POST-IRRADIATION ANALYSIS OF LOW ENRICHED U-Mo/A1 DISPERSIONS FUEL MINIPLATE TESTS, RERTR 4 & 5

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Argonne National Laboratory's work was supported by the U.S. Department of Energy, Office of International Policy and Analysis (NA-212), national Nuclear Security Administration, under contract W-31-109-ENG-38.

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ABSTRACT

Interpretation of the post irradiation data of U-Mo/A1 dispersion fuel mini plates irradiated in the Advanced Test Reactor to a maximum U-235 burn up of 80% are presented. The analyses addresses fuel swelling and porosity formation as these fuel performance issues relate to fuel fabrication and irradiation parameters. Specifically, mechanisms involved in the formation of porosity observed in the U-Mo/A1 interaction phase are discussed and, means of mitigating or eliminating this irradiation phenomenon are offered.

I. Introduction

Results from the postirradiation examination of miniplate test RERTR 4 and 5, as well as irradiation test data were presented at the 2004 RRFM meeting [1]. Examinations have continued since and isotopic burnup analyses have been performed. The results to date are briefly reviewed and summarized. A preliminary analysis of the nature and behavior of the U-Mo Al interaction product is presented, including proposed means of eliminating the porosity formation associated with the interaction.

II. Metallographic Analysis

Examples of transverse cross sections taken at the axial midplane of several miniplates are shown in Fig. 1. These sections represent the microstructure of mini-plates positioned along the length of the test assembly.

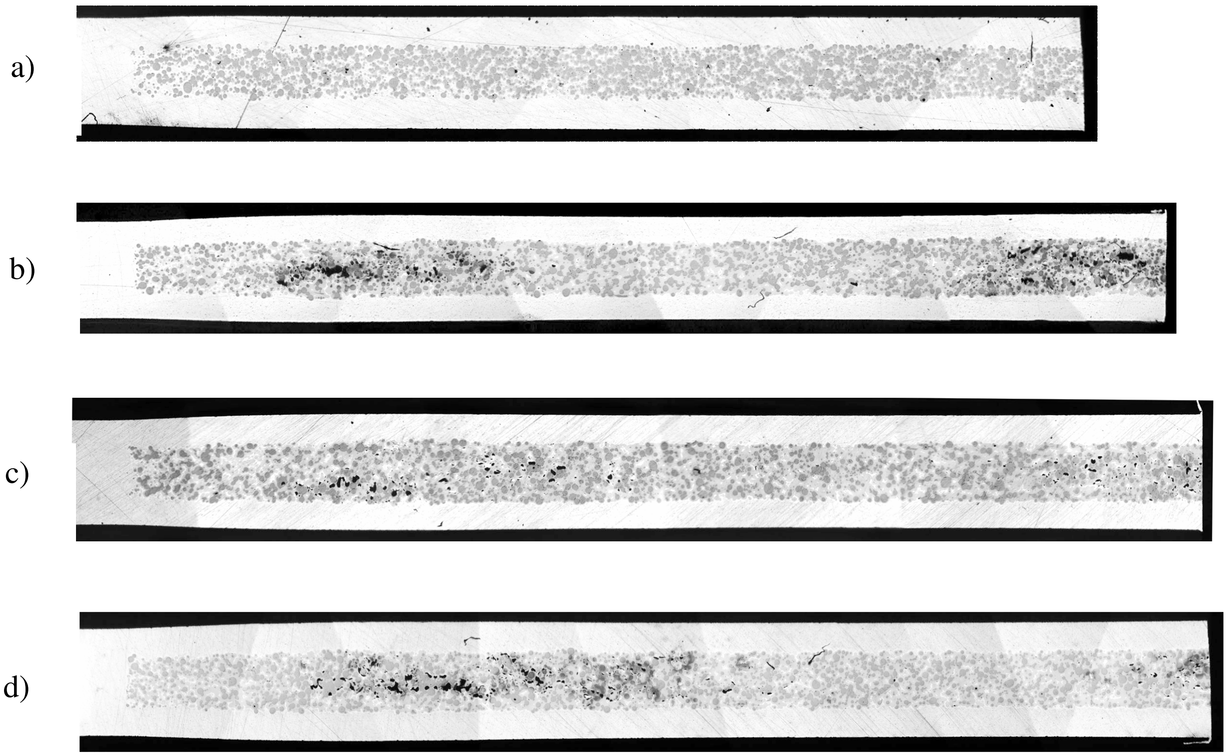


Figure 1. Transverse cross section of U-xMo RERTR 4 mini-plates containing atomized fuel powder. a) V6001M -10Mo, Capsule A, b) R6003F-7Mo, Capsule B, c) S6004C-6Mo, Capsule C, d) V6022M-10Mo, Capsule D.

