

POSTIRRADIATION ANALYSIS OF EXPERIMENTAL  
URANIUM-SILICIDE DISPERSION FUEL PLATES

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Abstract

Low-enriched uranium silicide dispersion fuel plates were irradiated to maximum burnups of 96% of  $^{235}\text{U}$ . Fuel plates containing 33 v/o  $\text{U}_3\text{Si}$  and  $\text{U}_3\text{Si}_2$  behaved very well up to this burnup. Plates containing 33 v/o  $\text{U}_3\text{Si-Al}$  pillowed between 90 and 96% burnup of the fissile atoms. More highly loaded  $\text{U}_3\text{Si-Al}$  plates, up to 50 v/o were found to pillow at lower burnups. Plates containing 40 v/o  $\text{U}_3\text{Si}$  showed an increase swelling rate around 85% burnup.

INTRODUCTION

Irradiation in the Oak Ridge Research Reactor of all miniature, low-enriched uranium silicide fuel plates in the initial test matrix of the RERTR fuel development program was concluded last year. The three fuel types included in the matrix,  $\text{U}_3\text{Si}$ ,  $\text{U}_3\text{Si-Al}$ , and  $\text{U}_3\text{Si}_2$ , all attained maximum burnups of 96% of the original 20%  $^{235}\text{U}$ . Postirradiation examinations of selected fuel plates are essentially completed and the results are reported here. Some miniplate parameters needed to follow the discussion of the irradiation behavior are given in Table 1. More detailed information on the fuel and the

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TABLE 1. Parameters for Miniplates from the First Irradiation Campaign of the RERTR Fuel Development Program

Fuel Type	Fuel Loading		Average Meat Porosity, %	Peak <sup>235</sup> U Bu, %	Peak Fission Density in Fuel Meat $10^{21} \text{ cm}^{-3}$	Number of Plates
	$\text{gU cm}^{-3}$	Average v/o				
U <sub>3</sub> Si	4.8	33	6	35	0.7	5
U <sub>3</sub> Si	4.8	33	6	90	2.2	4
U <sub>3</sub> Si	4.8	33	6	96	2.4	2
U <sub>3</sub> Si	5.8	40	17	73	1.7	4
U <sub>3</sub> Si <sub>2</sub>	3.7	34	5	90	1.6	4
U <sub>3</sub> Si <sub>2</sub>	3.7	34	5	96	1.8	2
U <sub>3</sub> SiAl	4.7	33	6	35	0.7	7
U <sub>3</sub> SiAl	4.7	33	6	90	2.2	4
U <sub>3</sub> SiAl	4.7	33	6	96	2.4	2
U <sub>3</sub> SiAl	5.7	40	17	73	1.7	4
U <sub>3</sub> SiAl	6.4	45	19	73	1.9	6
U <sub>3</sub> SiAl	7.0	50	19	75*	2.2	6

\*Preliminary values.

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fabrication of the miniplates has been reported by Domagala<sup>1</sup>, Dwight<sup>2</sup>, and Nazare.<sup>3</sup> Inspection of Table 1 reveals an emphasis on U<sub>3</sub>Si-Al (the intermetallic compound U<sub>3</sub>Si alloyed until 1.5 w/o Al, as it was initially seen as the prime candidate for low-enriched fuel. Experience gained in the Canadian reactor program with this particular variety of U<sub>3</sub>Si and the paucity of irradiation data on pure U<sub>3</sub>Si and U<sub>3</sub>Si<sub>2</sub> lead to the preference for U<sub>3</sub>Si-Al. Unfortunately, in our current application and geometry, as the following discussion will show, this fuel proved to be the inferior of the three. Conclusions on the irradiation behavior of these miniplates put forth in previous reports<sup>4,5</sup> are not fundamentally altered by the additional data accumulated since 1982. However, the new data on highly loaded plates now indicate an effect of volumetric loading on the U<sub>3</sub>SiAl and U<sub>3</sub>Si swelling rate at high burnup, and on the onset of pillowing in U<sub>3</sub>SiAl.

## IRRADIATION BEHAVIOR

### Miniplate Swelling

Irradiation swelling of the miniplates is shown in Fig. 1 in the conventional terms of volume change,  $\Delta V$ , of the meat versus the accumulated number of fissions in the meat, expressed in  $f/cm^3$ . The swelling in U<sub>3</sub>Si<sub>2</sub> and U<sub>3</sub>Si at a loading of 33-34% continues to be moderate up to the very high burnup value of 96%, i.e.,  $1.8 \times 10^{21}$  and  $2.4 \times 10^{21}$   $f/cm^3$  in U<sub>3</sub>Si<sub>2</sub> and U<sub>3</sub>Si, respectively. The ternary alloy U<sub>3</sub>Si-Al at the same loading had, as previously reported,<sup>4</sup> shown signs of breakaway swelling at 90% burnup ( $2.2 \times 10^{21}$   $f/cm^3$ ) and was found to have pillowed at 96% burnup ( $2.4 \times 10^{21}$   $f/cm^3$ ).<sup>4</sup> This instability and failure by pillowing has now also occurred in U<sub>3</sub>SiAl miniplates with 45 and 50 v/o fuel loadings at burnups of 75% ( $2.2 \times 10^{21}$   $f/cm^3$ ).

The data plotted in Fig. 1 indicate a general trend of breakaway swelling in the plates at about  $2.2 \times 10^{21}$  f/cm<sup>3</sup> in the meat. With the exception of one cluster of data points for 40 v/o plates at  $2 \times 10^{21}$  f/cm<sup>3</sup>, there appears to be no correlation with fuel loading (and its attendant porosity variation; see Table 1) or fuel material. It would appear, however, that when swelling and pillowing occur, the magnitude is greater in more highly loaded plates.

