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Projection of Irradiation Behavior of U_3Si_2-Al Dispersion Fuel at High Fuel Loading and High Power

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ABSTRACT

In view of the recently renewed interest in the use of low enriched silicide dispersion fuel for the conversion of research reactors that operate at the high end of the performance range, an assessment of the projected irradiation behavior under the more taxing conditions is required. To this end, the PIE data accumulated during the U_3Si_2 irradiation programs in the ORR were re-examined as well as a few more recent results of higher power experiments performed in BR-2 and ATR. The data show that at low-enriched-uranium (LEU) burnup of 85% or higher, coarsening of fission gas bubbles occurs in all experiments examined. However, the porosity does not form interconnected gas bubbles and appears to be of acceptable magnitude.

The ranges of operating parameters of the U_3Si_2 experiments in ORR, BR-2 and ATR, whose uranium loading are 4 – 5 gU/cm^3, are 100 – 165°C for fuel temperature and 2 – 7×10^{14} fission/cm^3-s for life-average fission rate. According to the post-irradiation-examination (PIE) results, the fuel swelling and the fuel-aluminum interaction are not significantly affected by the variation of the parameters within these ranges. However, the results of tests performed in HFIR, as part of the ANS project, show that at fuel temperatures of 200°C and higher, the behavior of U_3Si_2-Al dispersion fuel is clearly different. The fuel particles do not transform to an amorphous structure but remain crystalline – having different fission gas behavior from the fuels irradiated at lower temperatures. In addition, the U_3Si_2-Al interdiffusion, being temperature dependent, is quite extensive at these higher temperatures. Therefore, if the application of U_3Si_2-Al at 200°C and higher is contemplated, an accurate and more extensive temperature evaluation is recommended.

1. Introduction

The initial qualification tests of U_3Si_2-Al dispersion fuel took place in the ORR with fuel fabricated by three commercial vendors [1]. Fuel loading ranged from 4.7 to 5.2 gU/cm^3 and the peak heat flux was 140 W/cm^2 [1]. Subsequently, CERCA fabricated fuel plates with loadings of 4.8 to 5.8 and 6.0 gU/cm^3 that were successfully irradiated in OSIRIS and SILOE [2] at also modest irradiation conditions of about 150 W/cm^2.

More recently, as part of the development of fuel for the new French reactor JHR, 4.8 gU/cm^3 plates were irradiated in BR-2 at peak heat fluxes of more than 400 W/cm^2 [3]. Current plans are to qualify fuel at similar high heat flux at somewhat higher U loading of about 5.3 gU/cm^3.

2. Silicide/Al dispersion fuel swelling

Swelling is probably the most universal problem encountered in the irradiation of nuclear fuels. The major swelling mechanism is basically the same for all types of fuel: it consists primarily of nucleation and growth of bubbles of the insoluble fission gases Xe and Kr, in addition to the accumulation of so called "solid fission products". However, there are characteristic differences in the fuel behavior. Solid fission products may be soluble in the fuel or precipitate out. They are part of the fuel density decrease, up to 100% LEU burnup, which is a linear function of the accumulated conversion of U to a large variety of fission product elements. For uranium silicide, this swelling contribution accounts to 4% volume increase for every 1% burnup of U.

The growth of, primarily, Xe bubbles at low temperature is determined by radiation enhanced diffusion, a property that depends on the fuel composition and crystal structure. As it turns out both properties change during irradiation of uranium silicide. As shown in Fig. 1, these compounds become amorphous at very low doses at low temperatures of 200 – 250ºC. Also, as shown in Fig. 2, the relative Si content increases with U burnup.