All but the miniplates from the upper test tier of the assembly, exhibit local areas with relatively large size gas pores. These areas coincide with the peak fission rate positions in the plates.

Meat swelling data of individual plates at the various axial positions in the assembly area are shown in Fig. 2. The swelling data for RERTR -5 (peak ^{235}U burnup of 50%) are from plate immersion volume measurements, whereas the data for RERTR-4 (peak burnup of 80%) were derived from the center of the metallographic sections. The difference in swelling in test RERTR-5 between miniplates made with mechanical and atomized fuel powders is due to the larger as-fabricated porosity in the former. Although there were signs of initial local porosity formation in test RERTR-5, the magnitude did not register in the overall meat swelling.

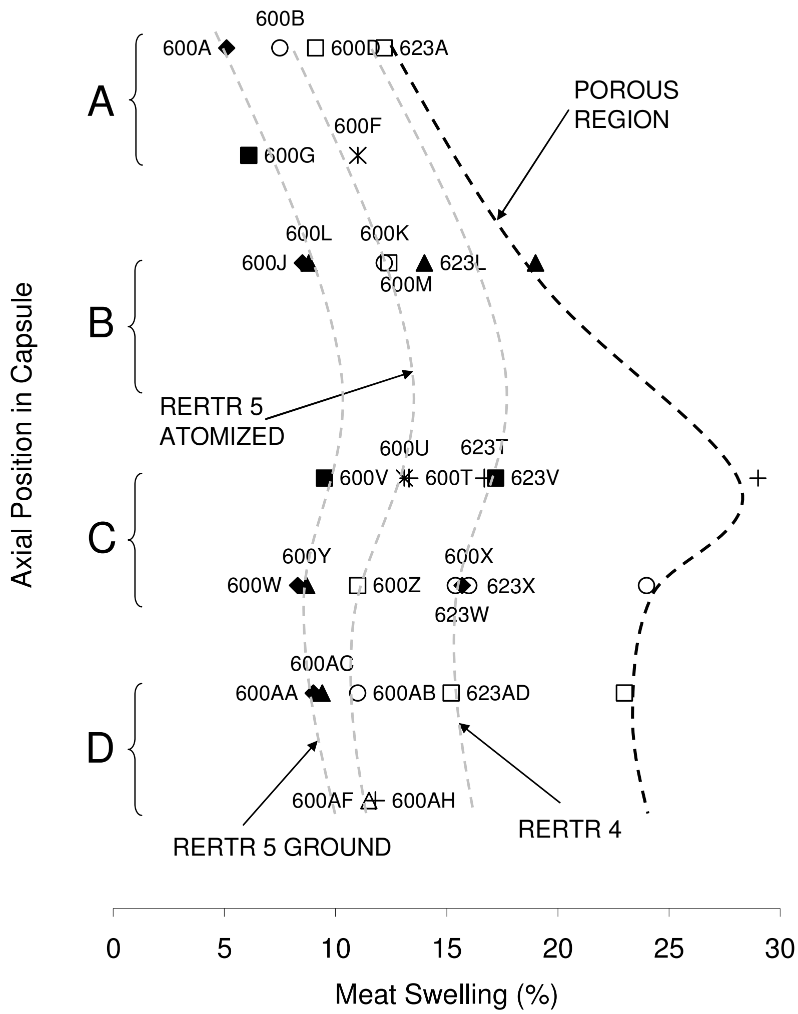


Figure 2. Meat swelling of mini-plates from RERTR-4 and RERTR-5 experiments as a function of their position in the irradiation device. The device consisted of 4 capsules, each capsule contained two rows of four plates. All measurements are taken in the non-porous, center region of the plate, except for the single data set as marked.