When the data are plotted as fuel volume increase versus fuel burnup, in f/cm<sup>3</sup>, a different picture of the effect of fuel loading appears. Figure 2 presents the data in this manner for U<sub>3</sub>SiAl miniplates, and Fig. 3 presents the data for U<sub>3</sub>Si and U<sub>3</sub>Si<sub>2</sub>. In generating these figures, it has been assumed that the meat porosity has been totally consumed in accommodating fuel particle swelling; this assumption is corroborated by the fuel microstructures shown in subsequent figures. When plotted in this manner, the data in Figs. 2 and 3 show that the threshold fuel burnup for breakaway swelling, followed by pillowing, is higher in the plates with lower loadings. Interestingly, the data for the 45 and 50 v/o U<sub>3</sub>SiAl plates (Fig. 2) cluster at a fuel burnup of  $\sim 5 \times 10^{21}$  f/cm<sup>3</sup>, with the 50 v/o plates generally showing the higher swelling at this burnup. The 40 v/o U<sub>3</sub>SiAl plates fall along the same swelling vs burnup curve, but in a lower swelling range. This suggests that the 40 v/o fuel may actually be following a swelling curve somewhere between those of the 33 v/o and 45-50 v/o plates. Unfortunately, there are no additional 40 v/o plates being irradiated to higher burnup to substantiate this hypothesis.

The swelling of U<sub>3</sub>Si in 33 and 40 v/o plates, as shown in Fig. 3, appears to be similar to that of U<sub>3</sub>SiAl, at least up to  $\sim 5.7 \times 10^{21}$  f/cm<sup>3</sup> in the fuel particles. The U<sub>3</sub>Si data also show a lower burnup threshold for breakaway swelling with increasing fuel loading. In fact, these data when overlaid on the U<sub>3</sub>Si-Al data would tend to support the hypothesis that the 40 v/o U<sub>3</sub>Si-Al

swelling curve is different from the 45-50 v/o curve.

Even with the paucity of data points,  $U_3Si_2$  shows lower swelling than  $U_3Si$  and  $U_3Si-Al$  at a loading of 33 v/o.

It may be important to note here that the swelling data pertain to the entire miniplate and thus reflect the average swelling for each plate. Plate thickness measurements show that the plates with lower loadings did swell very uniformly and the total swelling measurements are therefore representative of all the fuel in each plate. However, the highly loaded plates, particularly those with 50 v/o fuel, were found to have rather severe "dog-boning". Plate thickness measurements indicate that the swelling rate is higher in the dog-bone areas than in the more homogeneous areas of the plate. The effect of non-uniform meat thickness on breakaway swelling and pillowing is under study.

#### Microstructure

Miniplate and fuel microstructures were determined by optical and scanning electron microscopy to complement the observations made on miniplate swelling.

The overall microstructure of transverse sections through the meat of the three different fuels at the same loading of 33 v/o and burnup of 96% ( $7.3 \times 10^{21}$  f/cm<sup>3</sup> of fuel in  $U_3Si$  and  $U_3SiAl$  and  $5.3 \times 10^{21}$  f/cm<sup>3</sup> of fuel in  $U_3Si_2$ ) is shown in Fig. 4. The comparison illustrates the greatly different behavior of the fuels under the same irradiation conditions and almost the same burnup. The  $U_3Si-Al$  fuel is in an advanced state of breakaway swelling. Fuel particles have formed a continuous mass and lost their identity because of the link-up of large fission gas bubbles across this fuel mass. The linkage of large bubbles caused this fuel plate to fail by pillowing.

Swelling in the  $U_3Si$  fuel has also caused linking up of the fuel particles

to a large extent. Fission gas bubble growth is, however, much less extensive and appears to occur predominantly towards the perimeter of the fuel particles. These two observations have been linked to the presence of Al in the fuel, as described previously.<sup>4</sup>

In sharp contrast to both other fuels, the  $U_3Si_2$  fuel (Fig. 4, bottom) shows no signs of fission gas bubble formation at this magnification. The fuel swelling has eliminated the as-fabricated porosity and caused some fuel particles to link up. But on the whole, the meat morphology has changed minimally during the entire irradiation.

The microstructure of the high-burnup  $U_3Si_2$  is shown in higher magnification in Fig. 5, and the scanning electron micrograph shown in Fig. 6 reveals the fission gas bubbles in a typical  $U_3Si_2$  fuel particle. As is the case at a somewhat lower burnup (85%) the fission gas bubbles are small and uniformly distributed.<sup>4</sup>

The microstructure of the more highly loaded plates tends to explain the swelling data. For example, Fig. 7 shows a section of meat of a 50 v/o  $U_3SiAl$  plate with a calculated fuel swelling of 100% (see Fig. 2). The fuel in this plate is in a state of breakaway swelling just prior to pillowing. The fuel particles form a continuous network and large fission gas bubbles are beginning to link up across this network. In fact, the fuel ligaments between bubbles are already being stretched in the transverse direction. In more highly loaded plates, fuel particle linkage through fuel swelling will occur at a lower fission density than for plates with lower loadings because the particles are in closer proximity. The microstructures also show an increase in the bubble growth rate in the more highly loaded plates. This is most likely due to the effect of higher temperatures in these denser dispersions, which would increase fission gas mobility. The combined effect of earlier

interlinkage of fuel particles and the earlier onset of breakaway swelling due to enhanced bubble growth will lead to earlier bubble linkage across particles and thereby to earlier pillowing.

The effect of loading in  $U_3Si$  plates is not as clear, owing perhaps to the fact that no plates with over 40 v/o fuel loading were tested. However, as Fig. 3 indicates, fuel swelling in 40 v/o  $U_3Si$  plates is accelerated compared with that of 33 v/o  $U_3Si$  plates at high fission densities. The microstructures shown in Fig. 8 for a 40 v/o  $U_3Si$  plate are consistent with this swelling rate increase. Fission gas bubbles in a number of fuel particles appear to be in the breakaway stage as a result of higher fuel temperatures in the more highly loaded dispersion. Significantly, though, many fuel particles retain a more stable bubble morphology. Another illustration of this behavior is given in Figs. 9 and 10. The  $U_3Si$  fuel particle depicted in Fig. 9 is entering the breakaway swelling stage, while a neighboring particle shown in Fig. 10 exhibits the more common  $U_3Si$  bubble morphology of large bubbles at its periphery and very small bubbles in the center of the particle. The non-homogeneous bubble growth in  $U_3Si$  has been attributed to aluminum diffusion into the particles.<sup>4</sup>

#### CONCLUSIONS

1. Fuel swelling in 33 v/o  $U_3Si_2$  plates continues to be moderate and the fission gas bubble morphology is very stable at a burnup of 96% ( $^{235}U$ )  $2.4 \times 10^{21}$  f/cm<sup>3</sup> of meat).
2. Higher fuel loadings have the effect of lowering the fuel burnup at which breakaway swelling and pillowing occur in  $U_3SiAl$ . This is likely due to an increase in fuel temperature that occurs when the fuel loading is increased.

