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Irradiation Behavior of Experimental Miniature Uranium Silicide Fuel Plates

by

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1. Introduction

Uranium silicides, because of their relatively high uranium density, were selected as candidate dispersion fuels for the higher fuel densities required in the Reduced Enrichment Research and Test Reactor (RERTR) Program. Irradiation experience with this type of fuel, however, was limited to relatively modest fission densities in the bulk form, on the order of 7×10^{20} cm^{-3} (1), far short of the approximately 20×10^{20} cm^{-3} goal established for the RERTR Program. The purpose of the irradiation experiments on silicide fuels in the ORR, therefore, was to investigate the intrinsic irradiation behavior of uranium silicide as a dispersion fuel. Of particular interest was the interaction between the silicide particles and the aluminum matrix, the swelling behavior of the silicide particles, and the maximum volume fraction of silicide particles that could be contained in the aluminum matrix. The first group of experimental "mini" fuel plates have recently reached the program's goal burnup and are in various stages of examination. Although the results to date indicate some limitations, it appears that within the range of parameters examined thus far the uranium silicide dispersion holds promise for satisfying most of the needs of the RERTR Program.

2. Experiment Description

a. Mini Fuel Plates

The details of the fabrication of the mini fuel plates were presented by Domagala, et al. (2). It suffices here to present briefly some of the parameters and characteristics pertinent to the understanding of the results of the irradiations.

The mini plates are nominally 4.5 inches long by 2 inches wide and were made with two aluminum cladding thicknesses, resulting in 0.05 and 0.06-inch plate thickness. The fuel meat is nominally 0.02 inch thick, and nominal fuel dispersion densities were 30, 40, and 50%. Three fuel types were used, namely U_3Si, U_3SiAl, and U_3Si_2, all in an aluminum matrix. A ^{235}U enrichment of 20% was used for all fuel in this study. Further experimental details are summarized in Table 1.

b. Irradiation Data

The uranium silicide plates are being irradiated in the Oak Ridge Research Reactor (ORR) along with other experimental low-enrichment fuel plates. The plates are stacked in several experiment modules containing twelve plates each, not necessarily all uranium silicide. The plates are cooled by the ORR process water and operate at nominal cladding surface temperatures of ~90°C. The status of the modules that contain ANL silicide plates is given in Table 1.

The following discussion of the irradiation performance deals primarily with Modules 3 and 7, with emphasis on Module 7 because its plates attained the highest fission density of those thus far examined. Some preliminary data from Module 4 are used to indicate trends. The data from Module 2 are suspect because this module underwent a temperature transient in the ORR hot cell that was caused by interrupted decay-heat removal.

3. Postirradiation Data

The postirradiation examination performed at the Alpha-Gamma Hot Cell Facility (AGHCF) at ANL included dimensional measurements, immersion volume measurements, and gamma-scanning on all plates. Metallography was done on selected plates, and SEM and Auger microprobe examinations were done on selected samples. In addition, blister tests were conducted on all three fuel types.

a. Dimensional Changes

Average plate-thickness increases are plotted in Fig. 1 as a function of fission density. The thickness change for the 12 plates in Module 3 ranged from -0.3 to +0.4 mils, with no apparent difference between the U_3Si and U_3SiAl plates. The maximum numbers from this set of plates are therefore plotted - Module 3 at about 0.6×10^{21} cm^{-3}. The four points at the highest fission density are preliminary measurements from Module-4 plates measured at the ORR hot cells. The reported thickness changes are "net" changes after ~1 mil was subtracted for oxide-scale buildup in the reactor water.

At a fission density around 1.8×10^{21} cm^{-3}, the rate of plate thickness change for the U_3SiAl and the U_3Si seem to begin to differ. The Module-4 data show the U_3SiAl plates to be in a stage of rapid growth while the U_3Si plates appear to continue growing at a more moderate rate. Admittedly, the preliminary nature of these measurements and lack of statistical weight of only four points make this conclusion somewhat tenuous at this point. Cladding thickness has no affect on the measured plate-thickness changes.