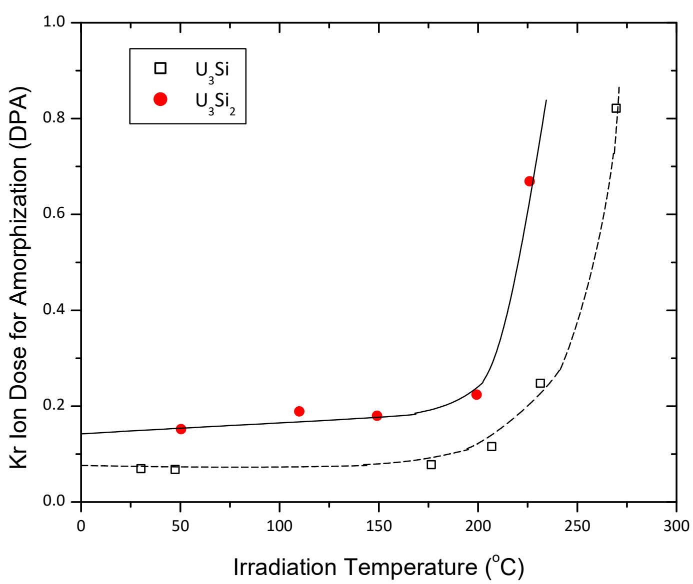

Figure 1: Temperature dependence of amorphization by ion irradiation of U-Si compounds. "U_3Si" data is from Ref. [4] and "U_3Si_2" data is from Ref. [5].

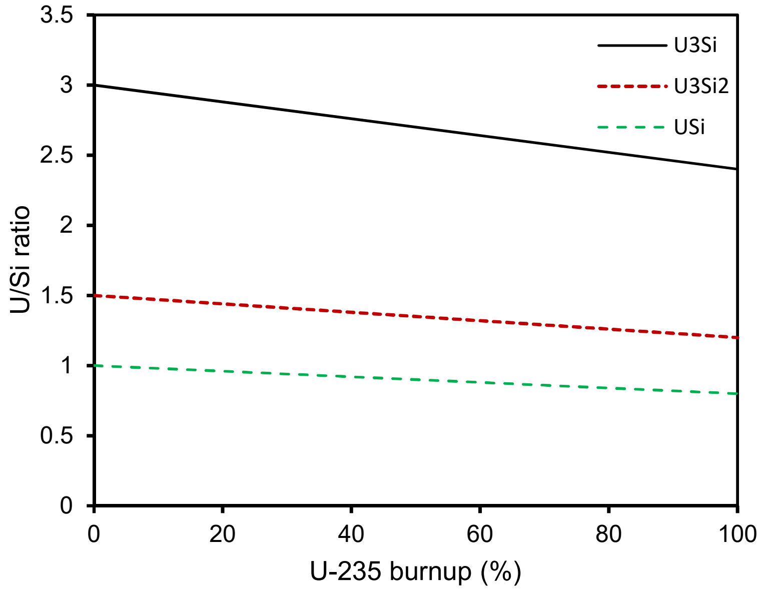

Figure 2: Change in U/Si ratio as uranium in 20% enriched uranium silicide is burned.

The effect of Si on the overall swelling of the uranium silicide is clearly evident in Fig. 3. The lowest Si composition alloys are generally swelling at a higher rate and are also approaching failure by pillowing at higher burnup, as shown in Fig. 4. The highest Si alloy maintains a low steady swelling rate, as shown in Figs. 3 and 5 for original USi, whereas the original U_3Si_2, the main purpose of this paper, shows signs of enhanced swelling behavior at exposures around 100% LEU burnup. This enhanced swelling is associated with a pronounced coarsening of fission gas bubbles, as shown in Fig. 6. When combined with a few recent experimental data on U_3Si_2-Al irradiated with highly enriched fuel, a general picture emerges. Original U_3Si_2 fuel experiences enhanced fission gas swelling by gas bubble coarsening as shown in Figs. 6 and 7 at values of maximum possible LEU burnup this appears to be well short of pillowing at this exposure. It should be pointed out that any increase in fuel loading will increase the likelihood of pillowing in high swelling plate fuel.