3. There are indications that this temperature effect is at work in  $U_3Si$  as well. However, because of compositional inhomogeneities in this fuel, bubble growth is not as extensive and the swelling is not as rapid as it is in  $U_3SiAl$ . No pillowing has occurred in 40 v/o  $U_3Si$  plates up to 73% burnup.

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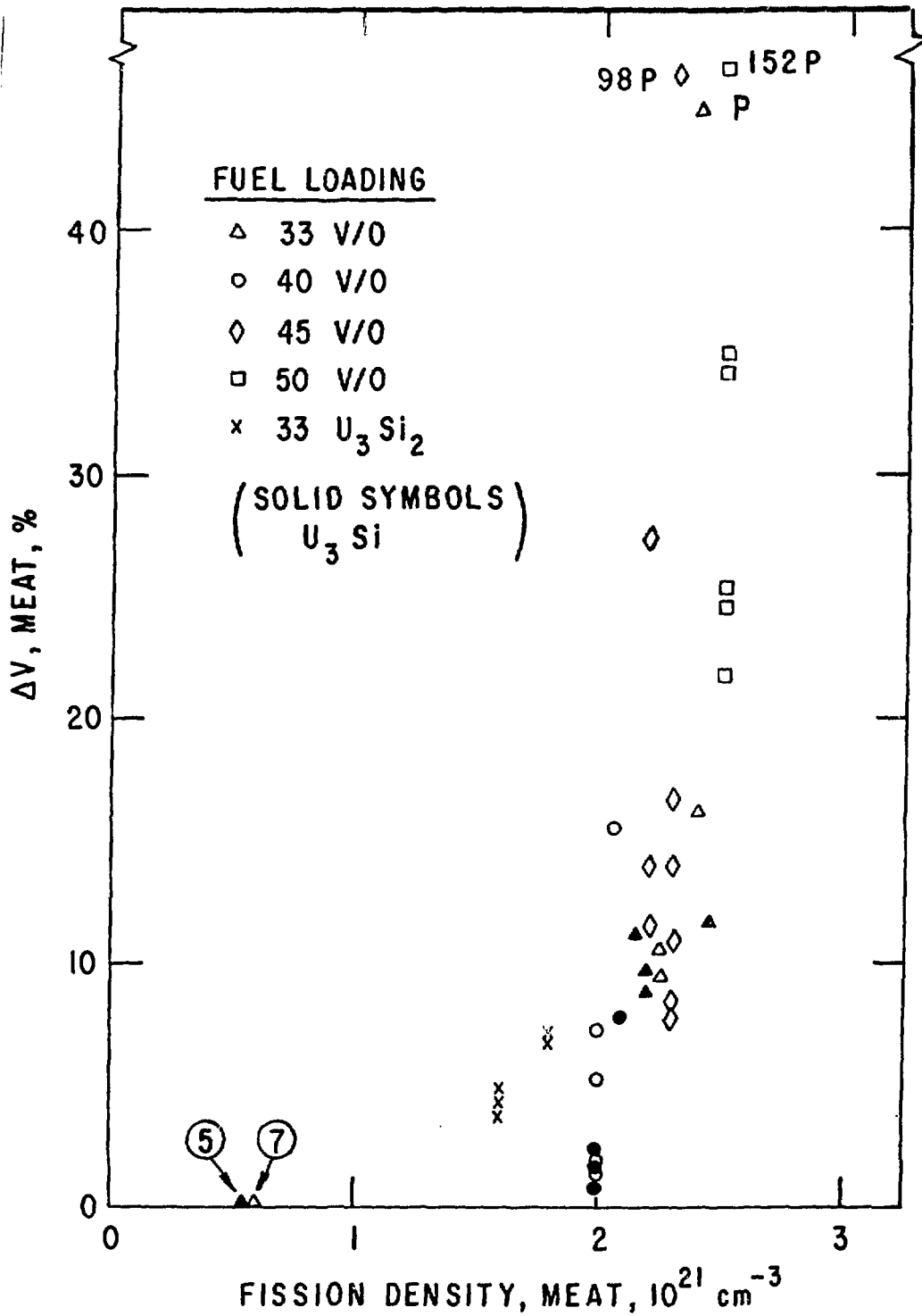


Fig. 1. Fuel Plate Meat Swelling in Miniplates with Various Fuel Loadings, as a Function of Meat Fission Density. P indicates pillowed plates.

