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RESULTS OF LOW ENRICHED U-Mo DISPERSION FUEL MINIPLATES FROM IRRADIATION TEST, RERTR-6

- Preliminary analysis of the effect of silicon on the irradiation behavior of U-Mo/Al dispersion fuel -

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ABSTRACT

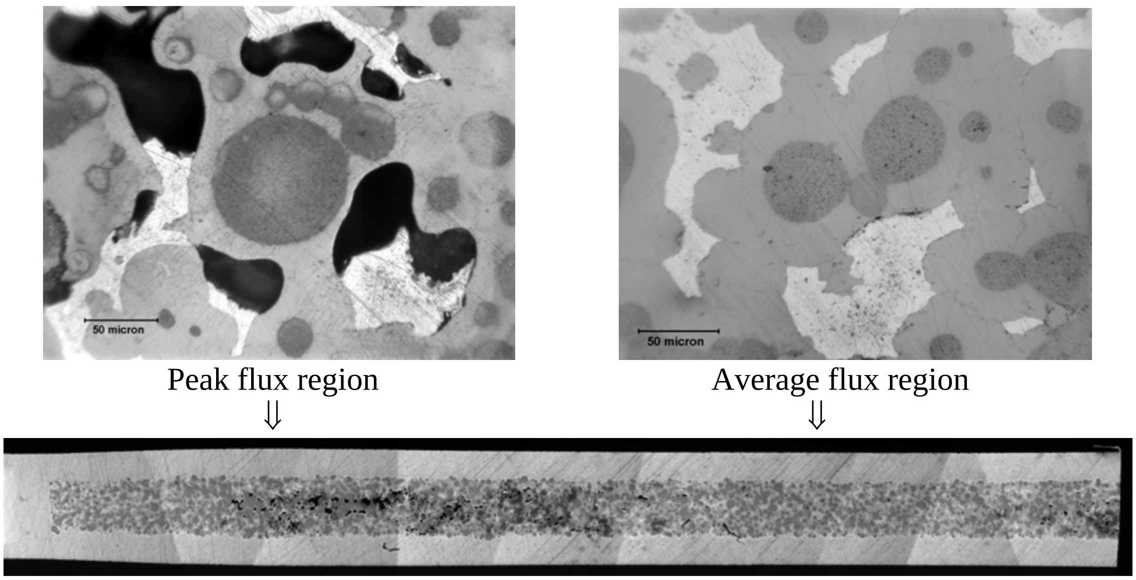
The irradiation of U-Mo fuel dispersed in a matrix of aluminum results in the formation of an interaction product that coats the fuel particle surface. In some instances pores form at the interface between the interaction product and the aluminum matrix. Depending on the irradiation conditions, the pores may grow, and linkup to form larger pores, or in severe cases form a continuous network that results in unacceptable pillowing of the fuel plate. Such observations have been made in the US and in other international irradiation experiments. Metallurgical and thermodynamic analysis suggests that the interaction product occurring in U-Mo/Al dispersion fuel may be rendered stable through the addition of Si to the Al matrix, and Zr or Ti to the U-Mo fuel alloy. The initial results of irradiation tests with Si additions are presented, showing that appropriate amount of Si additions to Al matrix substantially reduce the extent of U-Mo/Al interdiffusion and eliminate the formation of porosity.

1. Introduction

As the first destructive postirradiation examination (PIE) data from the RERTR-6 experiment are now available, it is possible to report a preliminary assessment of the effects on the irradiation behavior of the various amounts of Si that were added to the Al matrix of the dispersion miniplates in RERTR-6. The Si addition is our first attempt to eliminate the gross porosity that was observed during the PIE of miniplates from RERTR 4 and 5 [1], the French full-size plate tests, FUTURE [2] and IRIS-2 [3], as well as the Russian hexagon tube test IVV-2M [4].

An example of this porosity is shown in Fig. 1. The large pores (black) appear to grow in the interaction phase (grey) that was formed by interdiffusion of the U-Mo fuel (dark grey spheres) and Al matrix (white) during irradiation. The appearance of this porosity is identical in all tests mentioned above and is a true performance-limiting irradiation behavior feature of the UMo - Al combination.

