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COMPARISON OF IRRADIATION BEHAVIOR OF DIFFERENT URANIUM SILICIDE DISPERSION FUEL ELEMENT DESIGNS*

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ABSTRACT

Calculations of fuel swelling of U_3SiAl-Al and U_3Si_2 were performed for various dispersion fuel element designs. Breakaway swelling criteria in the form of critical fuel volume fractions were derived with data obtained from U_3SiAl-Al plate irradiations. The results of the analysis show that rod-type elements remain well below the pillowing threshold. However, tubular fuel elements, which behave essentially like plates, will likely develop pillows or blisters at around 90% ^{235}U burnup. The U_3Si_2-Al compounds demonstrate stable swelling behavior throughout the entire burnup range for all fuel element designs.

INTRODUCTION

Uranium silicides have been widely considered as a low-enriched dispersion fuel because of their relatively high density (15.2 g cm^{-3} for U_3Si and 12.2 g cm^{-3} for U_3Si_2). A variation of U_3Si containing from 1 to 2% aluminum, which we call U_3SiAl, has been favored by certain fuel developers because of superior corrosion resistance in water. This aluminum-containing compound is somewhat less dense (14.2 g cm^{-3}) than the pure binary compound.

Irradiation experiments have shown that U_3Si and U_3SiAl are prone to excessive swelling (breakaway swelling) that commences at cumulative fission densities of about 5.5 x 10^{27} fissions m^{-3} and at about 4.5 x 10^{27} fissions m^{-3}, respectively. U_3Si_2 on the other hand has consistently shown very stable swelling behavior[1].

Breakaway swelling leads to blistering or pillowing of fuel elements and therefore appears to render compounds afflicted with this property unsuitable for high-fission-density (high-^{235}U-burnup) applications. However, because breakaway swelling is associated with fission-induced high plasticity of the fuel, it is susceptible to external restraint. The magnitude of mechanical restraint imposed on swelling fuel particles in a dispersion fuel element depends on the fuel element design, i.e., on the amount of matrix aluminum surrounding the fuel particles and on the shape of the element, be it a flat, thin plate, a cylindrical tube, or a solid rod.

This paper examines the swelling behavior of these silicide compounds in various research reactor fuel element designs.

FUEL SWELLING

The swelling data presented here were obtained in experimental irradiations as part of the Reduced Enrichment Research and Test Reactor (RERTR) program. The majority of the data came from so-called "miniplates" that have nominal dimensions of 115 x 50 x 1.3 mm and are fabricated by hot rolling. The fuel core or meat is typically 33-40% of the plate thickness and contains up to 50 vol.% of the fuel particles, up to 20 vol.% voids, and a balance of pure Al powder. The cladding may be made of various Al alloys. These experimental plates were fabricated by NUKEM (Germany), CNEA (Argentina), and ANL (USA).

A separate group of experiments consisted of hot extruded rods of 7-mm diameter with a core of 5.5-mm diameter containing up to 29 vol.% of fuel particles and a small fraction, ~2 vol.%, of voids. These rod-type elements were made and tested by AECL-Chalk River[2]. Finally, recent Russian data from irradiation experiments with extruded tubular fuel elements that contain 34 vol.% U_3SiAl and 3% voids[3] are used in this study.

Fuel core swelling is determined by measuring the volume change of the miniplate or rod after irradiation using the Archimedean immersion method and by subtraction of the cladding volume. The fuel particle swelling $\frac{\Delta V^F}{V_0^F}$ is calculated from the core volume changes $\frac{\Delta V^C}{V_0^C}$ as follows:

$$\frac{\Delta V^F}{V_0^F} = \left[\frac{\Delta V^C}{V_0^C} - \frac{V_0^P - V^P}{V_0^C}\right]\frac{V_0^C}{V_0^F}$$

where ΔV^C is the change in core volume during irradiation, V^P is the amount of as-fabricated porosity remaining in the core after irradiation, and $\frac{V_0^F}{V_0^C}$ is the as-fabricated fuel volume fraction in the core.

Experimental data for both U_3SiAl and U_3Si are shown in Fig. 1. The different swelling behavior of the two compounds and the effect of as-fabricated fuel volume fraction on eventual plate failure by pillowing are apparent.