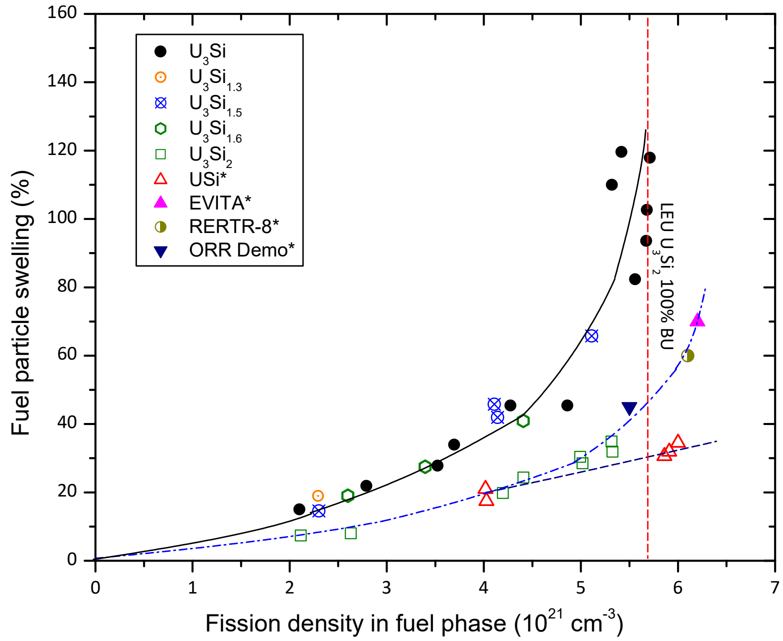

Figure 3: Fuel particle swelling in various U-Si/Al dispersion fuel plates (* Enriched in U-235 higher than 20%). "EVITA" data is from Ref. [6], "RERTR-8" data is from Ref. [7], "ORR Demo" data is from Ref. [1], and other data sets are from Ref. [8].

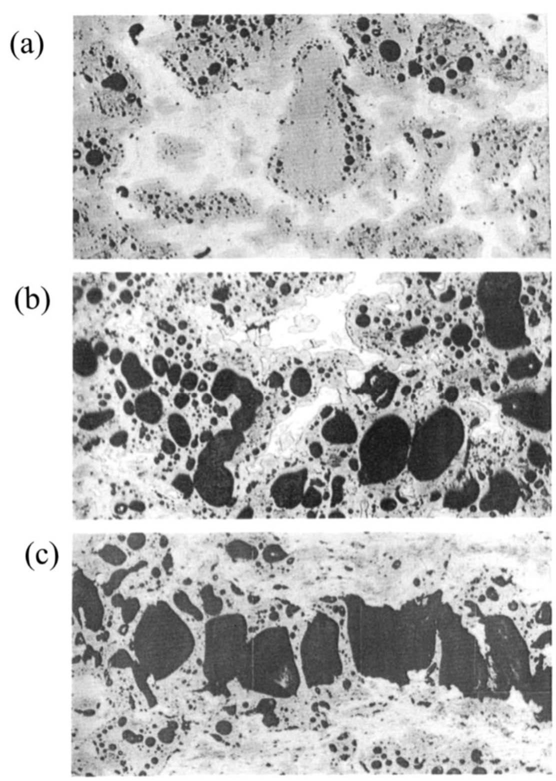

Figure 4: Fission gas bubble development in LEU U_3Si showing (a) initial swelling stage, (b) breakaway stage and (c) pillowing [9].