Although it is difficult to separate operational parameters such as fuel temperature and fission rate (power density), examination of all available PIE data suggests that fuel fission rate is the controlling factor determining the initiation and growth of the porosity. As shown in Fig. 2, only experimental fuel plates operating at fuel fission rates below ~ 2.5×10^{14} $cm^{-3}s^{-1}$ attained high enough U-235 burnup to be considered having acceptable irradiation behavior.



RERTR-4 V6022M (623AD) U-10Mo/Al 80% burn-up

Figure 1. An example of porosity in U-10Mo/Al dispersion fuel irradiated to 80% U-235 burnup (RERTR-4 623AD).

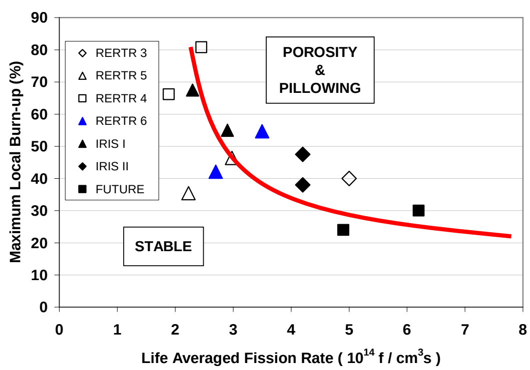


Figure 2. A burnup vs. fission rate diagram showing the boundary between stable and porosity-pillowing irradiation behavior.

Any further development of high density LEU dispersion fuel hinges, therefore, on our finding a way to eliminate the porosity problem. In our attempt to do this, we were guided by the following observations and analyses to the selection of Si as the most promising stabilizing element.

Table 1. Observations and analysis of Si effects on interaction product.

Related Literature [5-15]

- ❑ Al-12Si to reduce reaction between U in Al cladding: *(Si added in Al)*
  - D.R. Green (1957) L.S. DeLuca and H.T. Sumsion (1957)
- ❑ U-Al alloy studies to reduce UAl_4 formation in order to improve fuel fabricability: *(Si added in U-Al alloy)*
  - W.C. Thurber and R.J. Beaver (1959) R. Boucher (1959) N.E. Daniel et al. (1959) A.K. Chakraborty et al. (1970)
- ❑ Metallurgical analyses to select effective alloying elements
  - H.E. Exner and G. Petzow (1969)
- ❑ Indications from irradiation tests:
  - U-(Al,Si) : A.E. Richt et al. (1962) W. Dienst et al. (1977)
  - Difference between various U-silicide/Al dispersions : G.L. Hofman et al. (1996)
  - Interaction of (U,Mo)/Al and (U,Mo)/Al6061: M.K. Meyer et al. (2002)

Recent Analytical Work [16-23]

- ❑ Thermodynamic evaluation of stabilizing element additions to U-Mo/Al dispersion
  - Miedema analysis: Y.S. Kim (2005)
  - Atomistic modeling: G. Bozzolo et al. (2005)
  - Phase diagram establishment: F. Mazaudier et al. (2006)
- ❑ Ex-reactor high-temperature interdiffusion studies
  - Mirandou et al. (2004) J.M. Park et al. (2005, 2006) D.D. Keiser (2005)
- ❑ Ion beam irradiation studies
  - R. Birtcher (2005) N. Wieschela et al. (2005)

As discussed in some detail previously [24], the addition of Si to the Al matrix would not only reduce the rate of U-Mo/Al interdiffusion, as indicated by the work cited above, but also reduce the fission-induced high fluidity of the amorphous interaction product that is thought to be the root cause of the gross porosity formation.

2. Irradiation Details

RERTR-6 was a scoping test designed to evaluate the performance of a range of modified-matrix dispersion fuels and irradiate the first monolithic fuels. Results of the latter are presented by M. R. Finlay in this conference [25]. The experiment was loaded into the Advanced Test Reactor (ATR) in May 2005 in experimental position B-12. It remained in the ATR for three cycles for a total of 135 EFPD. Average burn-up of the 19.7% enriched fuel was 35 – 49 % depending on the location in the experiment. BOL fuel temperatures were < 200^{o}C and peak heat flux was 140 W/cm^{2}. The position of each plate within the experiment basket is shown in Fig. 3. The RERTR-6 experiment consisted of four capsules A-D, each containing eight plates. More information on experiment design is reported in [26]. The power history of plates from RERTR-4, 5 and 6 is shown in Fig. 3. The experiment basket is positioned so that one edge of the miniplates faces the core. A neutron flux gradient exists across the width of the plates and by rotating the basket by 180^{o} between cycles, which was done for RERTR 4 and 5, the burn-up becomes more uniform. IN RERTR-6 the basket was not rotated between cycles and hence one edge ran at ~35% higher power that the other.