The particular swelling behavior of the two compounds is due to a somewhat different fission gas bubble development. The bubbles in U_3SiAl are generally larger than those in U_3Si. The bubbles in U_3SiAl eventually interlink across fuel particles, leading to the development of large cavities, enormous (breakaway) swelling, and pillowing of the fuel plate (see Fig. 2). Breakaway swelling and pillowing also occurs in U_3Si at larger as-fabricated fuel loadings, but always at higher fission densities or burnups than in U_3SiAl because of the higher swelling rate of U_3SiAl.

The appearance of the interlinked bubbles suggests a very plastic, viscous behavior of the fuel during irradiation. This behavior has been ascribed to fission-induced amorphization (mictametization) of U_3Si and U_3SiAl.[4]

The effect of fuel loading on breakaway swelling and pillowing is due to the fact that at higher as-fabricated fuel volume fractions the swelling particles contact each other at lower fission density, giving rise to earlier translinkage of gas bubbles.

Since the fuel is evidently very plastic, it should be sensitive to external mechanical restraint. This is clearly demonstrated by the swelling data from rod-type elements, where the swelling fuel particles are under relatively high hydrostatic compression provided by the relatively thick cylindrical cladding.

Swelling of the U_3Si_2 is fundamentally different from that of U_3Si. Fission gas bubbles in the visible range (by SEM) form after a fission density of approximately 3.5 x 10^{27} cm^{-3} causing an increase in swelling rate. The fission gas bubbles, however, remain relatively small and do not interconnect as in the case in U_3Si. The result is the absence of break-away swelling, and of pillowing in highly-loaded dispersion, even at complete ^{235}U burnup. Because of the small bubble size, the affect of external restraint on swelling should be less pronounced in U_3Si_2 than in U_3Si.

IRRADIATION BEHAVIOR MODELING

Our dispersion fuel behavior code DART[4] was used to model the fuel swelling for three different fuel element designs. Modifications to the DART mechanical model were implemented in order to include the constraining effects of the cladding in plate, tube, and rod configurations. The DART mechanical (stress) model consists of a fuel sphere which deforms due to both solid fission product and fission gas-bubble swelling. The fuel sphere is surrounded by an Al matrix shell, which is assumed to behave in a perfectly plastic manner and which deforms (yields) due to fuel particle volume expansion. The effects of the cladding are included by a suitable adjustment of the effective Al volume fraction. Currently, the effects of creep are not included; instead, the stress relaxation is approximated by lowering the Al yield stress to an "effective" value. The deformation of the matrix and cladding material generates stresses within the expanding fuel particles which affect the swelling rate of the fission gas bubbles. Subsequent to the closure of the as-fabricated porosity, the swelling rate is primarily dependent on the plastic yielding of the Al matrix and cladding. At this point, the hydrostatic stress, σ, acting on the gas bubbles is given by

$$\sigma = \frac{2}{3} (1 - \ln \left(\frac{V^F \, x \, \Delta V^F}{V_0^C} \right) \beta \sigma_{Al}^Y(T)$$

where $\sigma_{Al}^Y(T)$ is the as-fabricated temperature-dependent yield strength of the Al, and β is a factor which accounts for the effects of irradiation (e.g., irradiation-enhanced creep).

U_3Si, U_3SiAl Fuel

The effective yield stress of irradiated Al matrix material and cladding (i.e., the value of β in the above equation was determined by comparing the results of DART calculations with postirradiation immersion volume measurement of U_3SiAl plates (shown in Fig. 1). This value of the yield stress was then used in all subsequent simulations. Results of the DART calculations are shown in Fig. 3. The lower calculated fuel swelling in the rod-type element is due an assumed biaxial stress state as compared to an assumed uniaxial stress state for the plate and thin-walled tube geometries. Elastic analysis and comparison of plate and tube swelling data[3] support the assumptions. Thermal stress analysis[5] of a thin-walled hollow cylinder shows that the circumferential and axial stresses at the inner surface are only half as great as those in a cylinder with a small bore for the same temperature difference. In addition, the thin-walled hollow cylinder can be treated as a cylindrical shell which can be shown to have a complimentary solution to that obtained for a rectangular beam on an elastic foundation. Fuel swelling in plates results in plate thickness increase only, while plate width and length remain relatively unchanged. Likewise, in tubes, only the wall thickness increases and the overall diameter remains unchanged. There is thus minimal lateral or circumferential strain in the cladding of these element designs and consequently much less restraint compared to the hoop stress state existing in a solid-clad rod.

