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THE EFFECTS OF FABRICATION VARIABLES ON THE IRRADIATION PERFORMANCE OF URANIUM SILICIDE DISPERSION FUEL PLATES*

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ABSTRACT

The effect of fabrication variables on the irradiation behavior of uranium silicide-aluminum dispersion fuel plates is examined. The presence of minor amounts of metallic uranium-silicon was found to have no detrimental effect, so that extensive annealing to remove this phase appears unnecessary. Uniform fuel dispersant loading, low temperature during plate rolling, and cold-worked metallurgical condition of the fuel plates all result in a higher burnup threshold for breakaway swelling in highly-loaded U_3Si fueled plates.

INTRODUCTION

At the present stage of development of low-enrichment uranium silicide dispersion fuel, where commercial fabricators have made and qualified fuel elements for use in research and test reactors, it is timely to devote attention to the effects of some fabrication variables that are dealt with in commercial production on the irradiation performance of the fuel.

The correlation between the irradiation behavior of fuels tested to date and a specific variable is not always unambiguous, because often more than one variable is changed between different irradiation test campaigns. However, the data collected thus far indicate trends in these correlations that may prove useful to the fabricators in their pursuit of superior and more economical commercial fuel.

DISCUSSION

Fuel Swelling

Before proceeding with the discussion of the topic of this paper, it will be helpful to review briefly the fuel swelling phenomena found in experimental miniplate irradiations in order to illustrate the characteristic irradiation behavior of nominally pure U_3Si and U_3Si_2.

An example of the fuel zone microstructure of a U_3Si_2 miniplate after more than 90% burnup is shown in Fig. 1. The U_3Si_2 fuel in this plate was carefully prepared in the laboratory to yield an essentially single-phase "pure" material, as contrasted to the nominal composition materials that are produced in commercial-type operations. Some of the noteworthy features in this optical micrograph are the absence of fission gas bubbles and the fact that as-fabricated porosity has been consumed by fuel particle swelling. Fuel/aluminum interaction is limited to a narrow band around the U_3Si_2 particles. SEM examination of fractured fuel particles reveals the gas bubble morphology typical of pure U_3Si_2 as shown in Fig. 2. The very uniform distribution of small gas bubbles, that show no tendency to interlink, explains the stable swelling behavior of U_3Si_2.

The microstructural changes in U_3Si miniplates resulting from irradiation to high burnups are quite different as shown in Fig. 3. Fission gas bubbles are clearly visible in the optical micrograph. The bubble morphology, more clearly shown in the SEM images in Fig. 4, reveals a basic difference in fission gas behavior between U_3Si and U_3Si_2. The fission gas bubbles in U_3Si are clearly not uniformly distributed and vary widely in size. The large bubbles are growing rapidly and linking up resulting in a much higher fuel swelling rate than that for U_3Si_2. Theoretical considerations on the difference in swelling behavior of these silicides are given in reference [1].

Fabrication Variables

Compositional variations in the as-cast commercial-type fuel alloy arise from imperfect mixing of U and Si in the melt. Melting of stoichiometric compositions of U_3Si and U_3Si_2 will in practice result in mixtures of U-Si: solid solution αU/Si (ss), U_3Si, U_3Si_2 and USi [2]. The formation of a nominal U_3Si fuel composition requires the heat-treatment of the casting to transform the αU/Si to U_3Si in the presence of U_3Si_2. To form a nominal U_3Si_2 fuel composition, however, does not require heat treating the casting because sufficient U_3Si_2 is formed on solidification. As shown in Figure 5, non-heat-treated U_3Si_2 can contain a significant quantity of αU/Si (ss). If U_3Si_2 is heat-treated, the resulting microstructure contains U_3Si and USi. The issue of heat treating with respect to irradiation revolves around the possibly detrimental behavior of the αU/Si and the U_3Si phases.

The microstructure of the as-cast and comminuted fuel is subject to changes during plate fabrication and irradiation. Both high temperature applied in plate rolling and irradiation-enhanced diffusion may cause transformation of the remaining unstable phases, such as U_3Si and αU/Si (ss) in U_3Si_2, and/or diffusion controlled reactions between these phases and the aluminum matrix. The final fuel microstructure will determine the irradiation behavior of the fuel, i.e., each phase will have its own swelling characteristics and will contribute to the overall swelling of the plate.