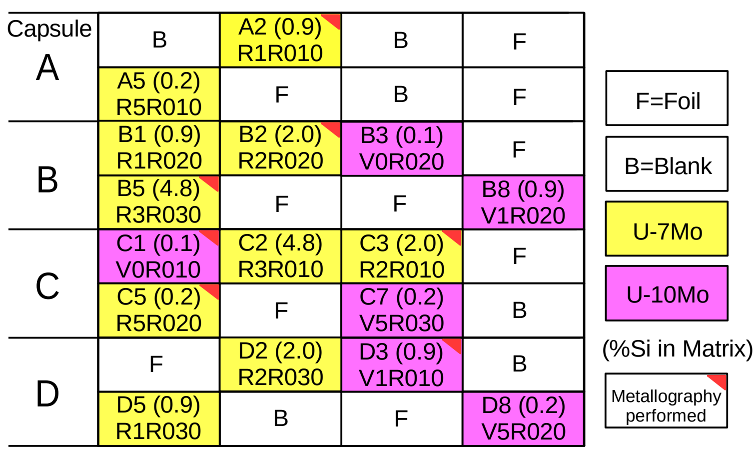


Figure 3. Schematic diagram of the RERTR-6 experiment showing location and composition.

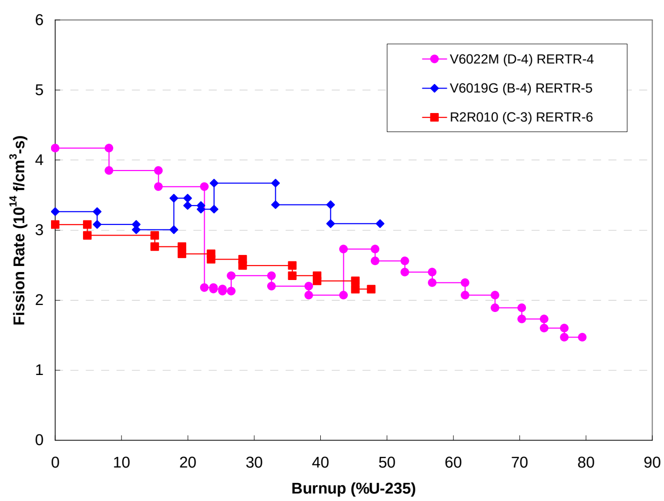


Figure 4. Power history of selected miniplates from RERTR-4, 5 and 6.