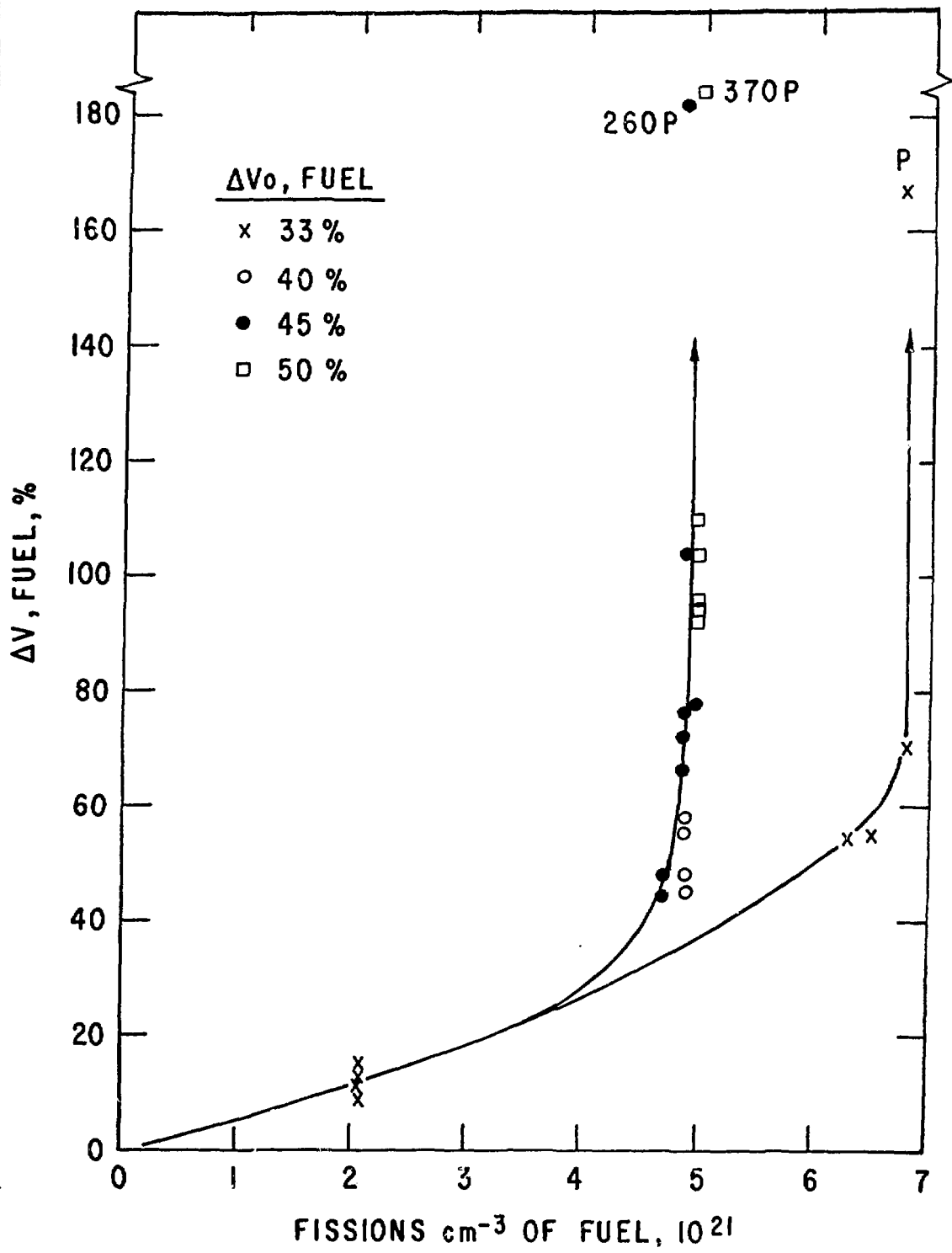


Fig. 2. Fuel Particle Swelling in  $\text{U}_3\text{Si-Al}$  Miniplates with Various Fuel Loadings, as a Function of Fission Density in the Fuel Particles Only.

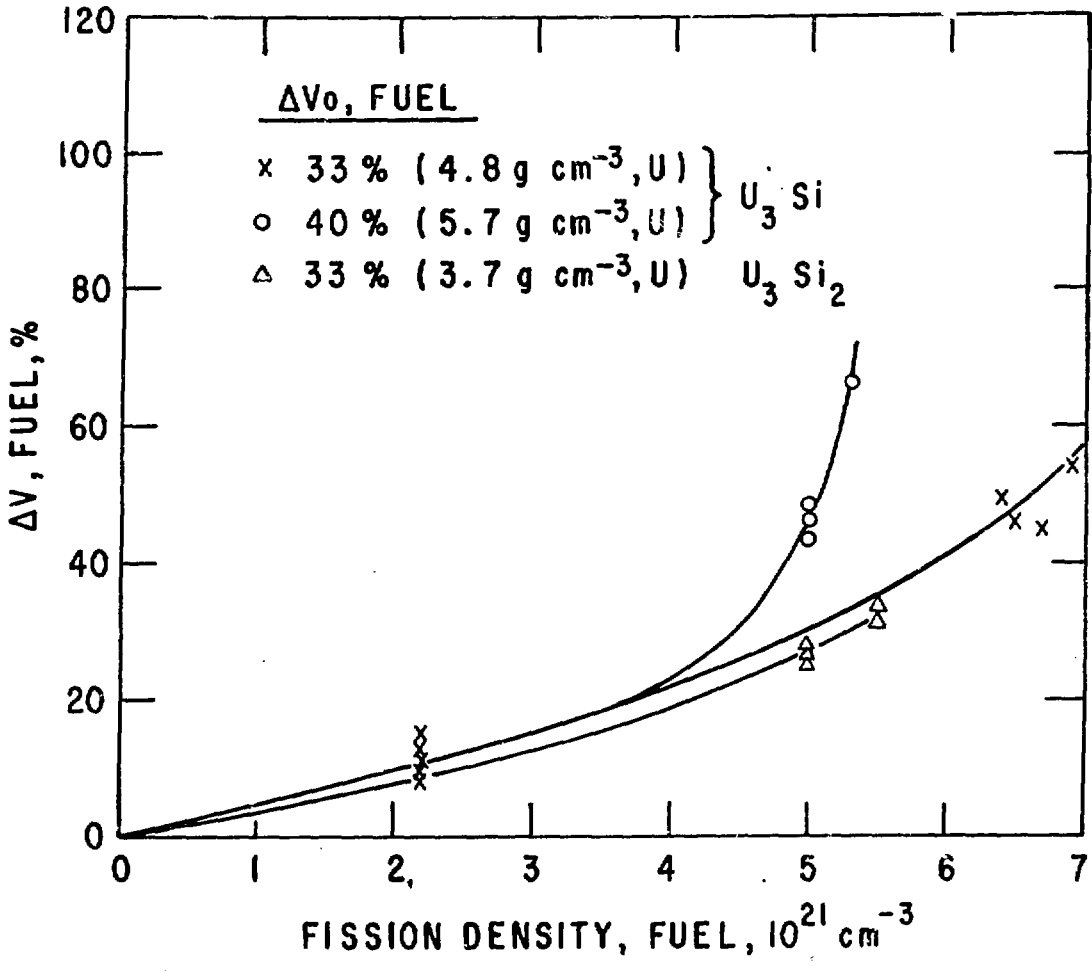


Fig. 3. Fuel Particle Swelling in U<sub>3</sub>Si and U<sub>3</sub>Si<sub>2</sub> Miniplates with Various Fuel Loadings, as a Function of Fission Density in the Fuel Particles Only