Some examples to illustrate this point are shown in Figs. 7 and 8. Figure 7 shows the irradiated microstructure of U_3Si_2 that was not heat-treated and contained significant amounts of αU/Si (ss) after comminuting. But the amount of U_3Si formed during rolling or irradiation was so small as to be undetectable after irradiation. Most of the αU/Si (ss) apparently transformed to $U(AlSi)_x$ by diffusion of Al from the matrix. The UAl_x compounds are extremely stable during irradiation and have never shown fission gas bubble formation [3], as appears to be the case here. This suggests that the possible presence of αU/Si at the beginning of irradiation is not a problem because it reacts with matrix aluminum to form an irradiation-resistant $U(AlSi)_x$ phase.

The U_3Si_2 fuel shown in Fig. 8, nominal U_3Si_2, was heat-treated to transform any remaining αU/Si and resulted in a mixture of U_3Si_2 and some locally large grains of U_3Si, and USi, which, from the phase diagram, has to be present as well. The USi is evidently finely dispersed and not generally recognizable in the microstructure. Both U_3Si_2 and U_3Si show their characteristic swelling behavior, i.e., large irregular bubbles form in the U_3Si while a stable uniform bubble morphology forms in the U_3Si_2. The implication of this dual behavior is obvious. If the fraction of U_3Si is so large as to form a somewhat continuous phase, the actual fuel will take on the less desirable swelling characteristics of the U_3Si.

Another important variable in high-loaded fuel plates is the non-uniform distribution of the fuel, a phenomenon associated with a problem commonly referred to in fuel plate fabrication as "dog boning." Because there is a direct correlation between loading and the onset of pillowing with fuels that exhibit breakaway swelling, such as U_3Si, an additional margin to pillowing can be achieved by avoiding local overloading (dog boning) in a fuel plate. Reference [4] deals with the effects of loading on pillowing in U_3SiAl fuel plates (this alloy, although inferior in burnup potential to U_3Si, behaves similarly). In every case where irregular distributions of fuel were present in test plates, pillowing started at the point of highest loading in the plate.

Two other noteworthy fabrication variables that are difficult to separate are rolling temperature and clad-matrix strength. There is evidence that U_3Si plates rolled at lower temperatures have a larger margin to breakaway swelling (see Fig. 9). It is believed that breakaway swelling in U_3Si is promoted by Al diffusion from the matrix. It follows, then, that lower temperatures during rolling minimize Al diffusion into U_3Si, thereby delaying breakaway swelling. The data in Fig. 9 could also be explained by the harder cold-rolled plates providing additional mechanical restraint on the swelling fuel particles. With either explanation reduced temperatures during plate rolling appear to be beneficial.

CONCLUSIONS

It can be concluded that careful fabrication procedures that result in uniform fuel distribution and composition, and mechanically strong cladding and matrix material play roles in providing the margin to breakaway swelling in highly loaded plates containing potentially unstable fuel (predominately U_3Si). Lower rolling temperatures may result in a delay of breakaway swelling in fuel afflicted by the phenomenon.

Heat-treatment of non-homogeneous U_3Si_2 to remove metallic αU/Si may indeed be counter productive in reducing swelling because αU/Si transforms to $U(AlSi)_x$ during rolling or irradiation, which is advantageous. The deliberate use of a non-heat-treated mixture of αU/Si and U_3Si_2 to achieve high loading therefore appears promising. The volume change on the transformation to the aluminide is insignificant compared to the volume change of swelling gas bubbles.

REFERENCES

1. Gerard L. Hofman, "Crystal Structure Stability and Fission Gas Swelling in Intermetallic Uranium Compounds," Journal of Nuclear Material 140 (1986), pp. 256-263.

2. R. F. Domagala, this conf.

3. Gerard L. Hofman, "Fission Gas Bubbles in Uranium Aluminide Fuels," to be published in Nuclear Technology April 1987.

4. G. L. Hofman and L. A. Neimark, "Postirradiation Analysis of Experimental Uranium-Silicide Dispersion Fuel Plates," Proc. Int. Meeting on Reduced Enrichment for Research and Test Reactors, Argonne, Illinois, USA, Oct. 1984.