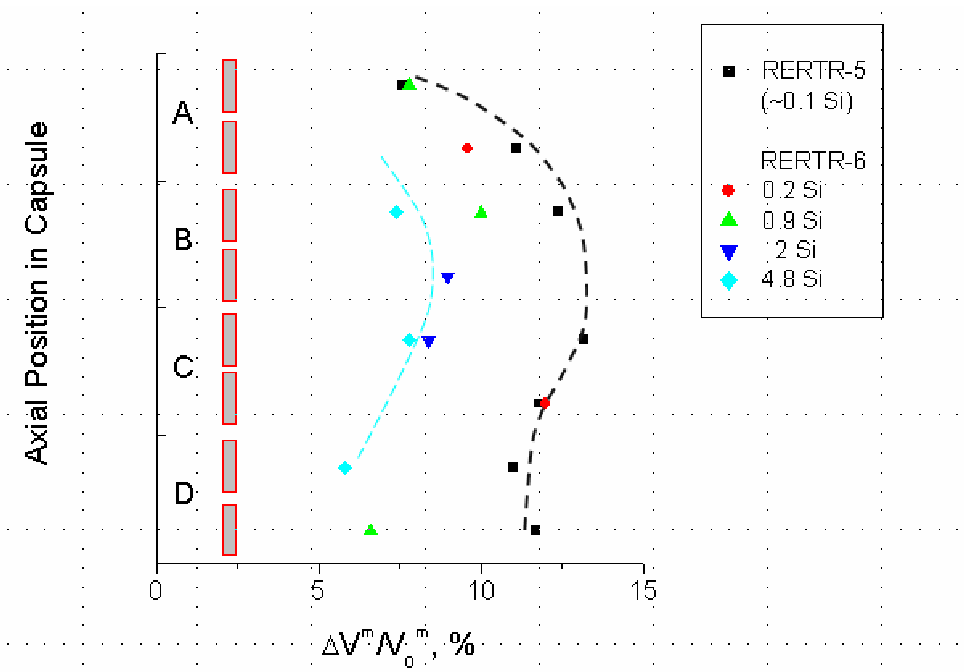


Figure 5. Comparison of meat swelling of miniplates from RERTR-5 and 6 at plate center (~50 % U-235 burnup)

3. Postirradition Examination Data

Results of the nondestructive examination, chiefly miniplate thickness measurement, have been reported previously [24]. These results showed that the fuel meat swelling, calculated from thickness data, was significantly reduced for miniplates that contained 2 % or more Si (see Fig. 5). This was presumed to be due to the expected effect of Si on the reduction of the amount of interaction and the increase of the density of the interaction product. Metallographic examination clearly confirms the former, as shown in a comparison of a plate with 2% Si and one with only 0.2% Si in Fig.6.

The difference in meat microstructure of the two plates is striking. There is substantial interaction and Al matrix reduction in the low-Si plate as well as gross porosity formation at the highest flux edge of the plate. The microstructure is virtually identical to that in similar plates from RERTR-5[1]. The interaction layer in the high-Si plate is not only much thinner, but there is no evidence of gross porosity formation in the interaction product.

A more complete comparison of microstructures for the range of Si addition, i.e. 0.2, 0.9, 2 and 4.8% is shown in Fig. 7. The interaction layer thicknesses in the 2% Si and 4.8% Si are very similar. The other interesting observation is that the microstructure at the highest flux position of the 0.9%-Si (Al6061) plate in the 'D' capsule appears similar to that in the 0.2%-Si plate and in the plates from RERTR-5, i.e., there appears to be no beneficial effect of a modest Si addition for the irradiation condition at this location in the fuel plate.