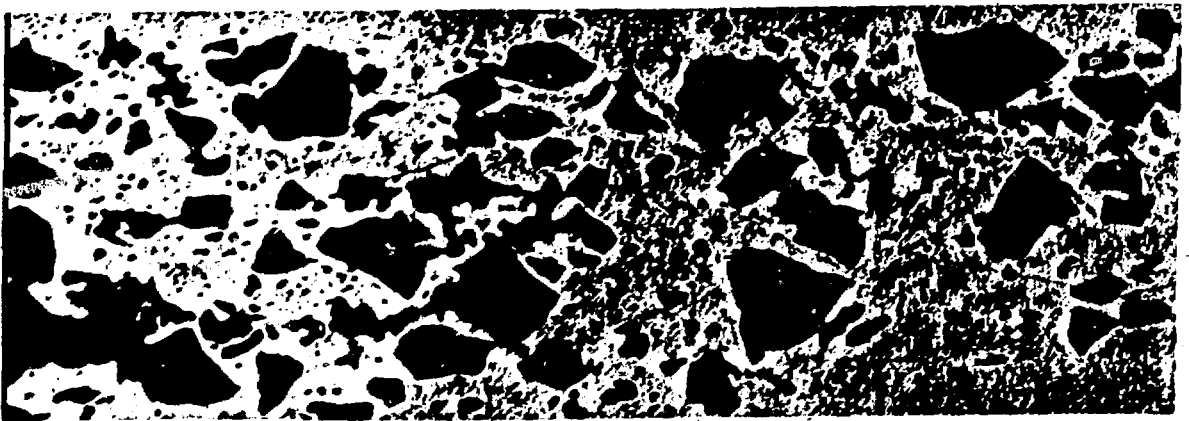
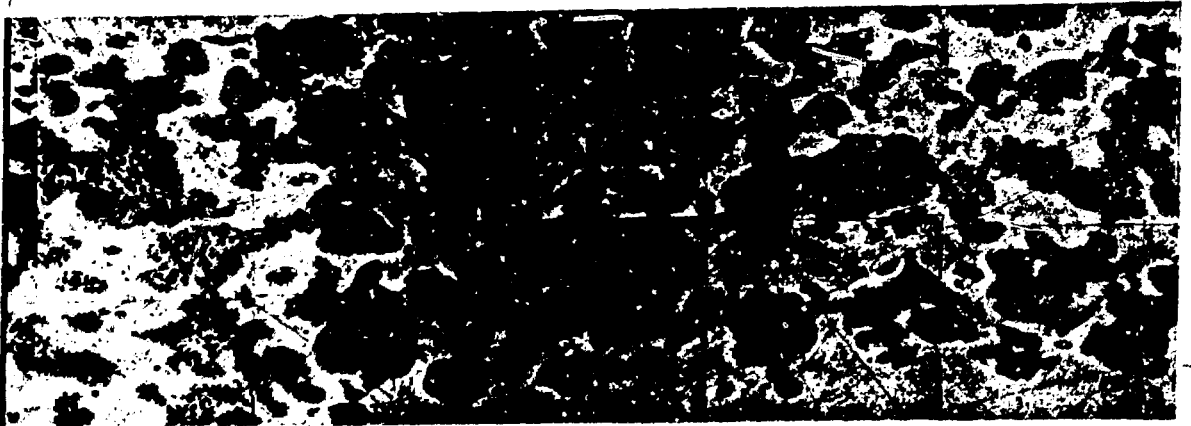
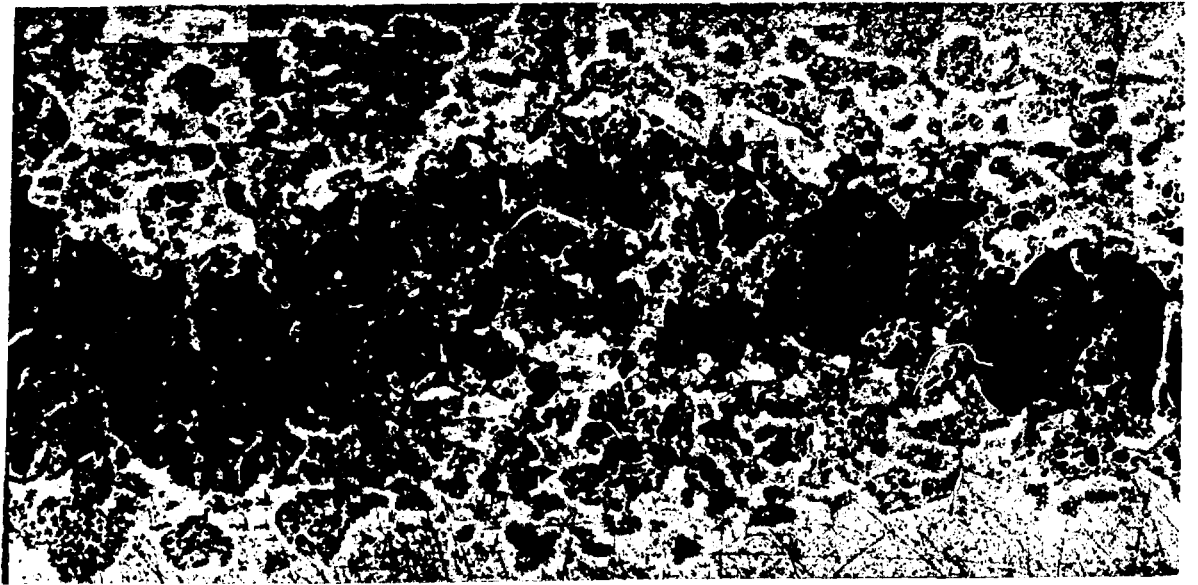


Fig. 4. Meat Microstructure of (Top to Bottom) As-polished  $U_3Si-Al$ ,  $U_3Si$ , and  $U_3Si_2$  after 96%  $^{235}U$  Burnup.