In the higher burnup test, RERTR-4 however, there is a clear difference in meat swelling between the central part of the plate and the location with porosity see Fig. 3. The axial distribution of these different locations is shown in Fig. 2. The additional swelling in the porous regions can be accounted for by the measured pore volume.

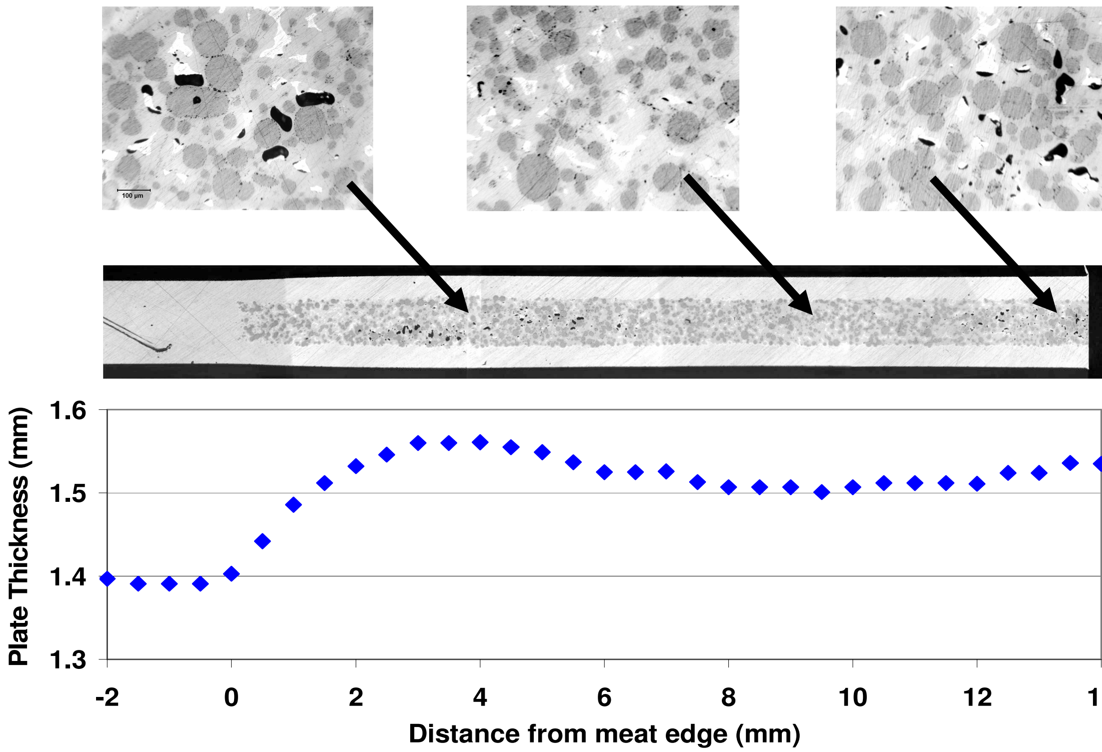


Figure 3. Plate thickness across the width of mini-plate S6004C-6Mo (RERTR-4).

Quantitative metallography was performed on several fuel plates to determine the volume fraction of the constituents of the fuel meat. The data in Table I combined with meat swelling values were used to determine the U-Mo fuel particle swelling as shown in Fig. 4 for U-10 wt. % Mo. The fuel swelling, as previously reported, can be divided in two regions, separated by the onset of recrystallization of the fuel.

The fuel swelling rate and magnitude over the entire range test conditions remain remarkably stable and predictable.

Table 1. Initial and final volume percent of fuel (VoF – initial), aluminum (VoAl – initial) and interaction (V I). Note final totals do not add up to 100 as values have been corrected for swelling. Y – Interaction layer thickness, ΔVm – meat swelling.