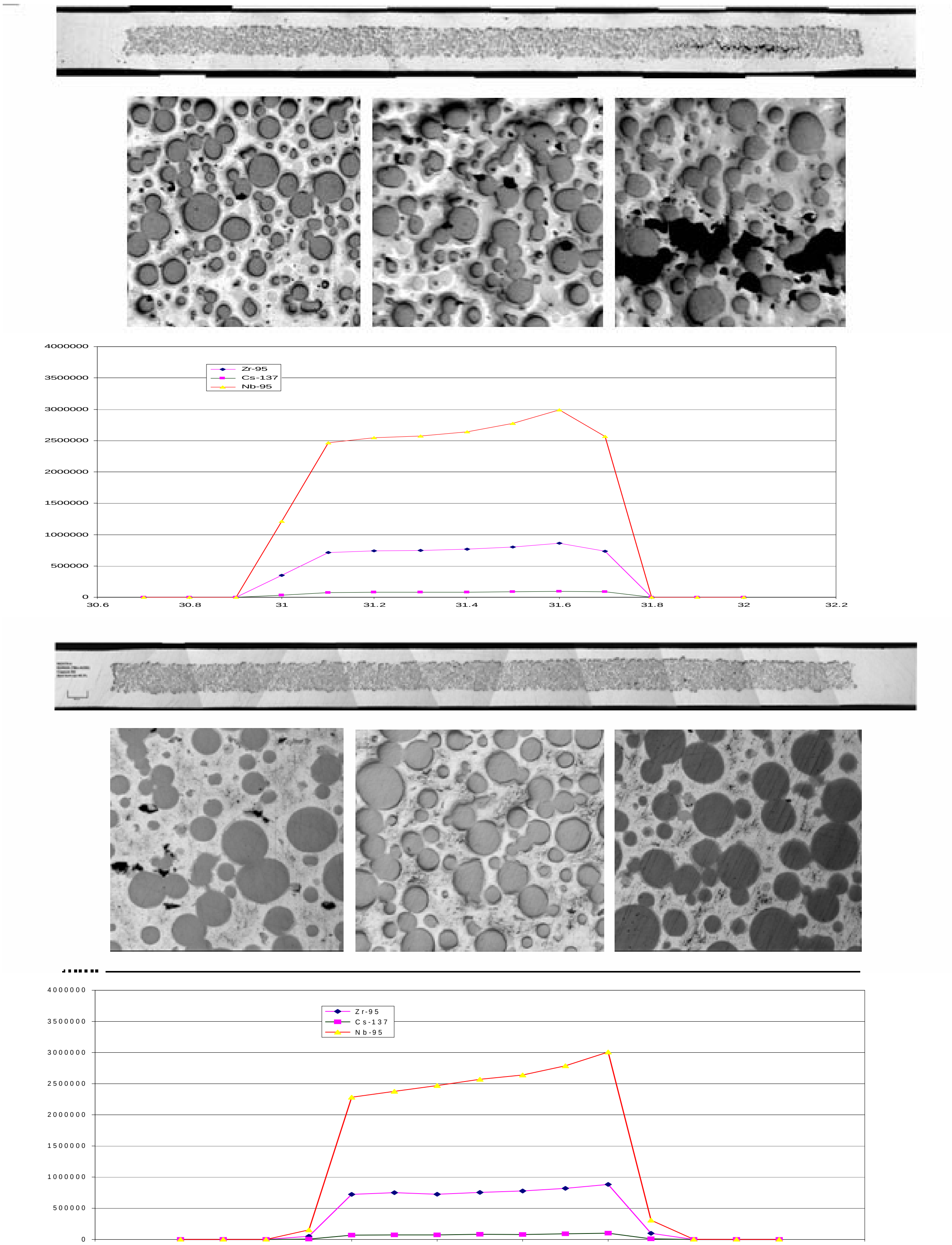


Figure 6. Fuel meat microstructure and gamma scan of low Si (0.2%, upper) and high Si (2%, lower) matrix miniplate from RERTR-6 (transverse section at mid-length).