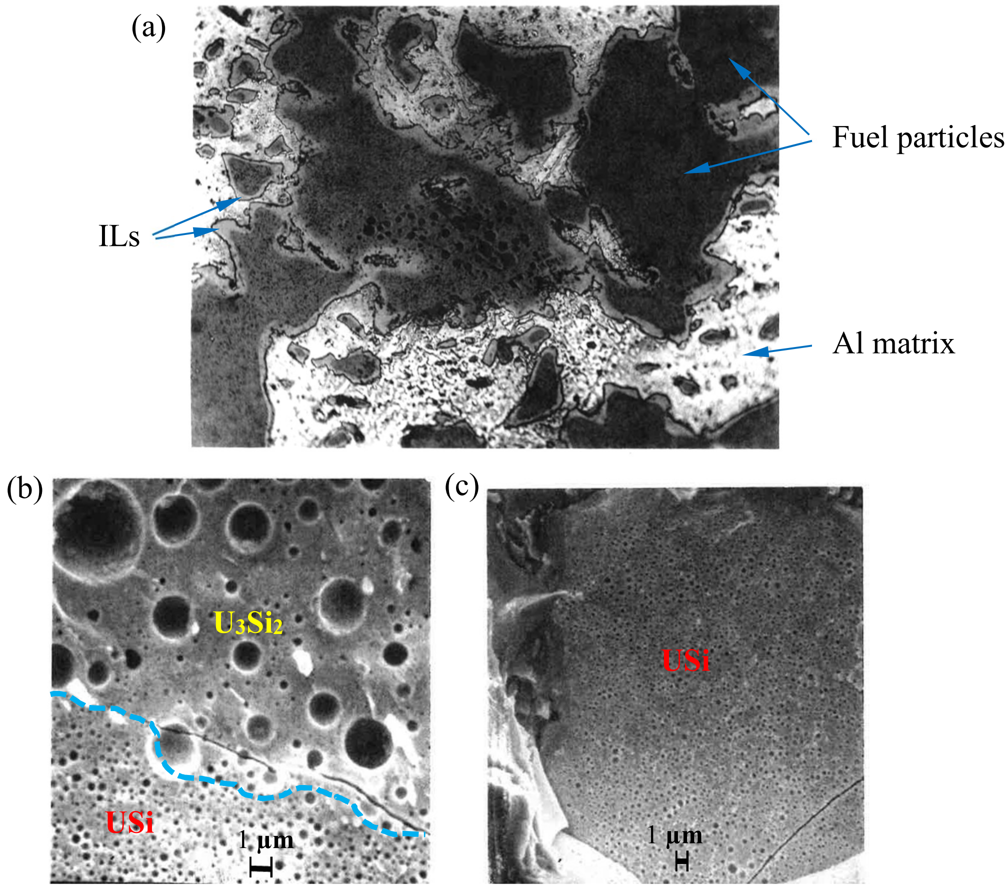

Figure 5: Micrographs of MEU USi irradiated to 5.9×10^{21} fission/cm^3 (65% LEU burnup): (a) overall microstructure, (b) microstructure of a region presumably composed of mixed U_3Si_2 and USi, and (c) general fission gas bubble morphology of irradiated USi. Note that scale bars are different in (b) and (c).