Plate No.	VoF (%)	VoAl (%)	Y (µm)	V F (%)	V I (%)	V Al (%)	ΔVm (%)
RERTR-5							
V6018G	39.3	60.1	7	36.9	30.5	29.6	9
V8005B	52.1	47	14	53.1	48.4	11.5	12
V6019G	39.3	60.1	15	39.7	58.4	17.2	15
RERTR-4							
V6001M	39.3	60.1	12	43.8	55.8	12.6	12
V6022M	39.3	60.1	17	39	67.1	9.1	15
V6015G	39.3	60.1	19	41.3	65.3	7	13

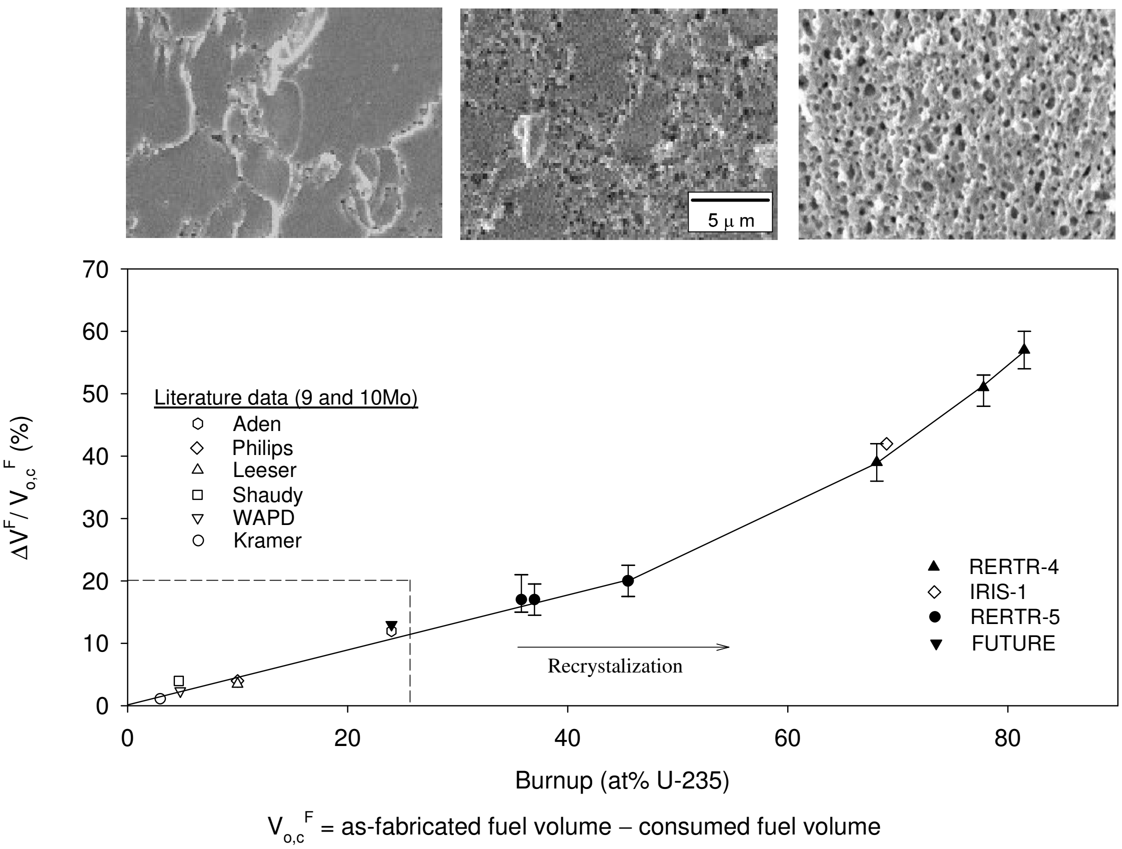


Fig.4 Swelling behavior of U-Mo vs. burnup

III. Irradiation Behavior of Fuel-Al Interaction product

The most striking observation to be made from the metallography of those areas of the miniplates, and the full size plate of the FUTURE experiments, that have a developed large pores is the, apparently, viscous nature of the material (the fuel-Al interaction product) in which the pores grow in Fig. 3. This behavior resembles that previously found in U_3Si and U_6Fe (see section B). In the case of these uranium compounds, this fluid-like behavior was attributed to fission induced amorphization (i.e. transformation to a metallic glass) of these compounds and a subsequent large decrease in viscosity and increase in gas mobility. Amorphization of a crystalline material is usually accompanied by an increase in volume-a quantity called 'free volume' which facilitates atomic mobility and shear deformation analogous to the liquid state.