b. Volume Changes

Immersion volume mesurements were performed on the Module-7 plates after a mild pickling treatment to remove the oxide scale on the surface of the plates. It was assumed that all the volume change occured in the meat; thus, the measurements are expressed in percent volume change of meat and so plotted in Fig. 2. The plates from Module 3, which were not pickled, had meat volume changes ranging from 1 to 2% without a clear pattern; the readings were therefore averaged and so plotted in Fig. 2. The points labeled with an asterisk are calculated from volume changes using the preliminary average thickness measurements and are included to show a possible trend in the data. It should be noted that the high point for the U_3SiAl is for a pillowed plate from Module 4. As was indicated for the thickness data, the U_3SiAl plates seem to have moved into a stage of rapid swelling. A comparison of measured thickness increases with those calculated from volume measurement show the swelling to be approximately anisotropic i.e., $\%\Delta t \simeq \%\Delta V$. This observation is supported by the negligible length change of 0.0-0.2% measured on the plates.

c. Metallography, SEM and Auger Analyses

Metallography has been done on U_3Si and U_3SiAl plates from Modules 3 and 7. A representative micrograph from Module 3 is shown in Fig. 3. The only noteworthy feature is the apparent interaction zone between the fuel particles and the aluminum matrix. The width of this zone ranges from 3 to 6 μm. Auger microscopy more clearly defined the interaction zone, as shown in Fig. 4. Composition gradients were measured in the zone with the Auger microprobe, identifying the zone as being caused by interdiffusion of uranium, aluminum, and silicon.

Some small voids are also visible in Fig. 4, and more clearly in the scanning electron micrograph of a fractured fuel particle shown in Fig. 5. The voids, or gas bubbles, have diameters of 1 μm and less. Metallography from Module 7 shows a drastic change in the bubble morphology, as shown in Figs. 6 and 7. In particular, the U_3SiAl plate, shown in Fig. 6, exhibits a proliferation of fission-gas bubbles. Some bubbles have grown quite large by linking up with their neighbors. The bubbles in the U_3Si plate, Fig. 7, however, are generally smaller; as are the bubbles in some of the fuel particles in the U_3SiAl plate. Both fuels exhibit a variety of bubble sizes, indicating an evolutionary state of bubble morphology that is moving toward a predominance of large bubbles at equilibrium. The bubble volume fractions in the fuel were determined by quantitative metallography to be 8% for the U_3Si plate and 18% for the U_3SiAl plate. The difference in this fuel swelling between the plates is consistent with the thickness measurement (Fig. 1).

A diffusion zone between fuel and matrix was again observed in all types of Module-7 fuel and was measured to be 5-10 μm wide. The zone growth appears to be diffusion controlled with a parabolic ($t^{1/2}$) time dependence. At this rate, the amount of interdiffusion will not be extensive during the lifetime of the plates.

d. Blister Testing

Several plates from both Modules 3 and 7 were blister tested according to accepted blister test procedures. The results are summarized in Table II. The blister temperatures are in an acceptable range and not substantially different from those determined for unirradiated plates.

4. Discussion

Clearly the most important result of these examinations is the observed increase in fuel swelling rate, particularly in the U_3SiAl, above a fission density of $\sim 1.5 \times 10^{21}$ cm^{-3}. The U_3Si may be following a similar path, but its swelling begins at a higher fission density. It is evident from the metallography that fission-gas-bubble growth is responsible for the increased swelling rate. The first evidence of fission-gas-bubble formation was found in Module 3 plates at a fission density of $\sim 0.6 \times 10^{21}$ cm^{-3}. The largest bubbles measured were approximately 1 μm in diameter (see Fig. 5), and the pore volume fraction was about 2%. At a fission density of 2×10^{21} cm^{-3} in Module 7, there were still many fuel particles in the U_3Si and some in the U_3SiAl that had low (2 - 6%) pore volume fractions. Gas bubbles in these fuel particles are probably in a quasi-equilibrium state, a state where the internal fission-gas pressure is balanced by surface tension and stress in the surrounding fuel. The gas atom mobility is evidently sufficiently enhanced by fissioning to allow for diffusion of gas into the existing small bubbles (perhaps originally irradiation-induced voids), resulting in a gradual increase of gas presure. At a certain pressure the voids become unstable and grow at a very high rate until a new quasi-equilibrium is reached, resulting in particles with pore volume fractions exceeding 50%.