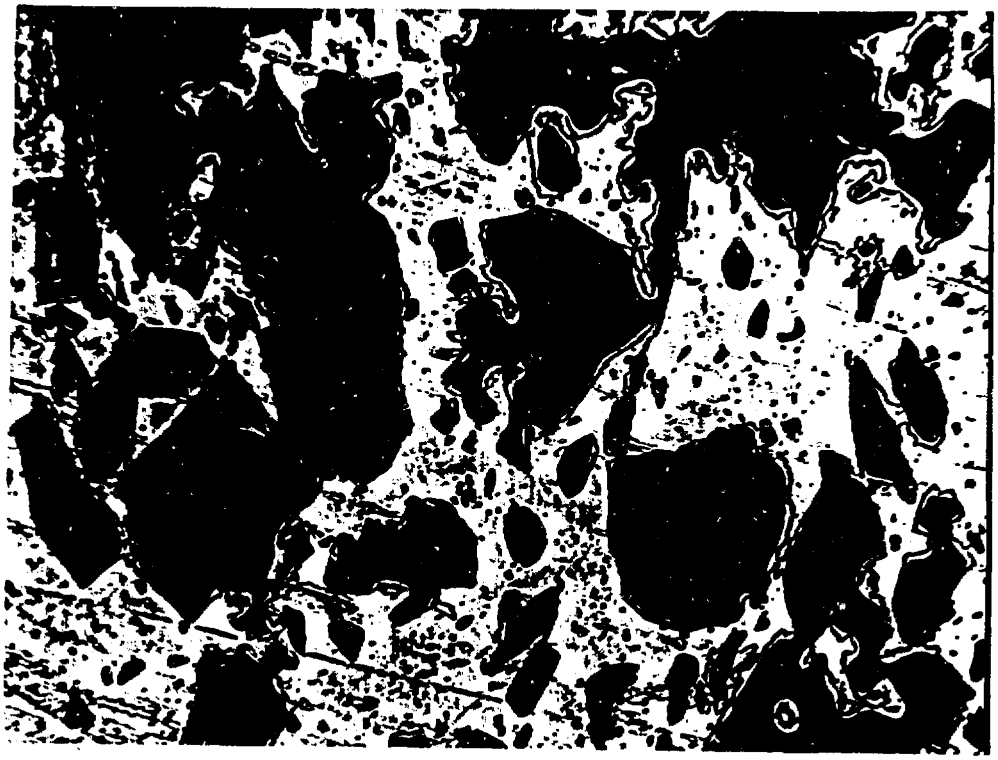

Fig. 1. Fuel Zone Microstructure of U_3Si_2 (250X) Miniplate After 90% Burnup (Optical).

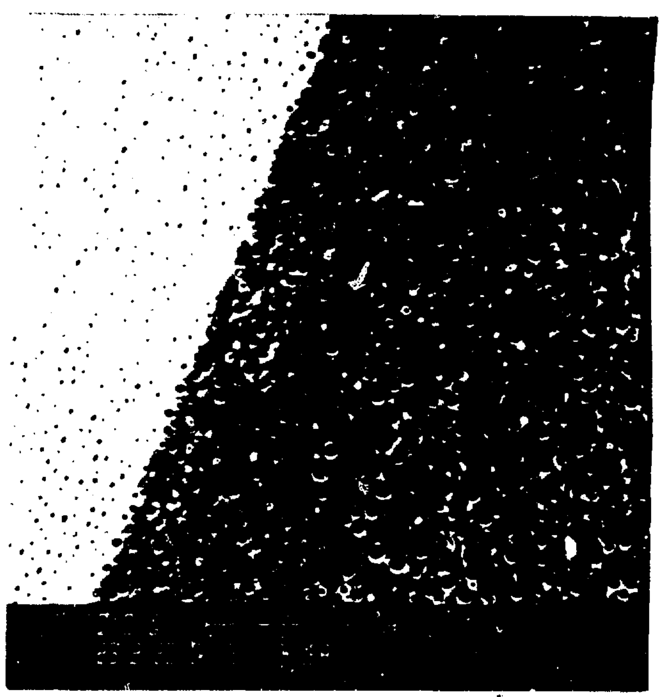

Fig. 2. Fission Gas Bubble Morphology in U_3Si_2 After 90% Burnup (SEM).

