195	47	15	20	9.5	22	0.43*/0.48**
V8005B$^\alpha$ 600 AH D-8	180	41	14	17	11.8	20	0.42/0.48
V6019G$^\alpha$ 600M B-4	149	49	13	20	12.4	28	0.42/0.57
V6018G$^\alpha$ 600D A-4	116	38	8	23	~8	22	0.59/0.58
A6008H 600Y C-7	160	49	13	27	8.7	22	0.56/0.45
A6005H 600C A-3	109	38	5	17	~6	17	0.46/0.45

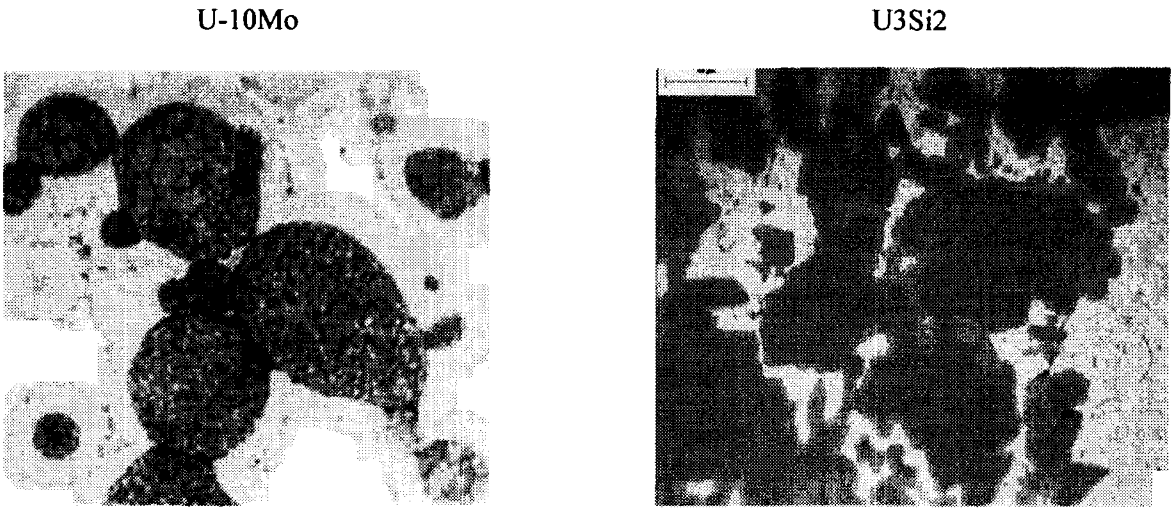
α atomized power
~ interpolated values
* by quantitative metallography
** by measured density

~3% versus 16% for the former. The main difference between U-Mo and U_3Si_2 fuel behavior is thus the kinetics and extent of fuel-aluminum interaction, making the U-Mo fuel swelling more sensitive to irradiation temperatures. The swelling of both unreacted fuel types is similar and athermal.

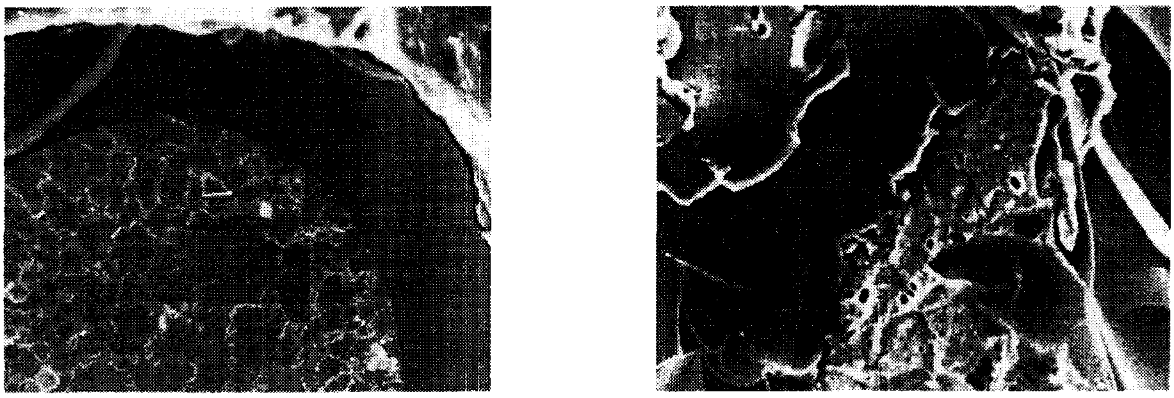
6. Conclusions

The following conclusions may be drawn from the initial results of the postirradiation examination of high-density dispersion fuel test RERTR-5.

- The extent of fuel plate swelling is acceptable and stable.
- Temperature dependence of swelling is due to temperature-dependent U-Mo/Al interdiffusion up to burnup where the matrix aluminum 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.



Optical



SEM

Fig. 5. Meat microstructure of U-10Mo and U_3Si_2 miniplates from RERTR-5 irradiated at BOL temperature of 149°C to ~50% U-235 Bu.

7. References

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