This accelerated swelling appears to occur first in the U_3SiAl fuel, but the evidence suggests that the U_3Si fuel will follow a similar path. It is likely that eventually all fuel particles will attain high porosity. The pillowed U_3SiAl plate with a thickness change of 12 mils in Module 4 (see Fig. 1) has probably passed this accelerated swelling stage and moved on to pillowing. Obviously, for the fuel plates to behave in a satisfactory fashion the unstable accelerated swelling range has to be avoided. The limiting factor in the behavior of the plates appears to be the burnup reached by the individual fuel particles not the fission density in the fuel meat. According to our interpretation of the data, this means keeping the U_3Al below 75% ^{235}U depletion and the U_3Si below 85% for 20%-enriched fuel in order to avoid excessive swelling.

In terms of fission density per unit-volume of fuel meat, then, 2×10^{21} cm^{-3} should be attainable by operating with fuel volume fractions in the 40% range but with more modest ^{235}U depletion levels. Miniplates containing 40 and 50 vol% fuel now in the RERTR irradiation program afford a means of verifying this concept soon.

5. Conclusions

The twelve experimental silicide dispersion fuel plates that were irradiated to approximately their goal exposure show the 30-vol% U_3SiAl plates to be in a stage of relatively rapid fission-gas-driven swelling at a fission density of 2×10^{21} cm^{-3}. This fuel swelling will likely result in unacceptably large plate-thickness increases. The U_3Si plates appear to be superior in this respect; however, they, too, are starting to move into the rapid fuel-swelling stage.

- Analysis of the currently available post irradiation data indicates that a 40-vol% dispersed fuel may offer an acceptable margin to the onset of unstable thickness changes at exposures of 2×10^{21} fission/cm^3.

- The interdiffusion between fuel and matrix aluminum was found not to be a problem.

- Blister temperatures were in the acceptable magnitude of higher than 500°C.

- Examination of the remaining plates that contain higher fuel dispersion densities, and more detailed analysis of already available data, are needed to fully characterize the irradiation behavior of the silicide and substantiate the potential of this fuel in the RERTR Program.

REFERENCES

1. M. A. Feraday, et al., *The Irradiation Behavior of U Si Elements to High Burnup*, AECL-4058, Chalk Raiver, Ontario, March 1974.

2. R. F. Domagala, et al., *U-Si-Al Dispersion Fuel Alloy Development for Research and Test Reactors*, This Conference.

TABLE 1. Miniplate Fabrication and Irradiation Data

Module Number	Number of Plates	Fuel Type	Nominal Fuel Vol. Fraction, %	^{235}U Depl., %	U Density, g/cm^3	Max. Fission Density, 10^{21} cm^{-3}	Current Location
2	3	U_3SiAl	30-40	77	4.6-5.9	2.1	ANL
3	12	U_3Si U_3SiAl	30	34	4.8 4.6	0.7	ANL
4	4	U_3Si U_3SiAl	30	88	4.8 4.6	2.0	ORNL Poolside
7	12	U_3Si U_3SiAl U_3Si_2	30	83	4.8 4.6 3.8	1.9	ANL
10	12	U_3Si U_3SiAl	40-50	65	5.7 5.8-6.4	1.9	ORNL Poolside
13	12	U_3SiAl	45-55	28	5.9-7.0	1.2	ORNL Poolside

Table II. Summary of Miniplate Blister-Threshold Temperature Data

Fuel Type	No. of Plates Tested	Uranium Density, Mg/m^3	Fission Density, 10^{27} fiss./m^3	Blister-Threshold Temperature, °C: Lower Limit	Blister-Threshold Temperature, °C: Most Probable
U_3Si-Al	2	4.80-4.81	1.9-2.0	475	500
U_3Si-Al	2	4.79-4.81	0.7	500	510
U_3SiAl-Al	1	4.51	1.8	475	500
U_3SiAl-Al	2	4.68	0.7	500	515
U_3Si_2-Al	1	3.75	1.5	515	530