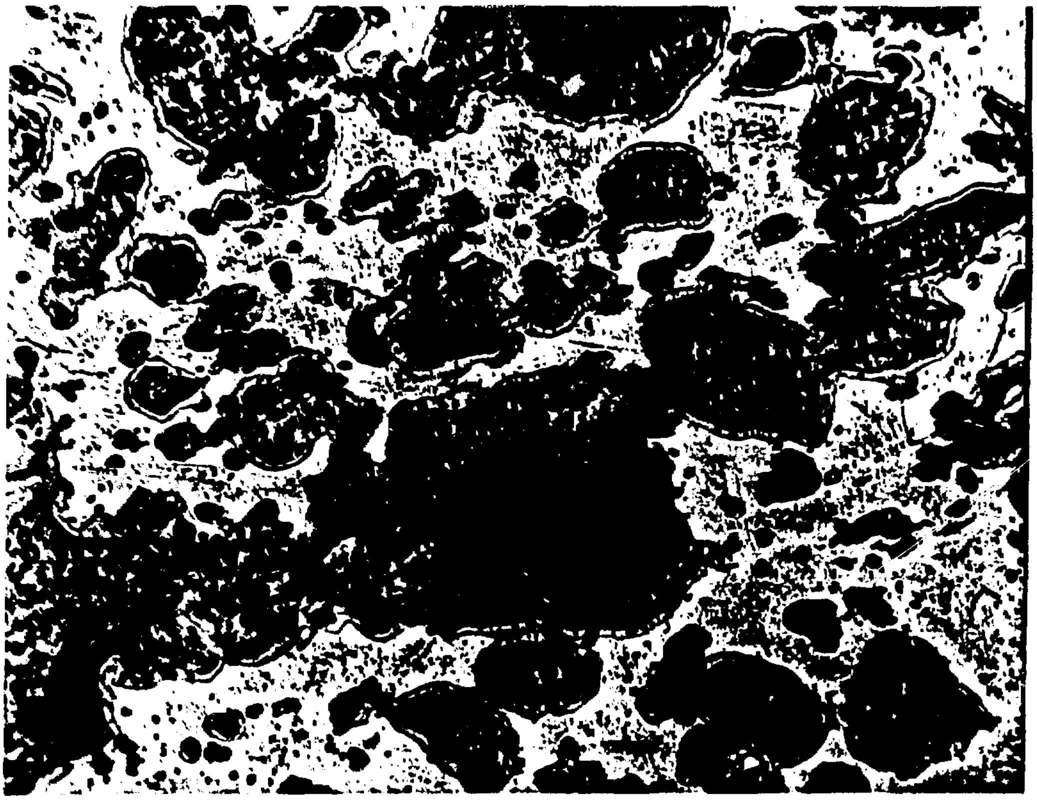

Fig. 3. Fuel Zone Microstructure of U_3Si (250X) Miniplate After 90% Burnup (Optical).

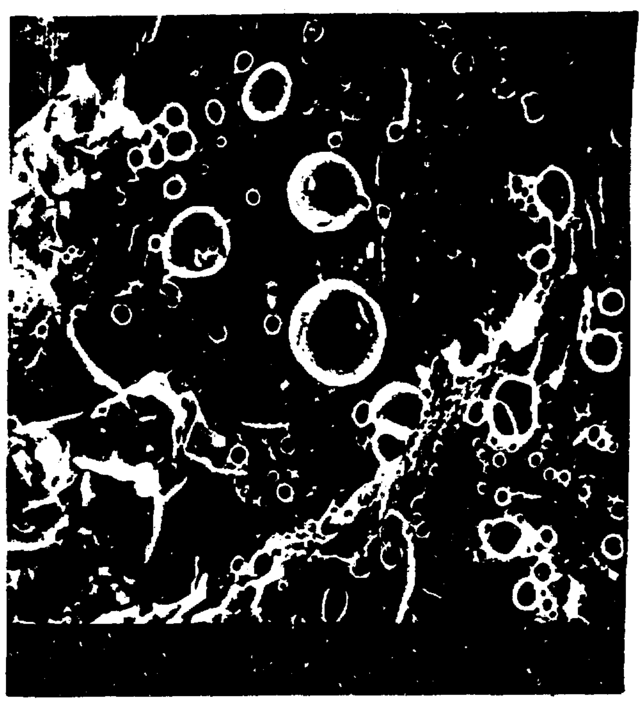

Fig. 4. Fission Gas Bubble Morphology in U_3Si After 90% Burnup (SEM).