— ( 50 $\mu$  )

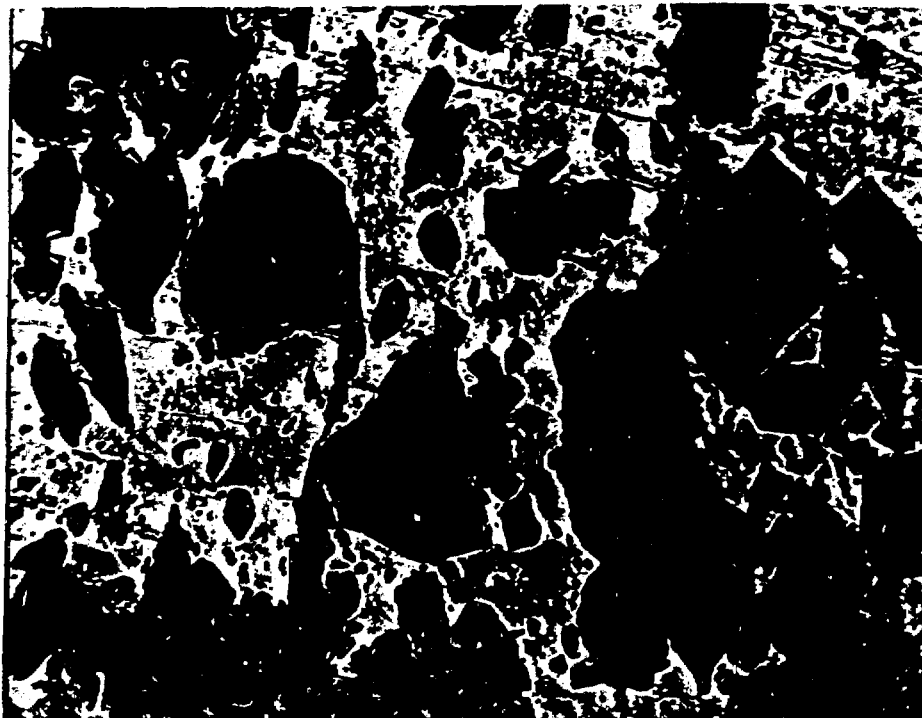


Fig. 5. Meat Microstructure of As-polished  $U_3Si_2$  after 96%  $^{235}U$  Burnup.

10 $\mu$

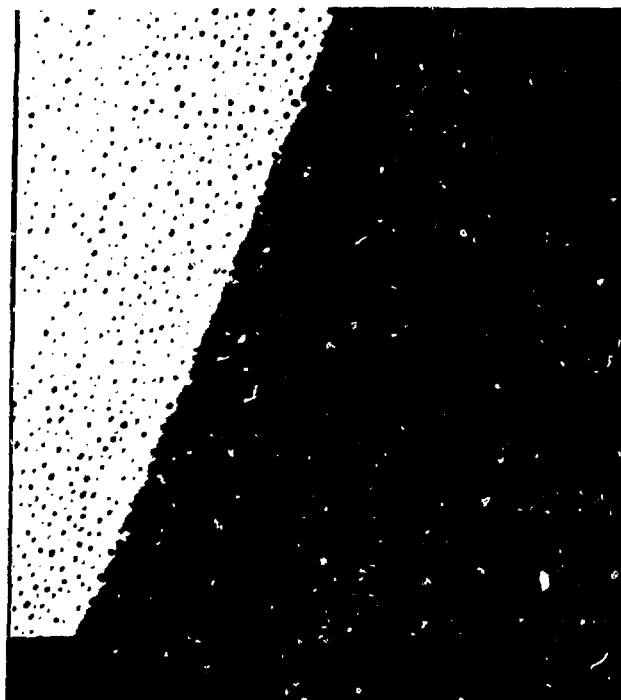
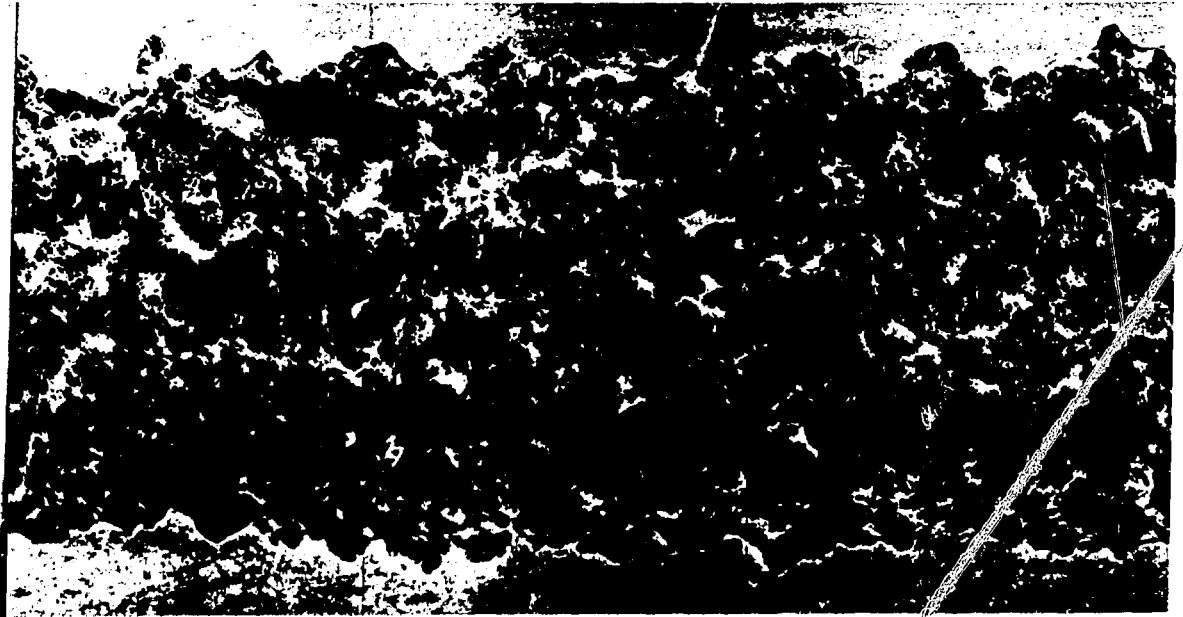
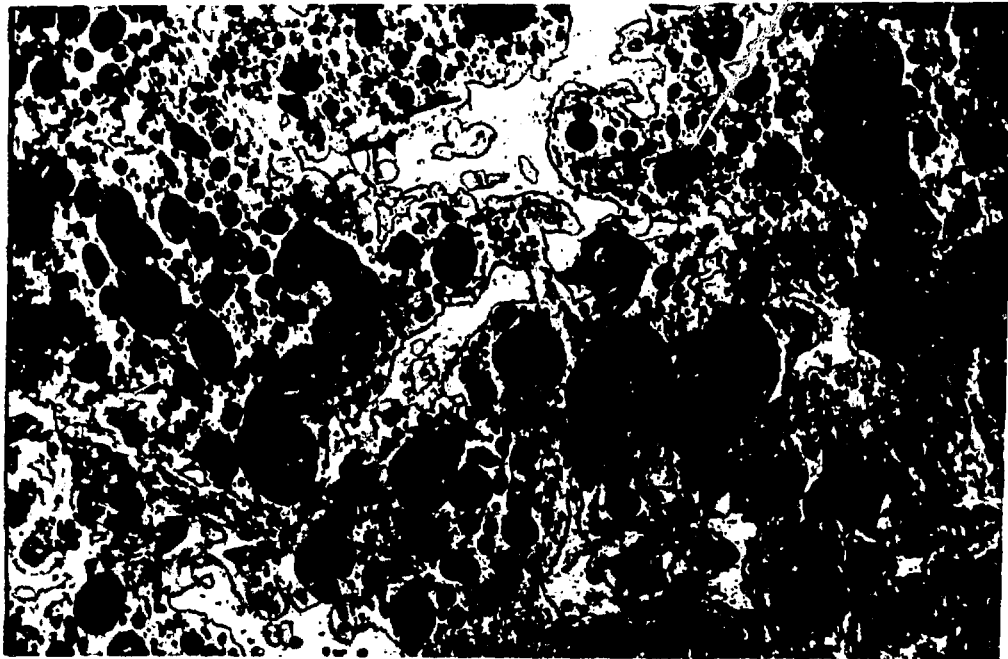