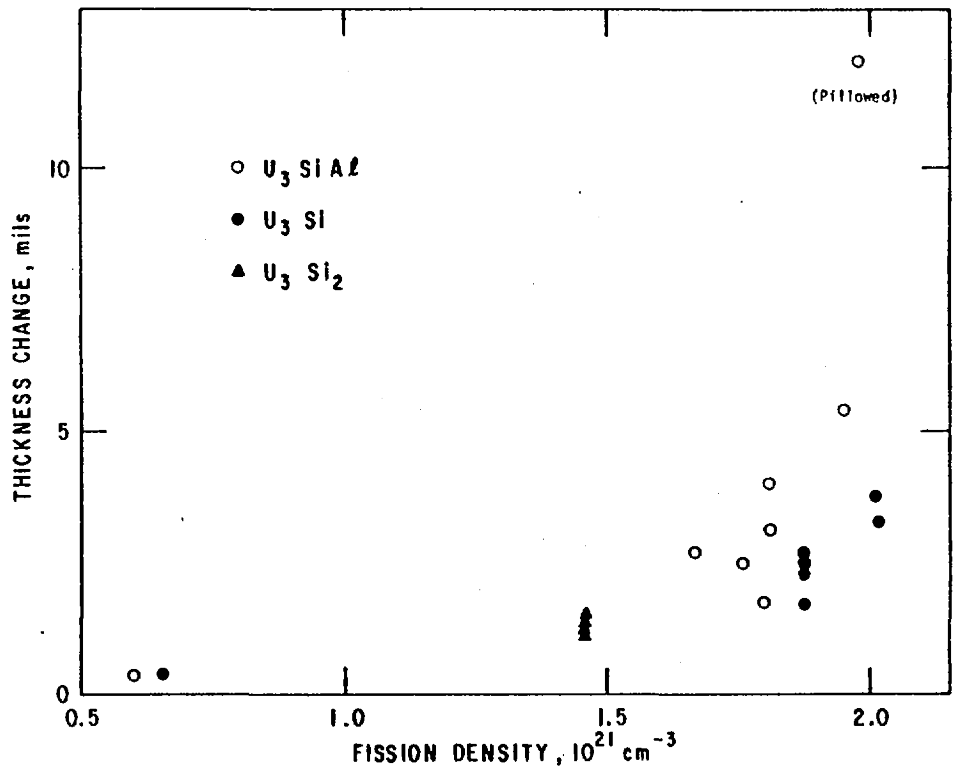

Fig. 1. Measured Increase in Fuel-plate Thickness.
Conversion factor: 1 mil = 25.4 μm