Doolittle [2] has developed an expression for the fluidity Ǿ or the viscosity η of amorphous materials.

$$Ǿ = \eta^{-1} = A'' \exp [-B^1/V_f] \quad (1)$$

Where V_f is the free volume

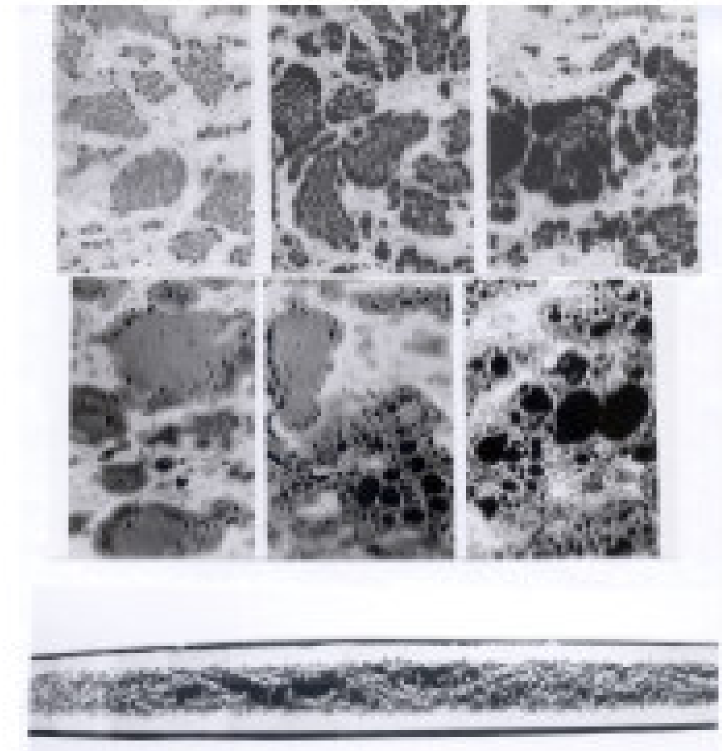
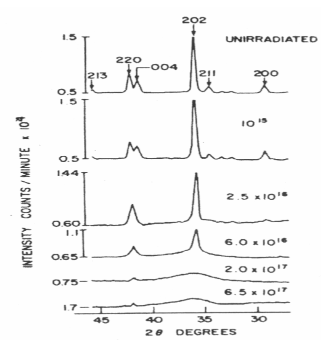
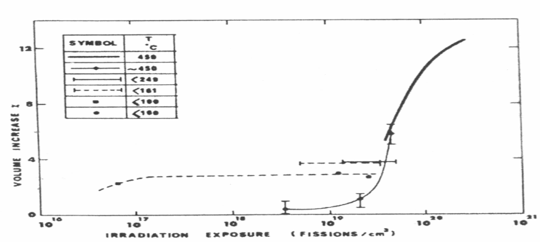


Fig.5 Examples of break-away swelling in amorphous U-compounds



X-ray diffraction patterns for irradiated U_3Si. Numbers indicate exposures in fissions/cm^3.



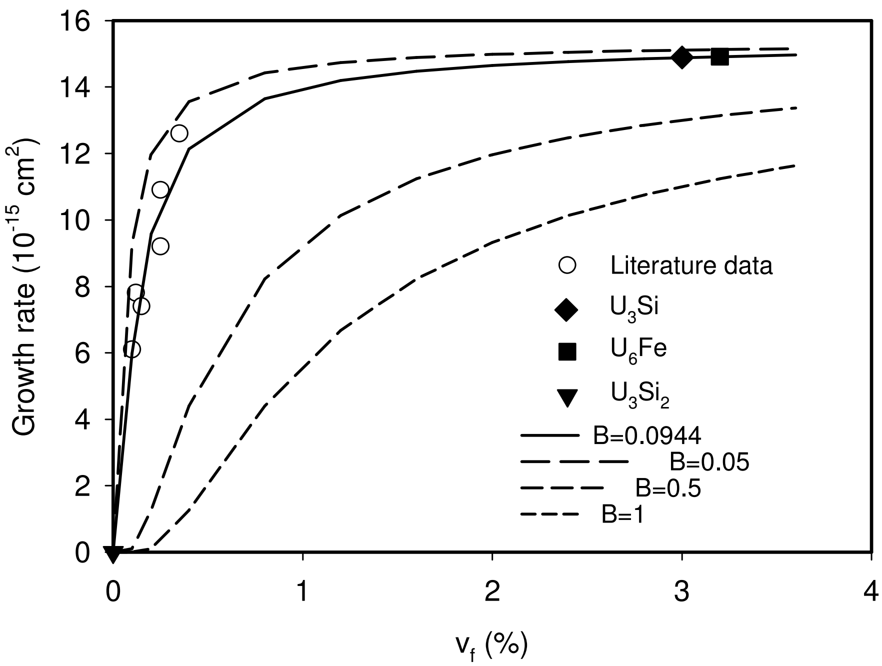
Volume increase as a function of exposure for U_3Si.