Results from postirradiation immersion volume measurements at a peak ^{235}U burnup of 70%[3], and a quantitative determination of the fission gas bubble volume fraction obtained by image analysis of fuel meat metallographs compare well with the calculated fuel swelling of the tubular fuel element as shown in Fig. 3. These results are also supported by comparison of calculated bubble-size distributions with the observed bubble morphology in the plate and rod configurations.

Breakaway swelling criteria in the form of critical fuel volume fractions were derived from data obtained from the U_3SiAl plate irradiations. As shown in Fig. 1, all U_3SiAl plates exhibit breakaway swelling and experience pillowing. A pillowing threshold was derived based on the observed effect of loading on pillowing, shown in Fig. 4. When a critical fuel volume fraction in the core is reached, translinkage of gas bubbles becomes prevalent, resulting in large cavity formation, breakaway swelling and pillowing. This situation is shown in Fig. 1 for plates and for the rod- and tube-type fuel elements modeled. Clearly the rod-type element remains well below the pillowing threshold up to complete ^{235}U burnup. However, the tubular fuel element, since it behaves essentially like a plate, will likely develop pillows or blisters at around 90% ^{235}U burnup.

U_3Si_2 Fuel

Figure 5 shows DART-calculated results for fuel-particle swelling of low-enriched (LEU) U_3Si_2-Al fuel plates as a function of fission density for two values of the Al-matrix yield strength (i.e., two values of β), and for U_3Si_2-Al rods for two values of the fission density at which recrystallization is predicted to occur. The calculations shown in Fig. 5 were made in the spirit of the theory presented in references 4 and 6 of irradiation-induced recrystallization in U_3Si_2 and UO_2 fuels. After recrystallization occurs, the gas-atom diffusion to the grain boundaries, bubble nucleation, and accelerated growth (relative to that of bubbles in the bulk material) result in an increased swelling rate, as shown in Fig. 5. The calculations shown in Fig. 5 were made for a homogeneous fuel at a constant temperature. In reality, time-dependent temperature and flux gradients exist across the plate and rod during irradiation. The two curves for the U_3Si_2-Al rod show the effect of such a gradient on the calculated results.

CONCLUSIONS

Irradiation experiments have shown that plate-type dispersion fuel elements can develop blisters or pillows at high ^{235}U burnup when fuel compounds exhibiting breakaway swelling such as U_3SiAl and U_3Si, are used at moderate to high volume fractions. Calculations indicate that tubular fuel elements behave similarly. Rod-type fuel elements, however, are inherently more resistant to pillowing. With a stable swelling compound, such as U_3Si^2, blistering or pillowing is entirely eliminated.