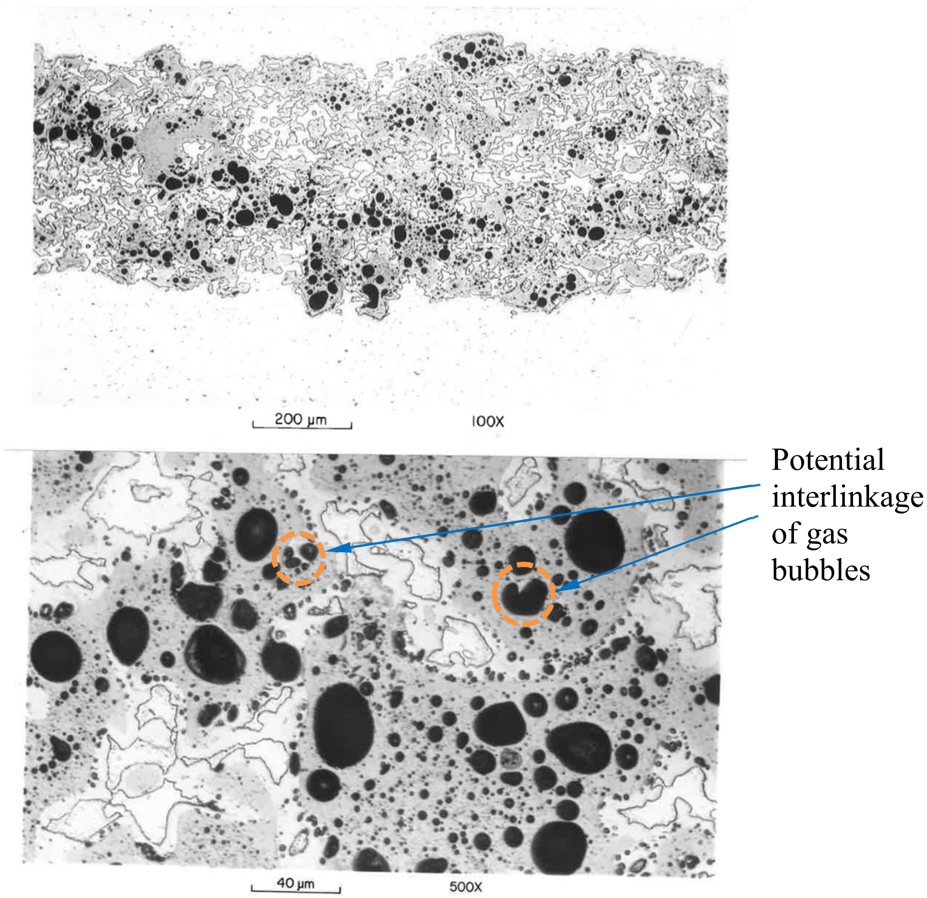

Figure 6: Microstructure of ORR Demonstration element CSI-202 at 97% burnup showing no fabrication porosity and early stage of linking of gas bubbles in some areas [1].

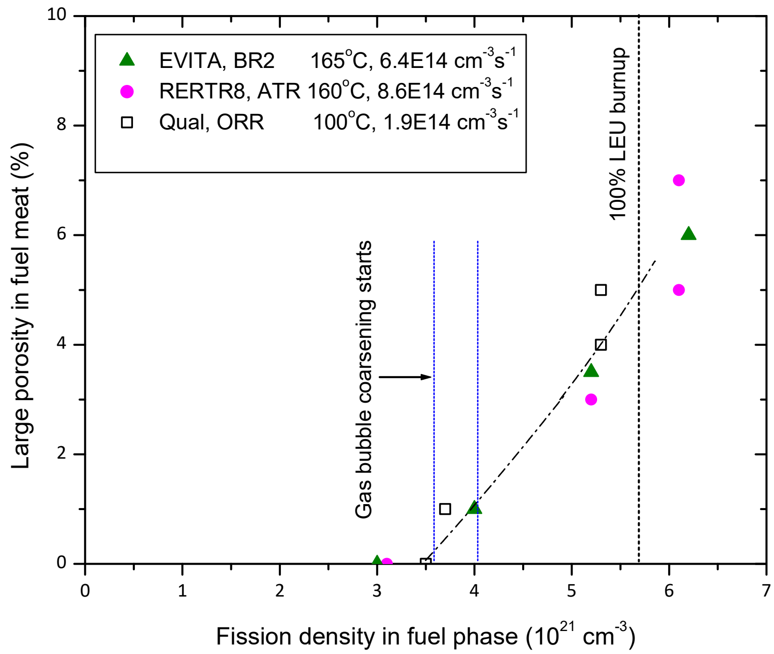

Figure 7: Coarsened fission gas bubble fraction in meat of various U_3Si_2-Al dispersion fuel plates. "EVITA" data is from Ref. [6], "RERTR8" data is from Ref. [10], and "Qual" data is from Ref. [1].

3. Effect of relatively high irradiation temperature on the behavior of U_3Si_2-Al

Clearly, at the low temperatures and moderate fission rates prevailing in most research reactors, the formation of thin interaction zones at the fuel particle surfaces has no discernable effect. However, as the HFIR tests have shown [8], the effects can be significant at higher operating temperatures and fission rates. This is shown in Fig. 8 for nominally 4.8 g/cm^3 (43 vol.% U_3Si_2) LEU dispersion samples irradiated at ~200°C, ~300°C and ~400°C, respectively. In the ~200°C sample (Fig. 8(c)), the interdiffusion zone width is substantial, but the overall fuel meat microstructure is essentially similar to that of the ORR samples (Fig. 6). At 300°C (Fig. 8(b)), however, the interdiffusion process has virtually consumed all matrix aluminum and crack-like voids have developed at the previous fuel particle surfaces. Had the samples not been constrained in the sample holders, the voids in the 400°C sample (Fig. 8(a)) should have developed to occupy approximately 50% of the meat volume; clearly any fuel plate should have pillowed under these conditions. It appears that this phenomenon is associated with the depletion (through interdiffusion) of the matrix aluminum. It is, therefore, sensitive to the amount of aluminum available, i.e., to the fuel loading as well as to the interdiffusion rate.