Fig. 5. U_3Si_2 Particle Containing Lacy αU/Si (ss) Phase. (Fuel Not Heat-Treated Before Comminuting and Plate Fabrication.)

Fig. 6. U_3Si_2 Particles Containing U_3Si (White Phase). (Fuel Heat-Treated Before Comminuting and Plate Fabrication.)

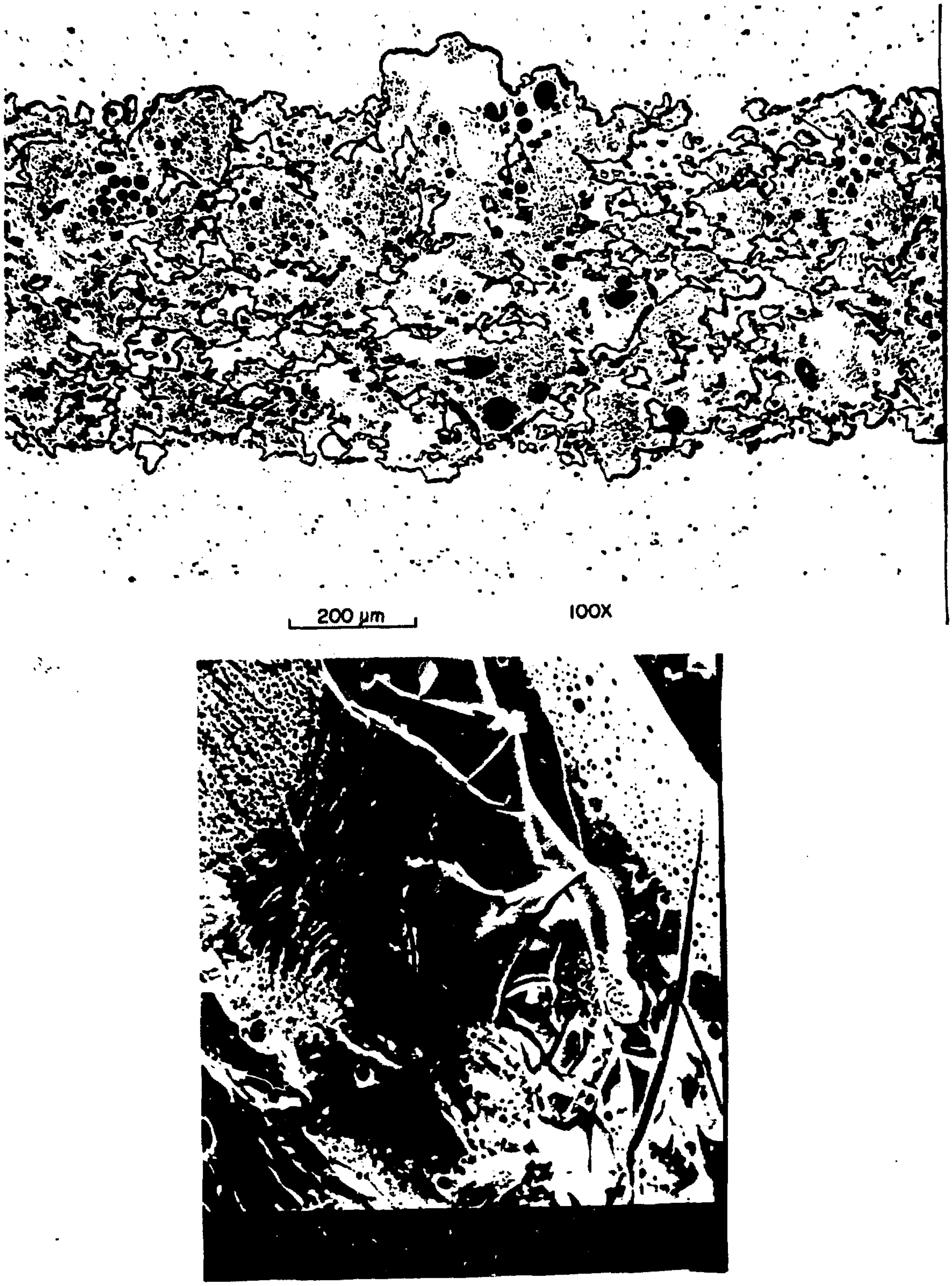

Fig. 7. Microstructure and Bubble Morphology in U_3Si_2 (Not Heat