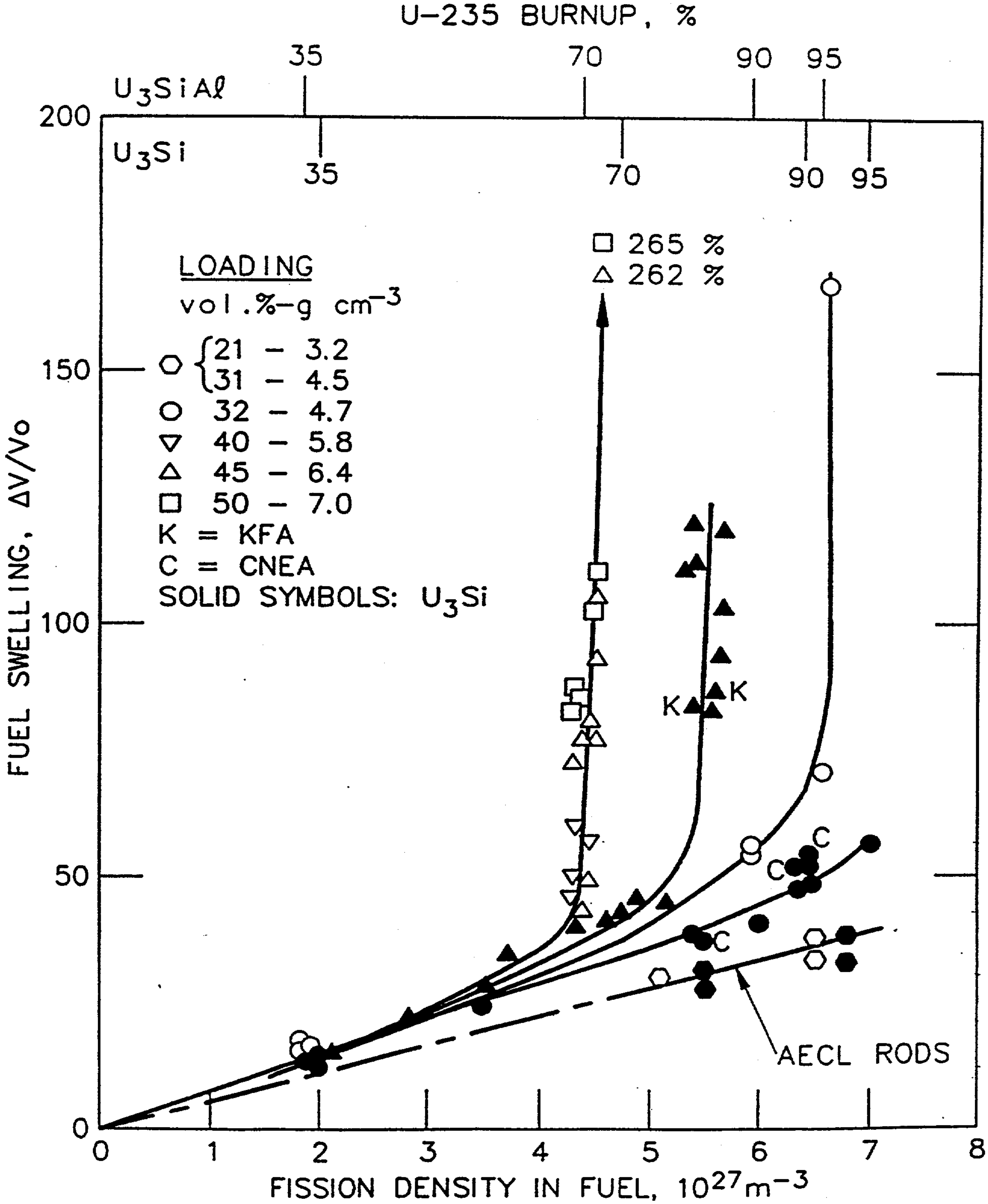

Figure 1. **Fuel Particle Swelling in Experimental U_3Si and U_3SiAl Dispersion Fuel Plates and Rods**

Figure 2. Metallographic Cross-section of High Burnup LEU, U_3Si Dispersion Fuel Plate Showing Breakaway Swelling