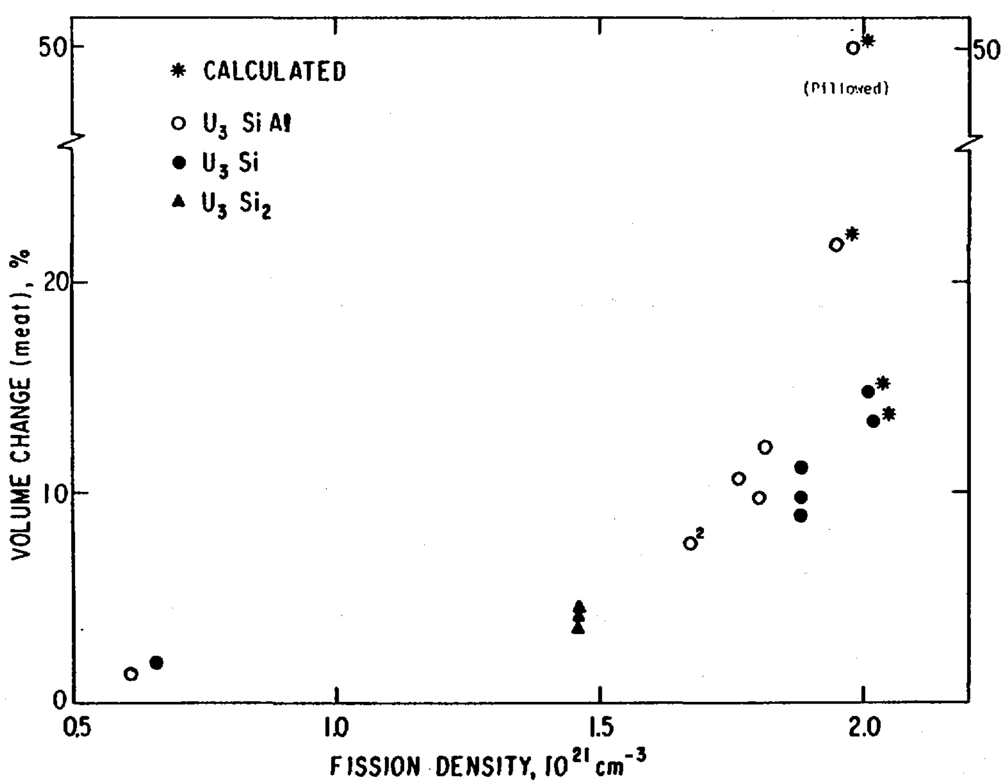

Fig. 2. Measured and Calculated Heat Volume Increase.