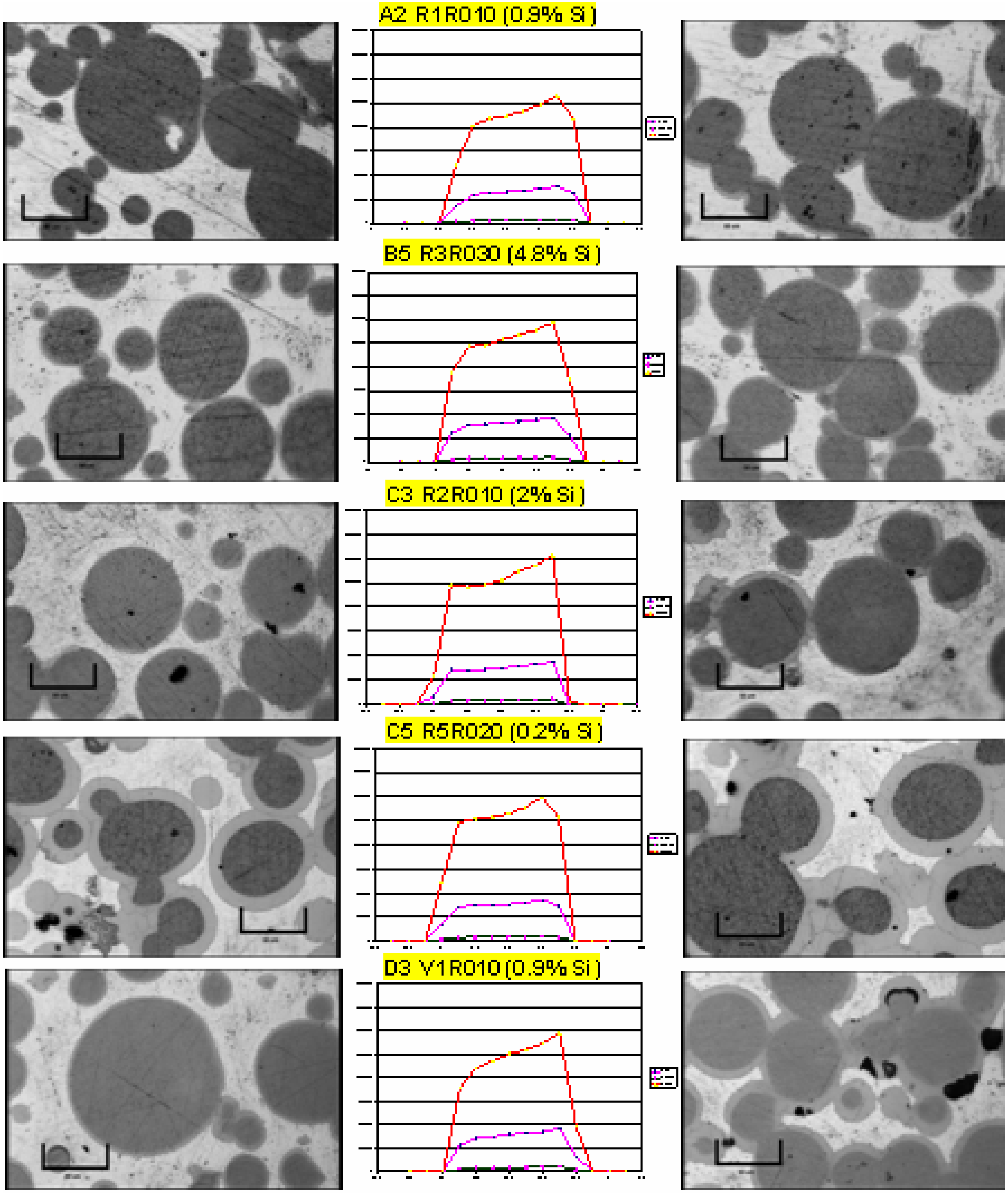


Figure 7. Comparison of fuel meat microstructures of RERTR-6 miniplates (transverse section at mid-length, BU ~ 50% U-235).

4. Discussion

The main purpose of the present irradiation experiment was to prove our hypothesis that if Si could be incorporated in the UMo-Al interaction product, the fission gas diffusivity and fission-induced fluidity of the interaction product would be reduced so as to eliminate gross porosity formation. The PIE data of the RERTR-6 test show that for the experimental conditions of the test, an appropriate amount of Si added to the Al matrix has indeed eliminated the porosity problem. In addition, the extent of interaction is drastically reduced.

The quantitative difference between low Si (0.2%) and high Si (2%) is shown in Fig. 8, where a correction to our diffusion correlation has been made to account for the small difference in power density and temperature. At the final burnup, there is a factor of 4 reduction in the width of the interaction layer.

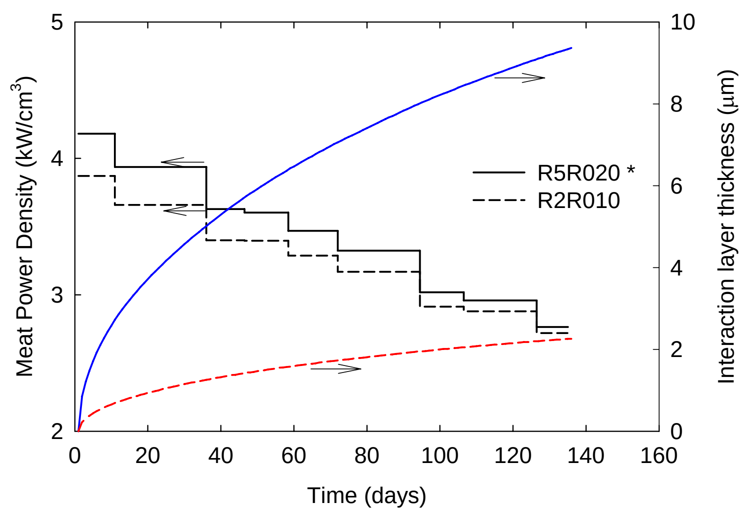


Figure 8. Comparison of power density and interaction layer thickness of R5R020 (0.2% Si) and R2R010 (2% Si).

Because of the rather wide range of matrix Si compositions employed in the test, the data permit an evaluation of the minimum amount of Si required. This amount appears, for the present test conditions, to lie somewhere between 0.9% Si (Al6061) and 2% Si (see Fig. 6).