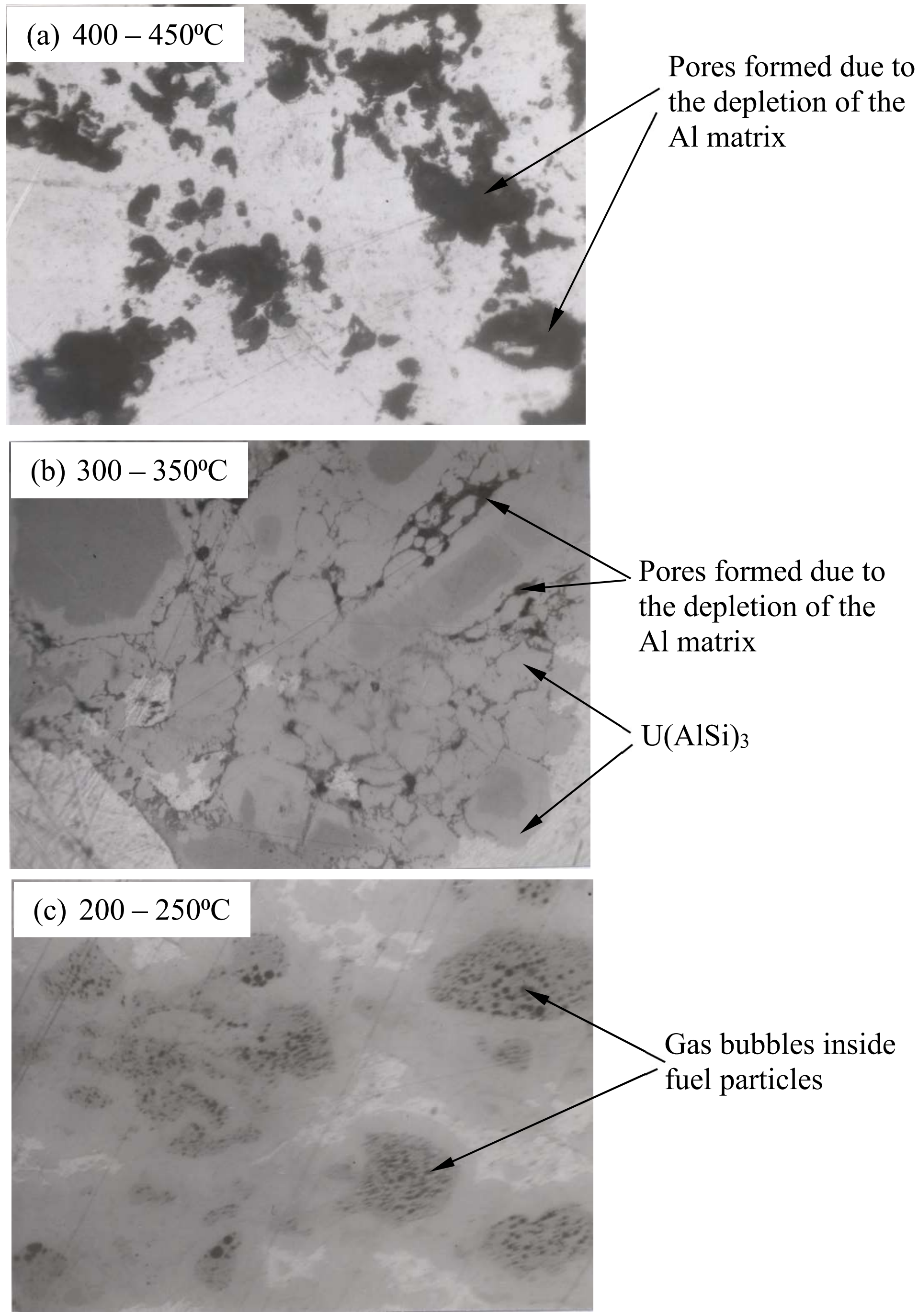

Figure 8: Low-enriched U_3Si_2, 4.8 g/cm^3 dispersion fuel samples irradiated in HFIR for one cycle of 22.9 EFPD to 74-91% U-235 depletion, at indicated fuel temperatures [8].