Fig. 6. Scanning Electron Micrograph Showing Bubble Morphology in  $U_3Si_2$  after 96%  $^{235}U$  Burnup.

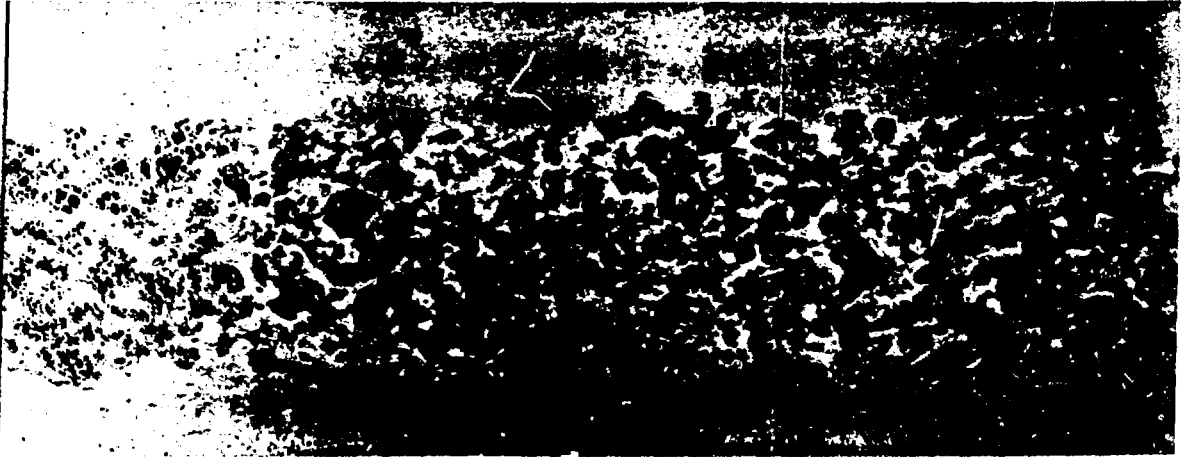


100 $\mu$

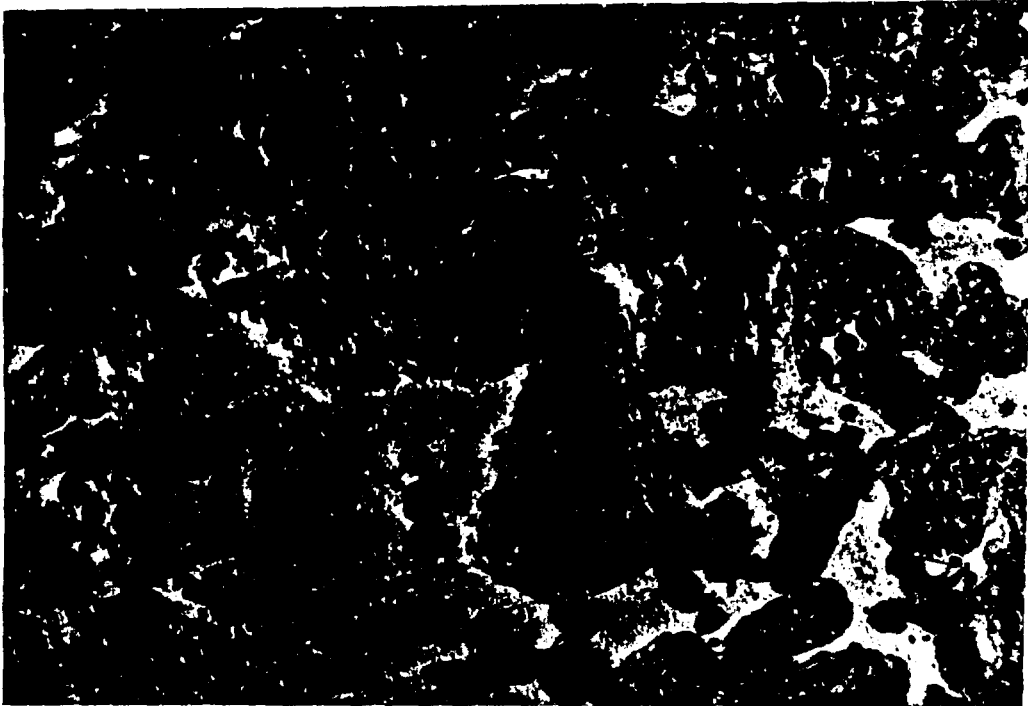


10 $\mu$

Fig. 7. Meat Microstructures As-polished 50 v/o  $U_3Si-Al$  after 75%  $^{235}U$  Burnup.