The microstructure of the 0.9%-Si plate in position D3 captures this minimum amount. If we proceed on the assumption that a concentration of at least 5 at.% is needed in the IL to “stabilize” its behavior – this level is based on the equilibrium U-Al-Si diagram and the assumption that Mo and irradiation effects do not significantly alter this – we have a situation as shown in Fig. 9.
The upper curve is obtained based on the assumption that all available Si in the matrix can accumulated in the IL, and the lower curve is based on the assumption that only Si from the fission fragment recoil zone is available to accumulate in the IL. The IL thickness at the low flux region is 2.5 µm. For both of the curves, the Si content in the IL can be maintained at the 5 at.%

limit. In the high flux region, the IL is 7 µm thick, and the available Si is not sufficient to maintain the composition of the IL at the 5 at.% Si limit.

Thus, somewhere between the two extreme flux levels, lack of Si supply to the IL causes its irradiation behavior, i.e., high rate of formation and susceptibility to gross porosity formation, to revert to that of low-or no-Si conditions.

Figure 10 shows the relation between expected stability of the IL, its thickness and the minimum amount of Si. Choice between the upper curves (all Si in Al matrix available) and lower curves (only Si from recoil zone available) is clarified by the microstructure of a high Si (4.8%) plate shown in Fig. 11. It is clear that only the Si from the recoil damage zone around the fuel particles participates in the interdiffusion process that forms the IL. Evidently the neutron flux damage in the rest of the Al matrix is not affecting the process. For fuel loadings higher than 6 $gUcm^{-3}$, however, the recoil zones will increasingly overlap, and this should be taken into account.

In general, analysis of the present results and the forthcoming data from the higher-power test, RERTR-7, are expected to permit design and qualification of UMo-Al dispersion fuel with predictable and satisfactory irradiation behavior.