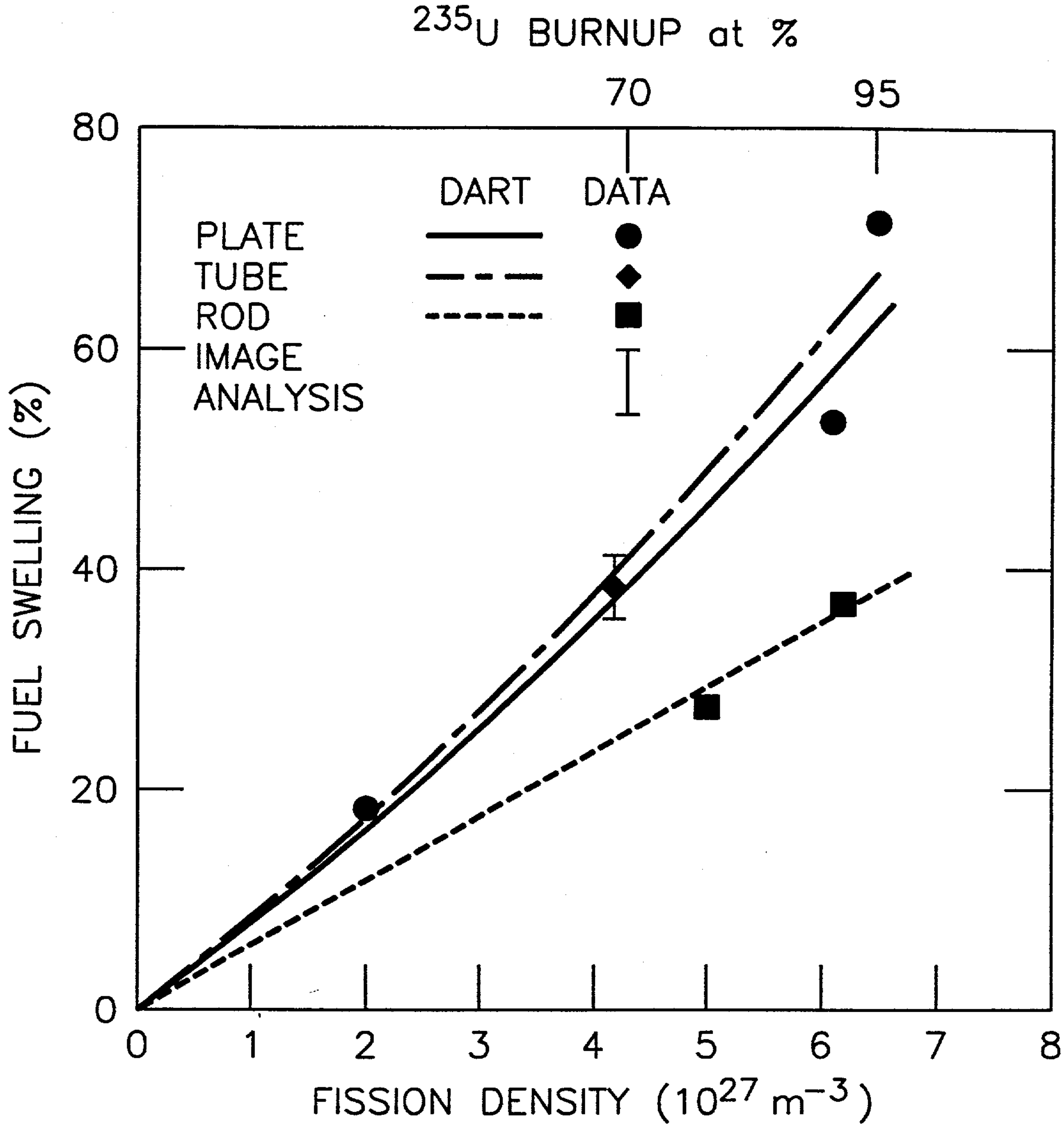

Figure 3. **DART-Calculated Fuel Swelling for U_3SiAl-Al as a Function of Fission Density for Different Fuel Element Designs Compared with Data**