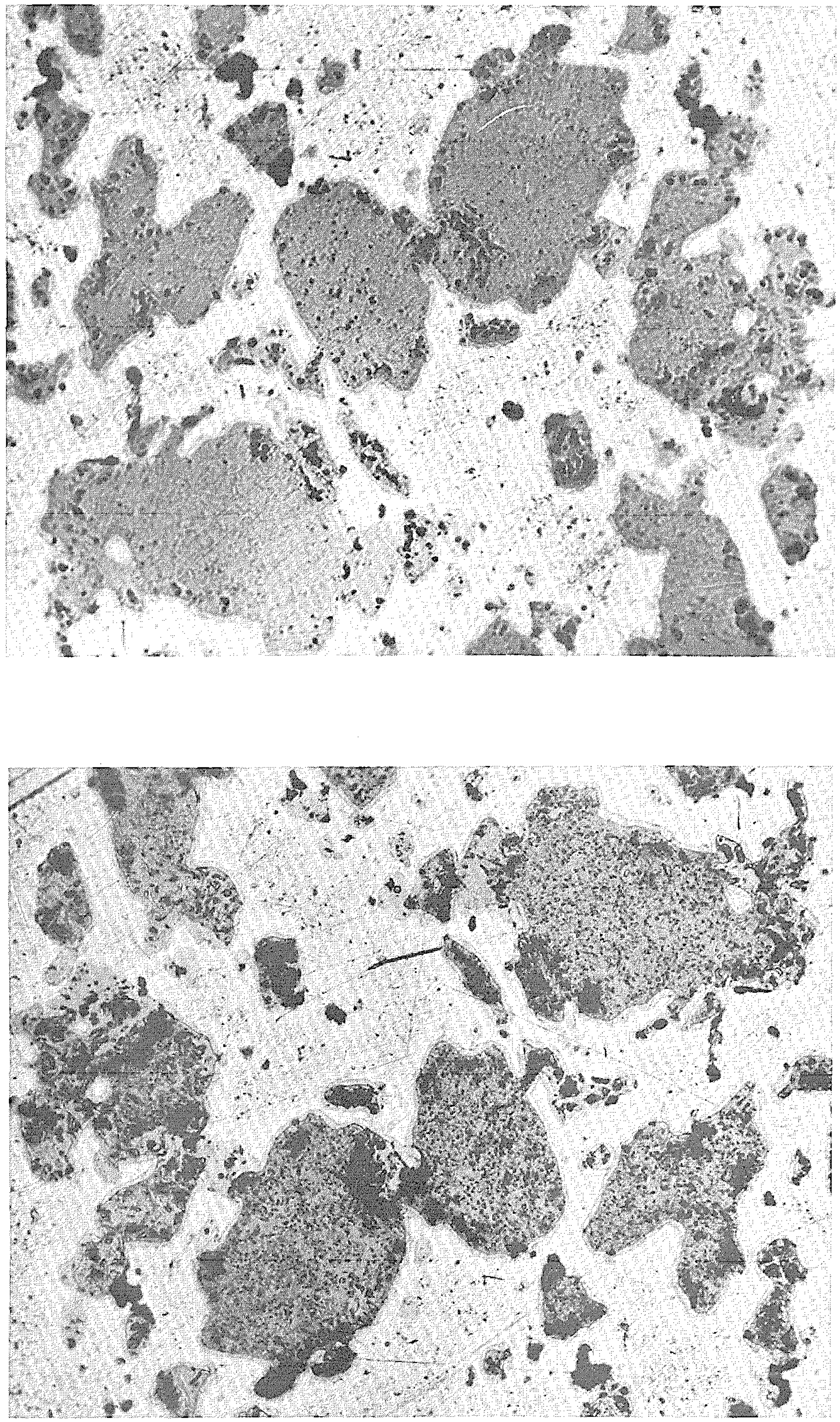

Fig. 3. Microstructure of U_3SiAl Plates from Module-3 (as polished and etched); 250x

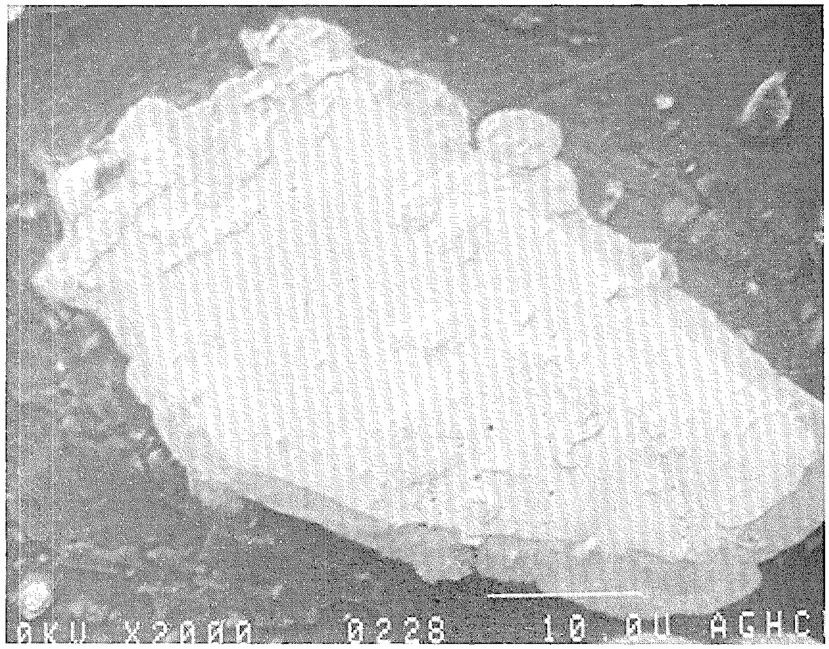

Fig. 4. Auger Micrograph of U_3SiAl Fuel Particle from Module-3 Plate

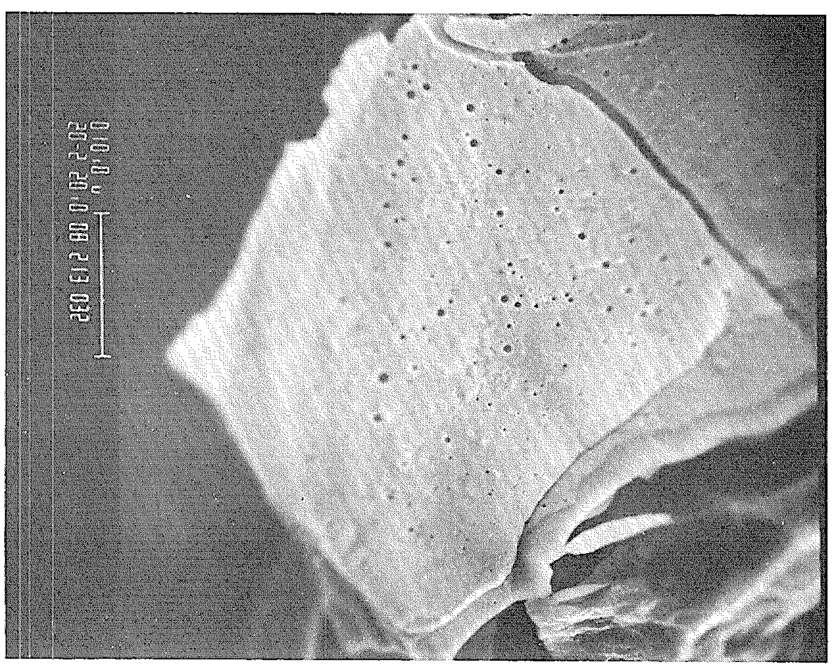

Fig. 5. Scanning Electron Micrograph of Fractured U_3SiAl Fuel Particle from Module-3 Plate