It is believed that the miscibility gap in the UAl_3-USi_3 tie-line shown in Fig. 9 is responsible for the formation of the crack-like voids upon depletion of the matrix aluminum. As long as an IL-Al interface exists, the outer portion of the IL will be composed of the $U(AlSi)_3$ phase. But when the Al matrix is depleted, Al continues to diffuse inward, resulting in a compound whose Al-to-Si ratio is less than 3.5. At some point, the IL composition "jumps" to the $U(SiAl)_3$ phase, which is on the other side of the miscibility gap; the consequent increase in density of the phase results in shrinkage of the IL and the formation of the crack-like voids. To avoid the formation of such cracks, one must avoid the situation where the matrix Al is depleted locally. Because of the inhomogeneous distribution of fuel particles inherent in a dispersion fuel, it is recommended that, in order to avoid the formation of the crack-like porosity in the higher-density portions of the fuel meat, the volume fraction of the matrix Al should not be allowed to drop below ~10% during irradiation.

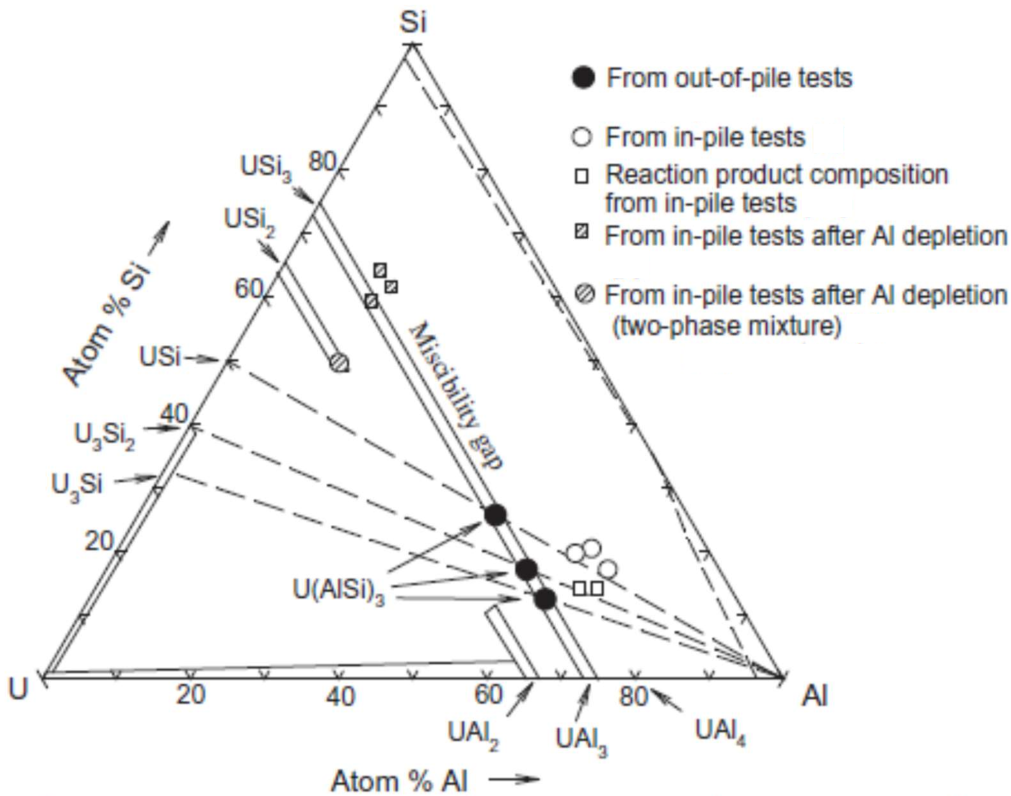

Figure 9: U-Si-Al ternary phase diagram showing compositions of reaction products between U_3Si_2 and Al [11].