Treated) Irradiated to >90% Burnup. Optical and SEM Images, Showing Bubble Free $U(AlSi)_x$ (In Both Optical and SEM Images), Small Bubbles in U_3Si_2, and Large Bubbles in U_3Si.

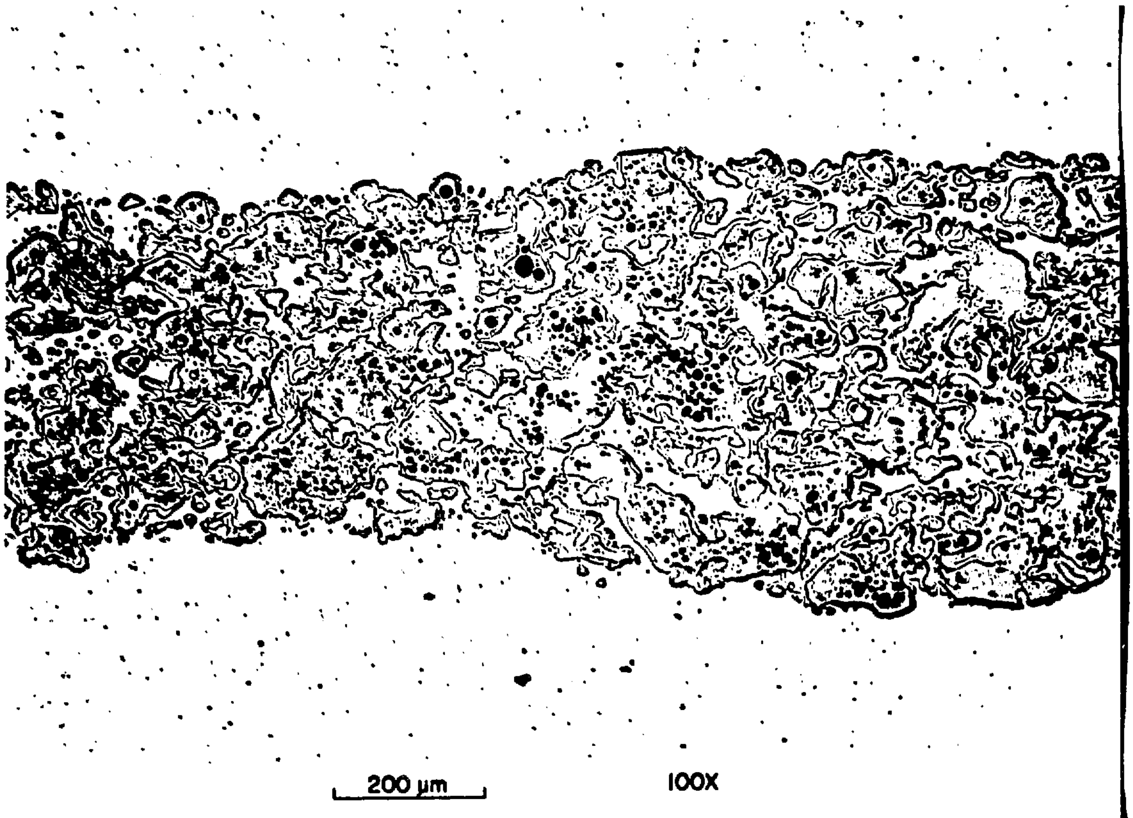

Fig. 8. Microstructure and Bubble Morphology of Heat-Treated U_3Si_2 Irradiated to >90% Burnup, Showing Large Bubbles in Particles Containing U_3Si.