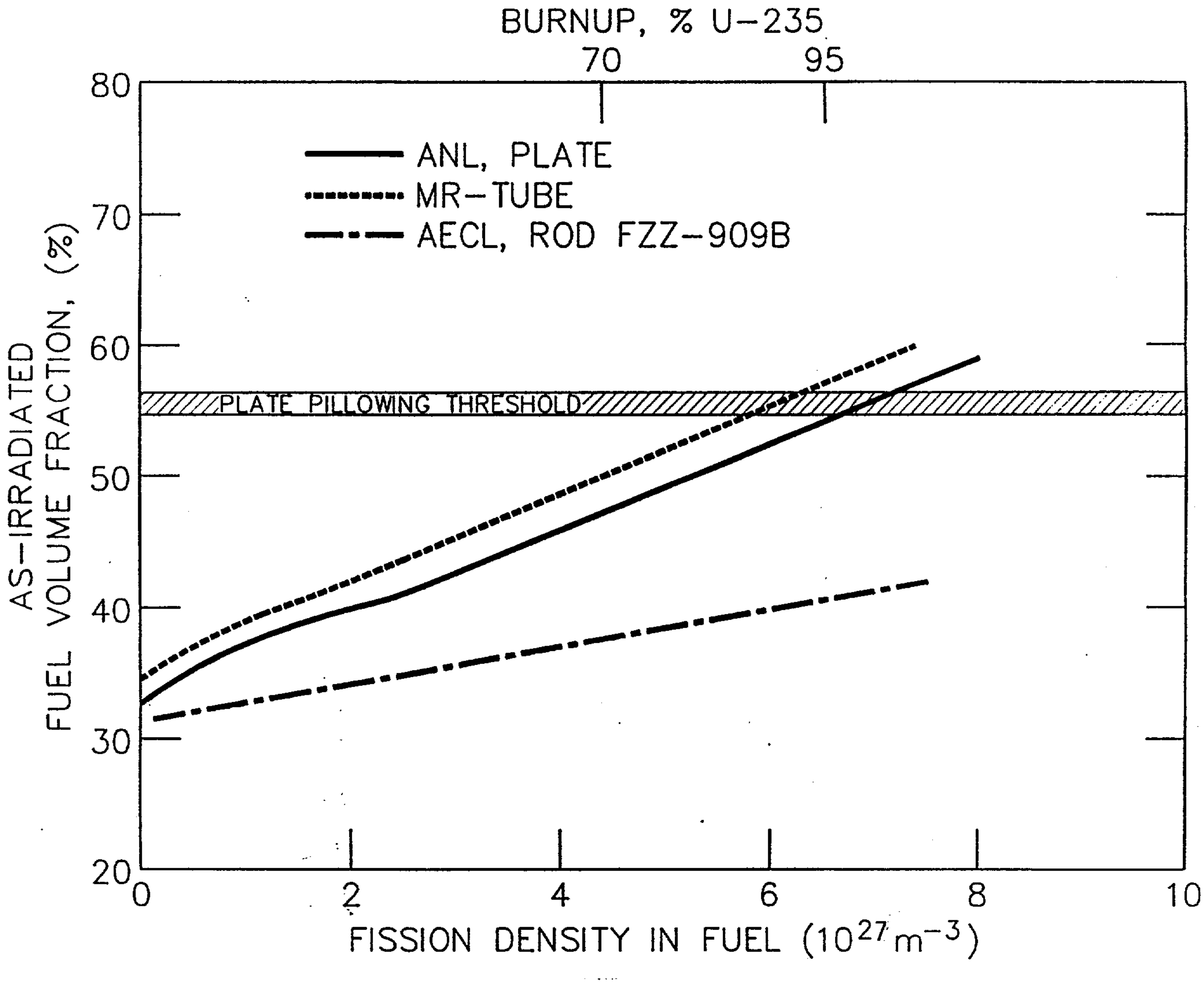

Figure 4. **DART Calculation of U_3SiAl Volume Fraction in Meat of Rod, Tube, and Plate Dispersion Fuel Elements and Pillowing Threshold Derived from Plate Irradiation Experiment**