Another consequence of the consumption of the matrix Al is the accompanying decrease in thermal conductivity of the fuel dispersion, which, in turn, promotes fuel temperatures to be well above beginning-of-life (BOL) temperatures. Such elevating effects on fuel temperature due to the matrix consumption are demonstrated in the U_3Si_2-Al dispersion fuel plates tested in the Belgian BR-2 reactor, exhibited in Fig. 10. The centerline fuel temperatures at the position subjected to 550 W/cm^2 is estimated to be over 400°C [12], caused by the poor heat transfer due to excessive corrosion of the cladding. Indeed, the resulting fuel microstructure (Figs. 10(d) and (f)) is remarkably similar to what was shown in the HFIR samples irradiated at higher temperatures (Figs. 8(a) and (b)). Note that there is considerable uncertainty on the fuel meat thermal conductivity of irradiated fuel. With the high heat fluxes generated in fuel meat, relatively small changes in its thermal conductivity can lead to significant variation in fuel temperature.

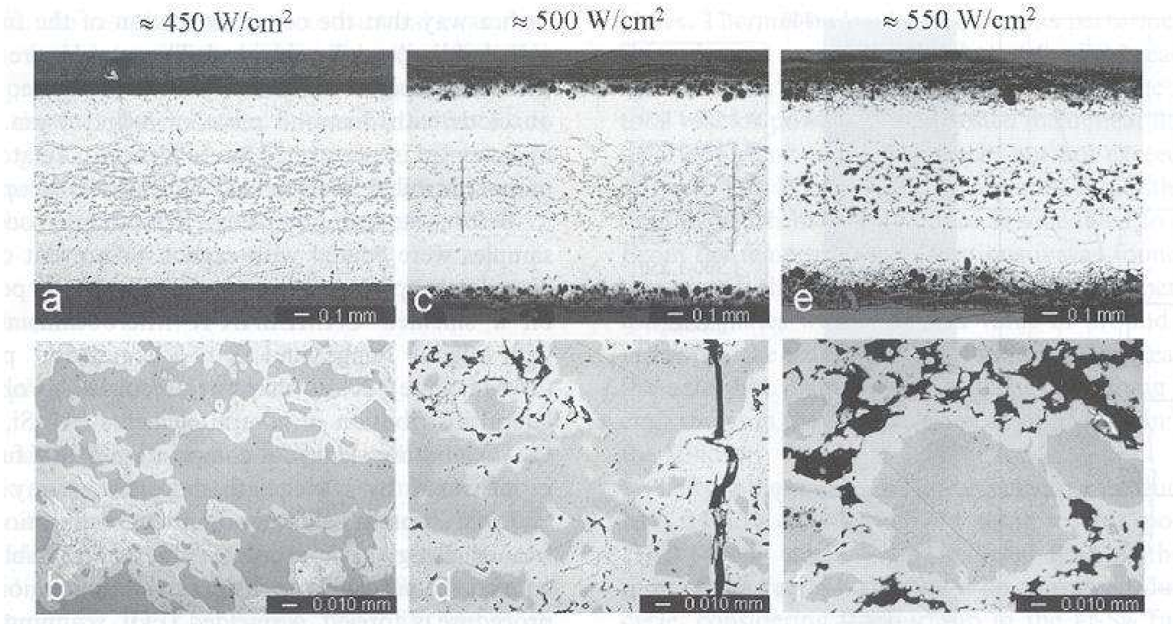

Figure 10: Images of the U_3Si_2-Al dispersion fuel showing the effect of fuel power (or temperature) [12].

4. Conclusions

Based on the available experimental information on the fabricability and irradiation performance of U_3Si_2-Al dispersion fuel, a plate-type fuel containing 6.0 gU/cm^3 of LEU [2] appears to be feasible. Main performance limits of such a fuel design are: a max burnup of ~ 90% of LEU and a max fuel meat temperature, including realistic uncertainties and variation, of 200ºC. Because original U_3Si_2 at these conditions has entered a stage of fission gas bubble coarsening, requirements on fuel homogeneity and fuel performance calculations are more important than before to ensure positive performance of this fuel design.

Acknowledgements

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