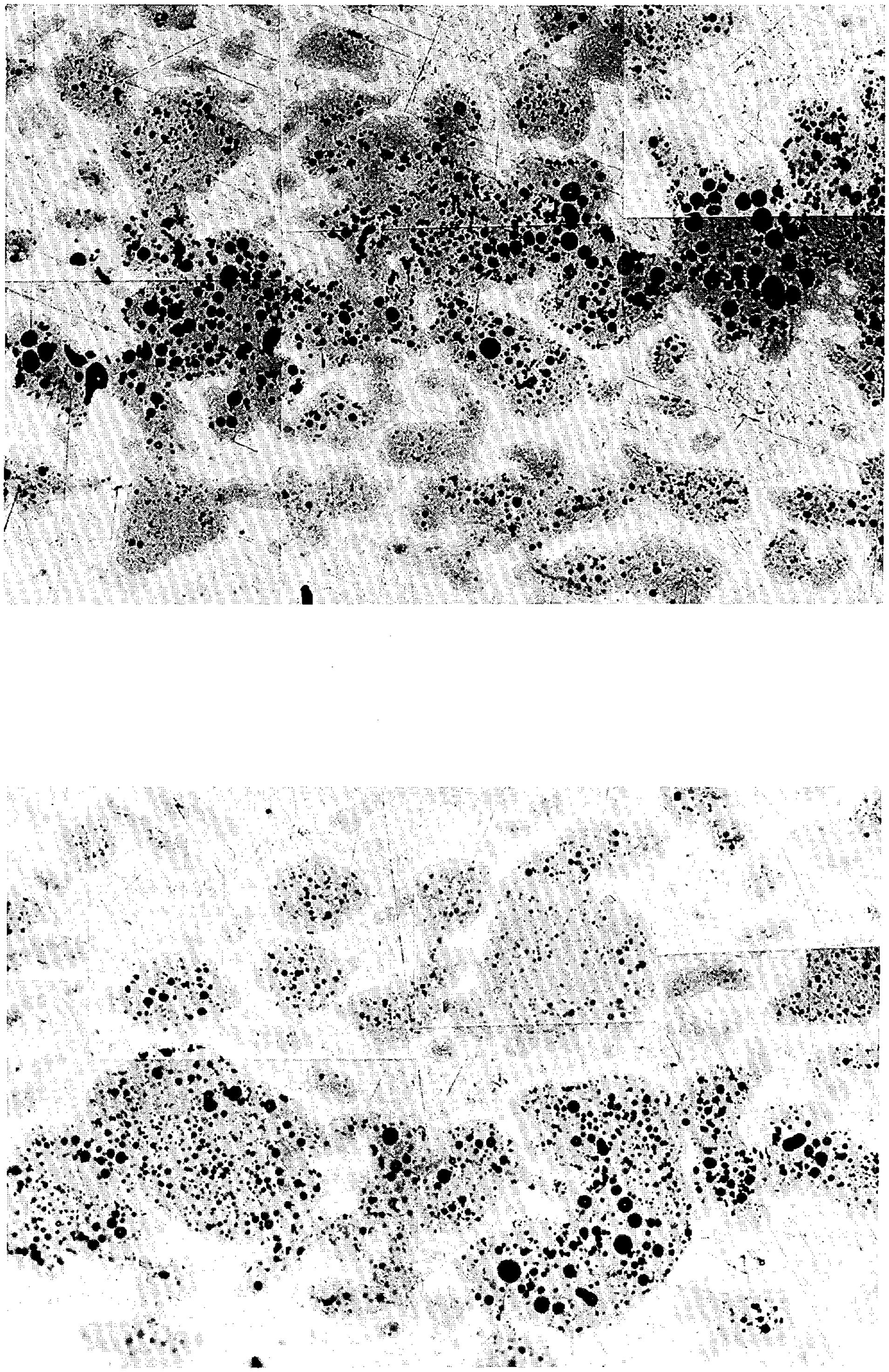

Fig. 6. Microstructure of U_3SiAl Plate from Module 7 ; 200x