— (100 $\mu$ )



— (10 $\mu$ )

Fig. 8. Meat Microstructures of As-polished 40 v/o  $U_3Si$  after 74%  $^{235}U$  Burnup.



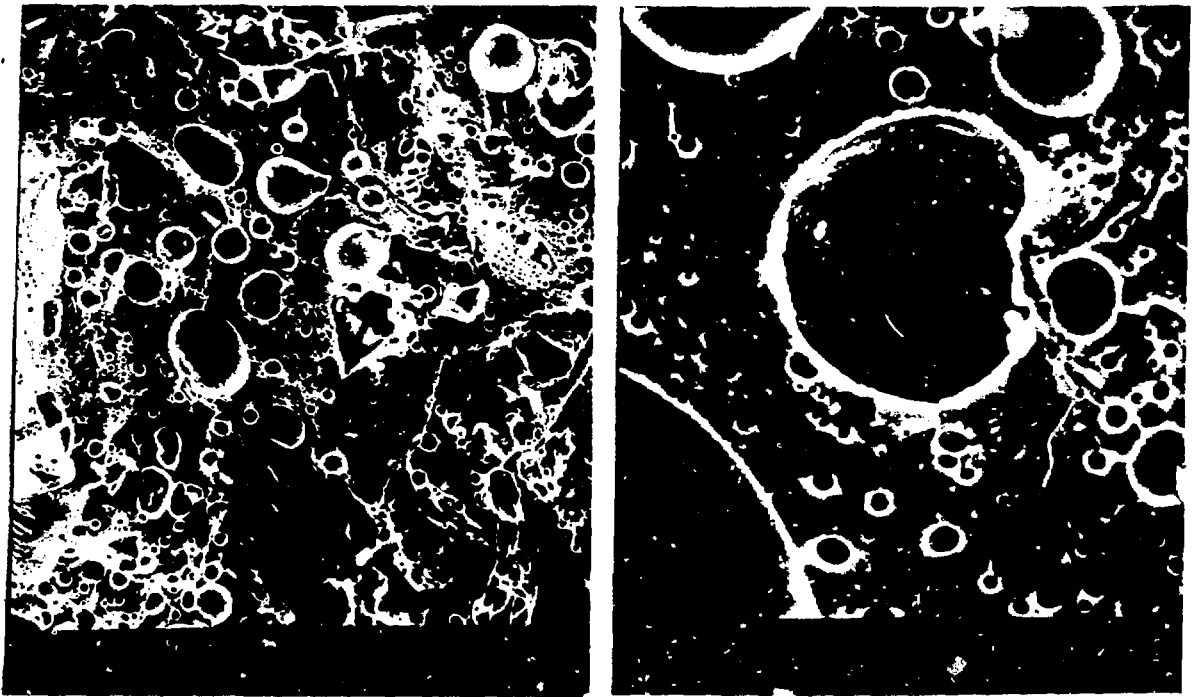


Fig. 9. Scanning Electron Micrographs Showing Breakaway Fission Gas Bubble Growth in  $U_3Si$  Particle in 40 v/o Loaded Plate after 75%  $^{235}U$  Burnup.

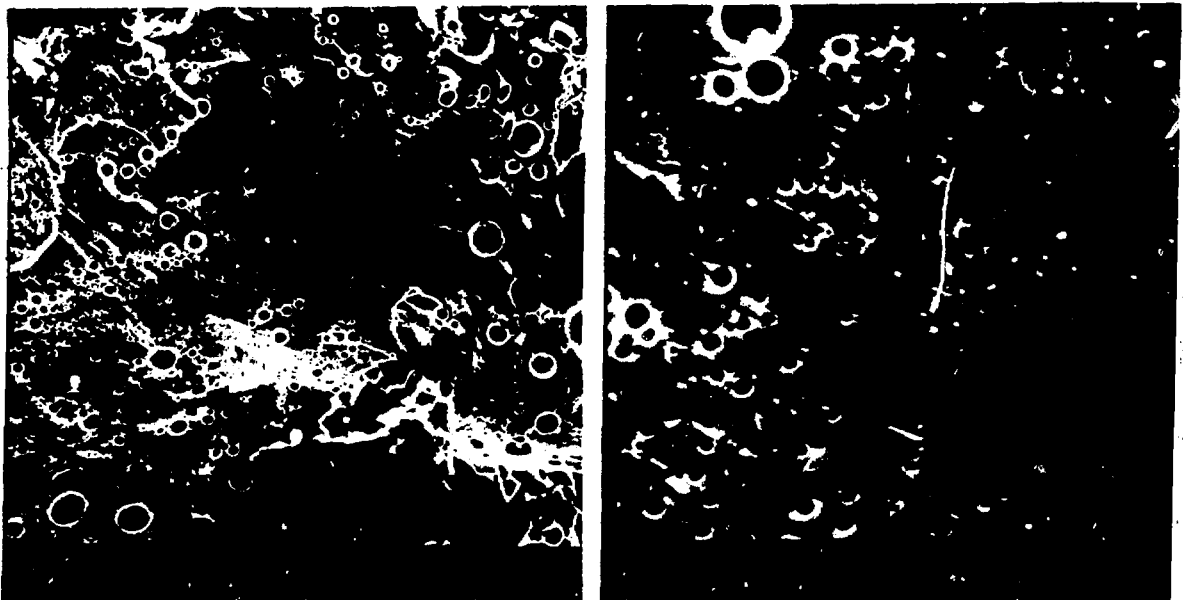


Fig. 10. More Stable Bubble Morphology in Particle Adjacent to That Shown in Fig. 9.