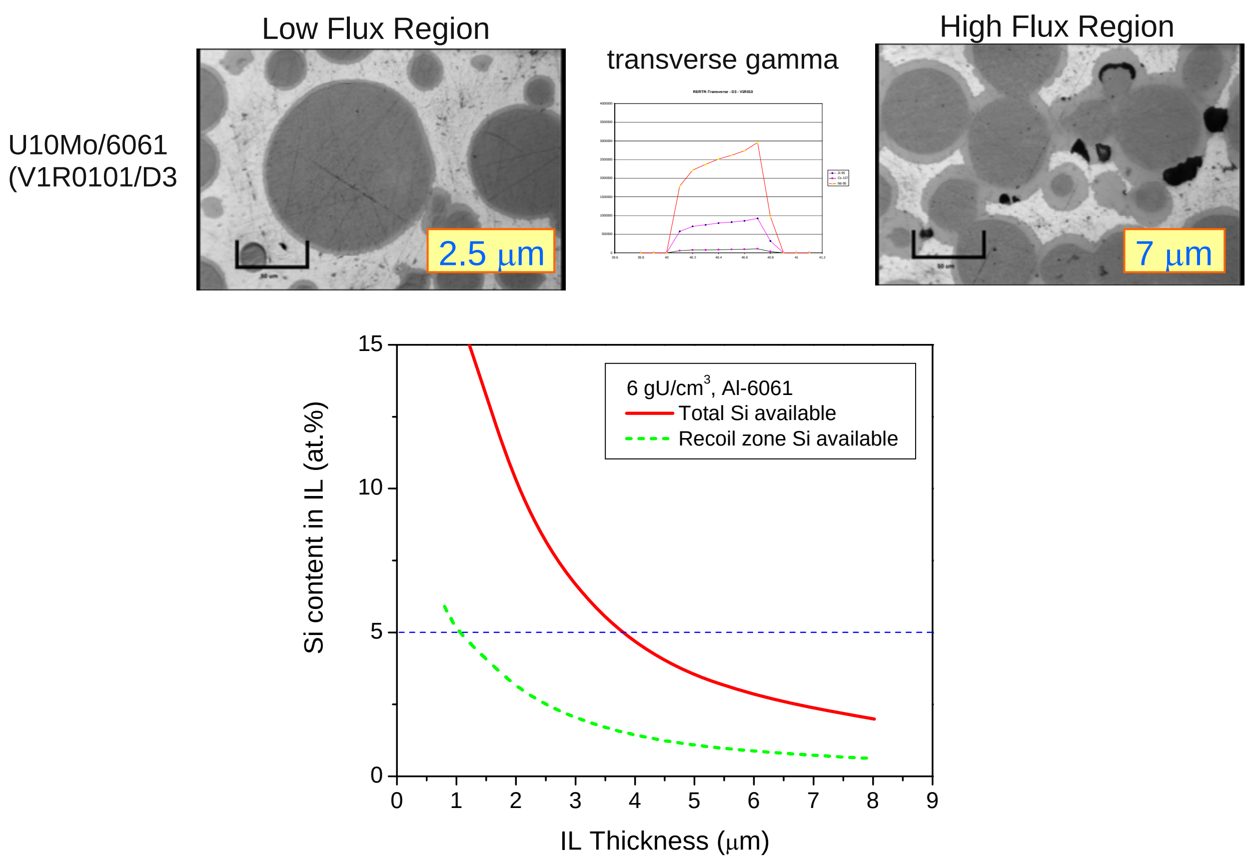


Figure 9. Estimation of Si content in the IL for two regions in miniplate V1R010. The fuel loading density is 6 gU/cm^3, the matrix is Al6061, and the average fuel particle size is 75 µm.

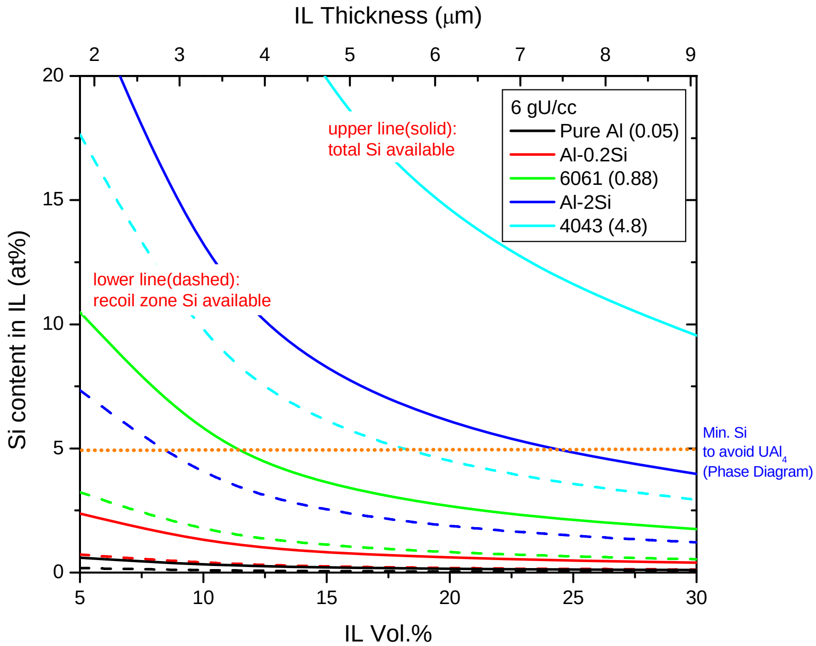


Figure 10. Amount of Si available for diffusion into U-Mo/Al interaction layer for various Si-content in Al matrix

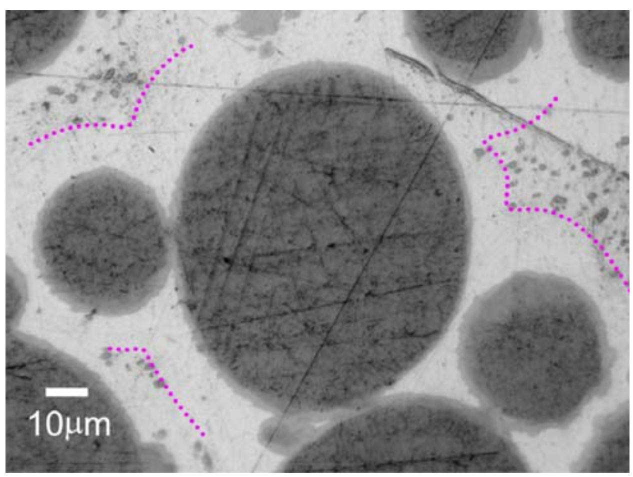


Figure 11. Distribution of remaining Si-rich precipitates in U-7Mo/4043Al (R3R030) from RERTR-6 test. The small dark particles in the bright Al matrix are the Si precipitates as in the diffusion couple above.

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