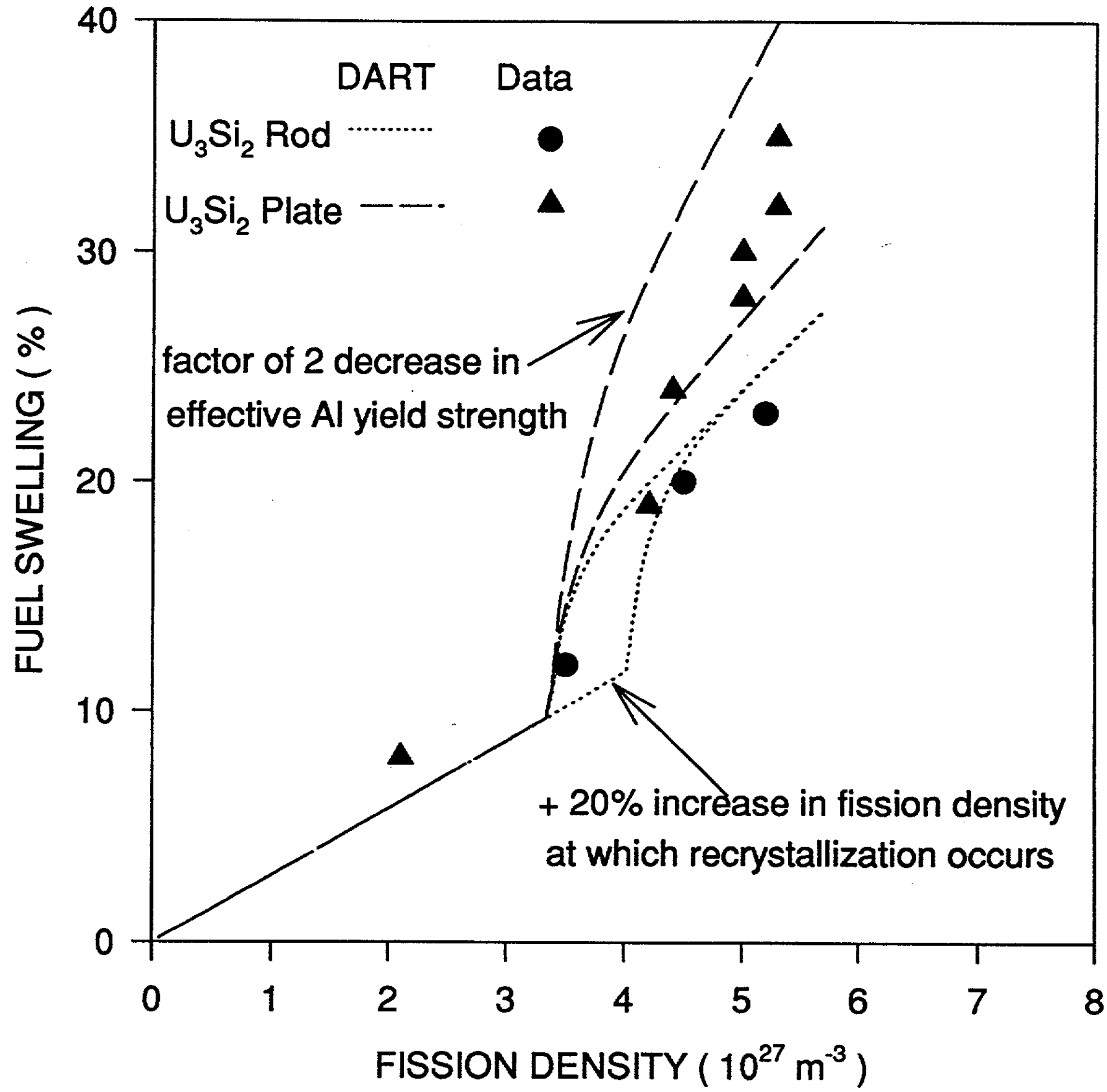

Figure 5. **DART-Calculated Fuel Swelling for U_3Si_2-Al as a Function of Fission Density for Rod and Plate-type Geometries Compared with Data**

REFERENCES

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

2. D. F. Sears, "The Development and Irradiation Testing of Al-U_3Si_2 fuel at Chalk River," Proceedings of the 14th International Meeting on Reduced Enrichment for Research and Test Reactors, Jakarta, Indonesia, November 4-7, 1991,(to be published).

3. G. A. Sarakhova, Y. A. Stetsky, V. D. Zaboikin, and V. B. Suprun, "Development of Fuel Elements for Russian Research Reactors with Fuel of Lower Enrichment," Proceedings of the 16th International Meeting on Reduced Enrichment for Research and Test Reactors, Oarai, Japan, October 4-7, 1993, JAERI-M-94-042 (1994).

4. J. Rest and G. L. Hofman, "Fundamental Aspects of Inert Gasses in Solids," S. E. Donnelly and J. H. Evans, Plenum Press, 443 (1991).

5. Z. Zulows, T. C. Yen, and W. H. Steiselmann, Thermal Stress Techniques in the Nuclear Industry, Chap. 3, American Elsevien Publishing Company, Inc., New York (1965).

6. J. Rest and J. L. Hofman, "Dynamics of Irradiation-induced Grain Subdivision and Swelling in U_3Si_2 and UO_2 Fuels," Journal of Nuclear Materials, pp. 187-202 (1994).