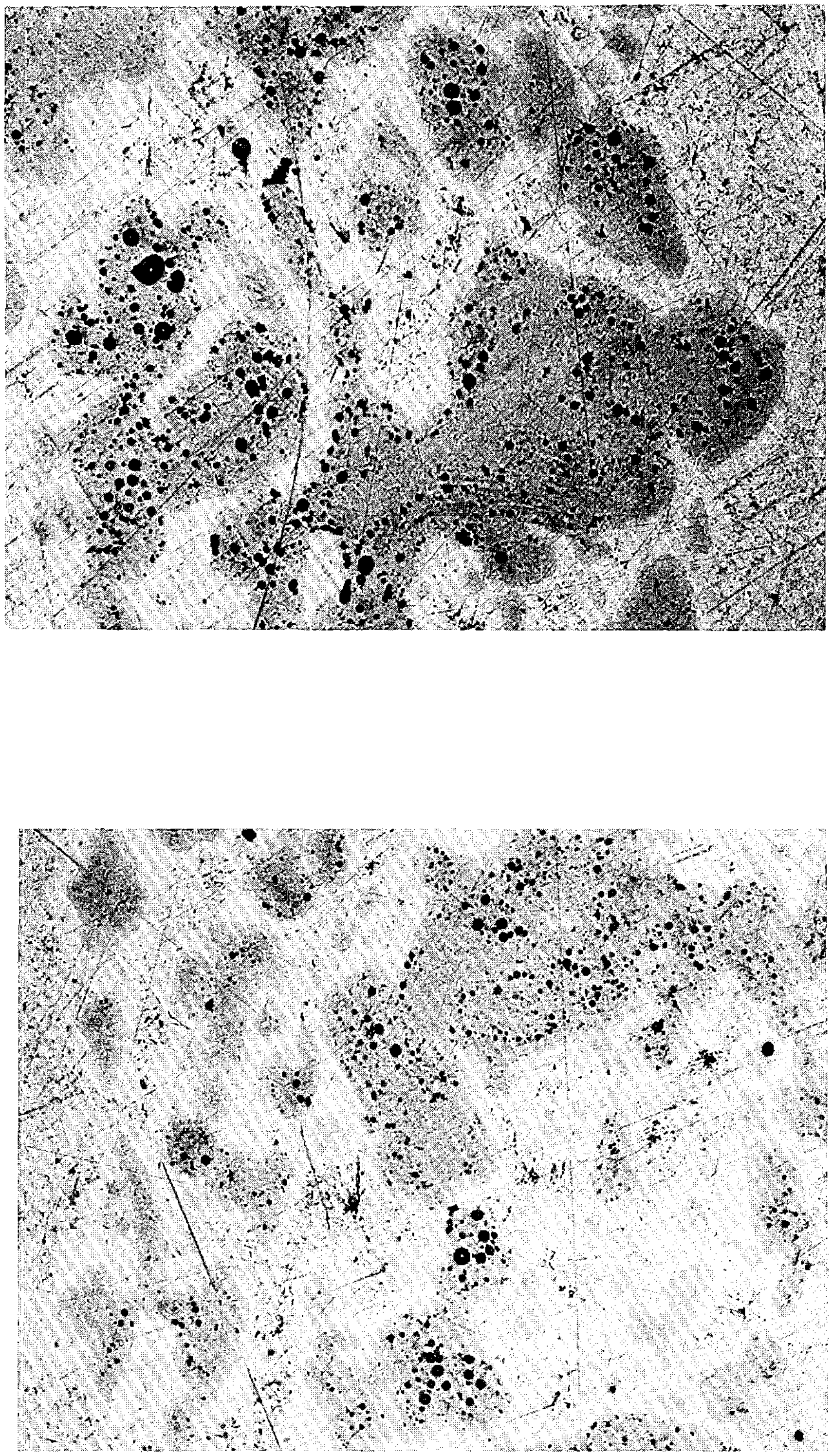

Fig. 7. Microstructure of U_3Si Plate From Module 7; 200x