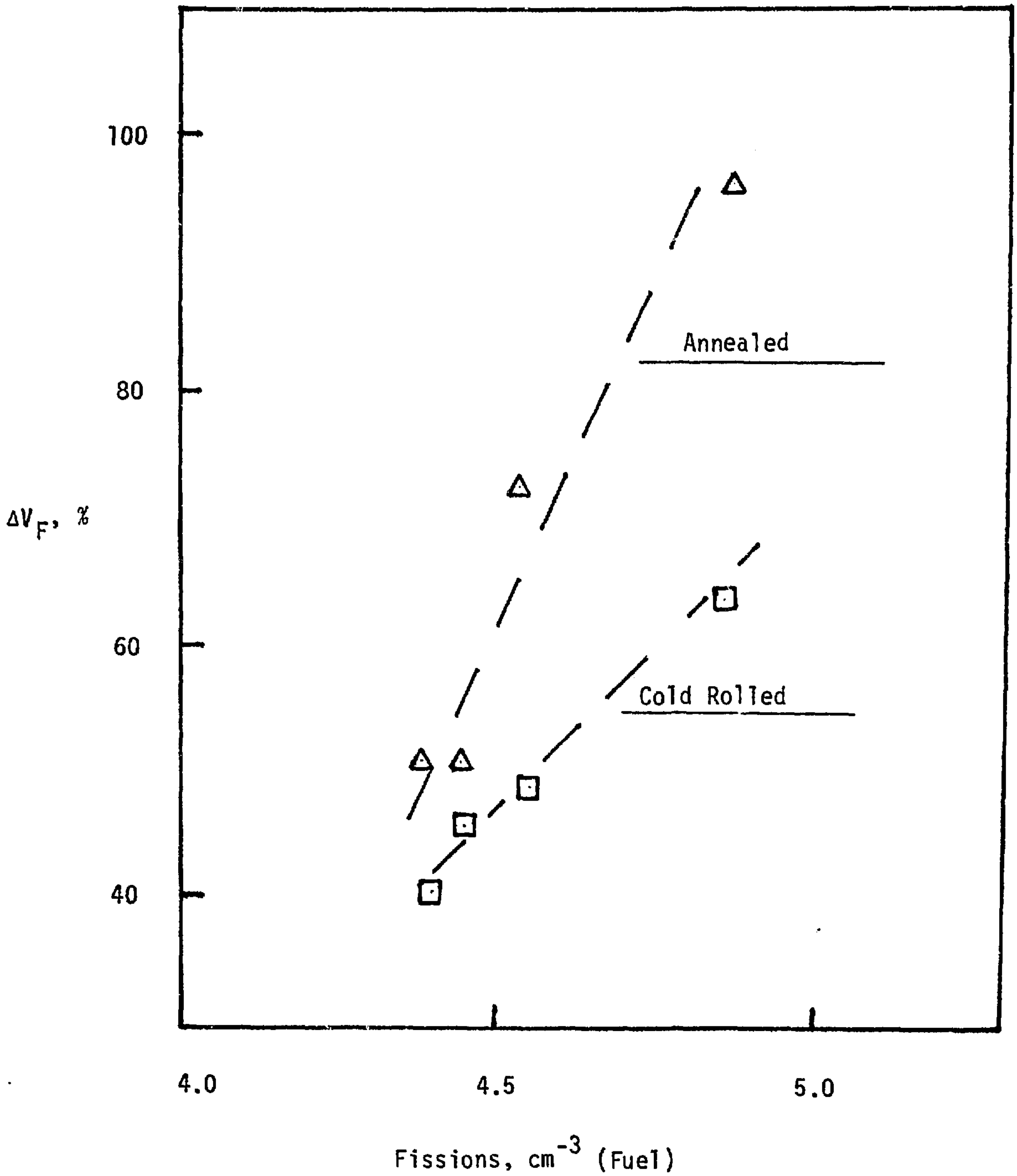

Fig. 9. Fuel Swelling in Central Region of U_3SiAl Mini-Plates Loaded With 6.2 g cm^{-3} of Uranium Showing Effect of Annealing Treatment.