Fig.6 Volume increase as a function of exposure for U_3Si

A” is the fluidity per unit fision
And B^1 = Nv*f the number density per volume entities with a value larger than some critical value that allows for a diffusion event to occur.

The free volume V_f is a material-specific property, its value is reached at a relatively low damage dose before any appreciable fission product swelling has occurred, as for example in U_3Si in Fig. 6. The quantity B^1 however, depends in some way on the irradiation damage rate in the case of nuclear fuel materials.

Figure 7 depicts the, in this case, ion irradiation induced fluidity for several compounds, showing the correlation between Ǿ, (η) and V_f. The free volume model is consistent with the irradiation experience, amorphous U_3Si and U_6Fe having a large value for V_f whereas amorphous U_3Si_2 with virtually zero V_f does not. Although this analysis is far from rigorous, it does allow the following conclusion: Amorphized uranium compounds having appreciable free volume exhibit fluid-like behavior and growth of large fission gas bubbles when gas accumulates with burnup. In the absence of hard evidence i.e. neutron or x-ray diffraction, it is presumed that the (U-Mo) Al_x interaction product is amorphous and has a relatively large value for free volume, accounting for its fluid-like behavior under fission damage.



$$\phi = 15.4\, exp\left(-B/\mathrm{v}_f\right)$$

Fig.7 Fluidity vs. free volume

Experiments with ion irradiation on metallic glasses [3] have shown that Ǿ has a relatively weak temperature dependence in the operating range of our fuel tests. The fission (damage) rate dependence that may enter the parameter B^1 in equation (1) needs further study. However, it appears that the fluidity is overwhelming controlled by the free volume, V_f. It appears therefore, that the solution to the problem of porosity formation lies in the reduction of this parameter. As this parameter is primarily a function of composition, in essence in the nature and strength of the chemical bonds of the materials, appropriate alloy additions to the constituents involved in the formation of the interaction product appears to be the most effective means to stabilize the U-Mo/Al interaction product.

IV. The Case for Silicon

Alloy theories show that elements of group IV A in the periodic table to the right of Al should strengthen the bonds in U-Al compounds. Indeed Si, Ge and Sn promote the formation of UAl_3 and suppress the formation of the higher Al compound, UAl_4 in U-Al alloys [4]. As the interaction product formed in U-Mo/Al dispersions consists of a mix of relatively weak components in the range of Al_{4-7}, it is likely that these group IV elements will have a similar effect, i.e. promote the formation of (U-Mo) $(Al,Si)_3$. Early diffusion couple tests with U-Mo against a Al-Si alloy appear to substantiate this [5]. The interaction zone formed in this experiments was indeed (U-Mo) $(Al,Si)_3$, very similar to the interaction product formed between U_3 Si_2 and Al, which was found to be stable in dispersion fuel irradiation tests.

Although this compound amorphized in pile, the extra Si bonds evidently reduced the free volume in the amorphous interaction product and the next mini-plates irradiation test (RERTR-6) will therefore include several U-Mo dispersions with a range of Al-Si alloy matrix composition to hopefully establish the efficacy of Si in stabilizing the interaction product against large pore formation.

V Conclusions

Irradiation test of LEU, U-Mo dispersion fuel have shown that the irradiation behavior of this uranium alloy fuel is very stable and predictable. However, the interaction phase formed by interdiffusion of U-Mo and matrix Al develops large fission gas pores at high power densities. It appears that this interaction phase is amorphous and of low viscosity under fission damage. Incorporation of Si into the interaction phase is thought to improve the irradiation behavior of the U-Mo/Al dispersion fuel.

VI References

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