ADVANCES IN THE UNDERSTANDING OF U_3Si_2-Al DISPERSION FUEL IRRADIATION BEHAVIOR

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SWELLING BEHAVIOR DURING IRRADIATION

- Around 1987, at the beginning of ANL's participation in the ANS Project, we had found that certain intermetallic compounds, e.g., UAl_x, U_3Si_2, and USi, exhibited very stable swelling behavior to full burnup with LEU, while other compounds, e.g., U_6Fe, U_6Mn, and U_3Si became unstable before full burnup was achieved.

- We concluded that the unstable, or break-away, swelling is associated with irradiation-induced amorphization and that the compounds exhibiting stable swelling remained crystalline during irradiation.

SWELLING BEHAVIOR DURING IRRADIATION (CONT'D)

- Irradiation of MEU and HEU U_3Si_2 showed that the swelling fell below the LEU values. Detailed analysis revealed a two-stage swelling behavior with an apparent fission rate effect.

- The fission gas bubbles in U_3Si_2 were distributed in a very regular pattern to the highest fission densities reached in our tests in the ORR (for ~65% burnup in HEU particles). The bubbles grew in diameter with increasing fission density but did not coalesce.

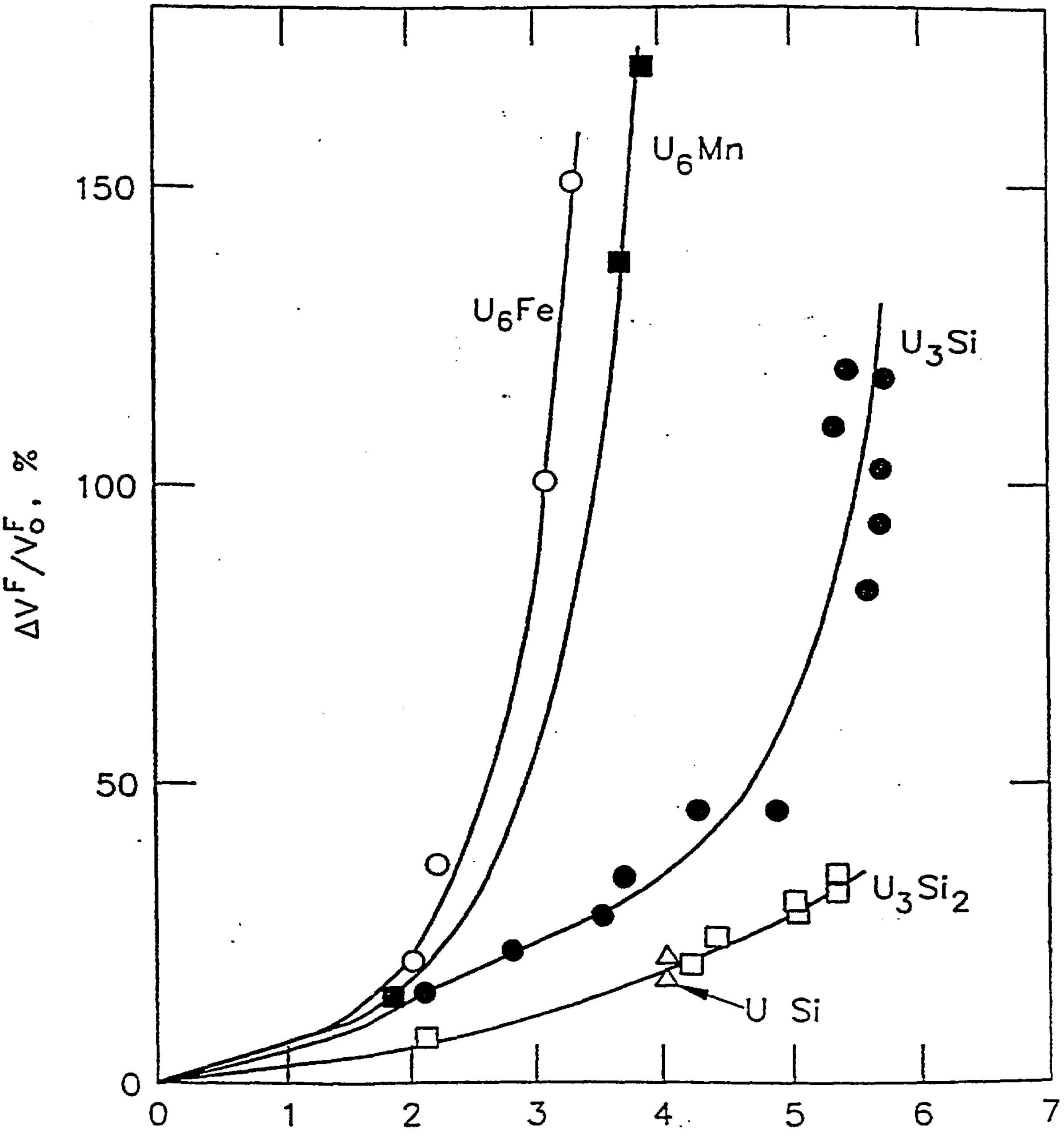
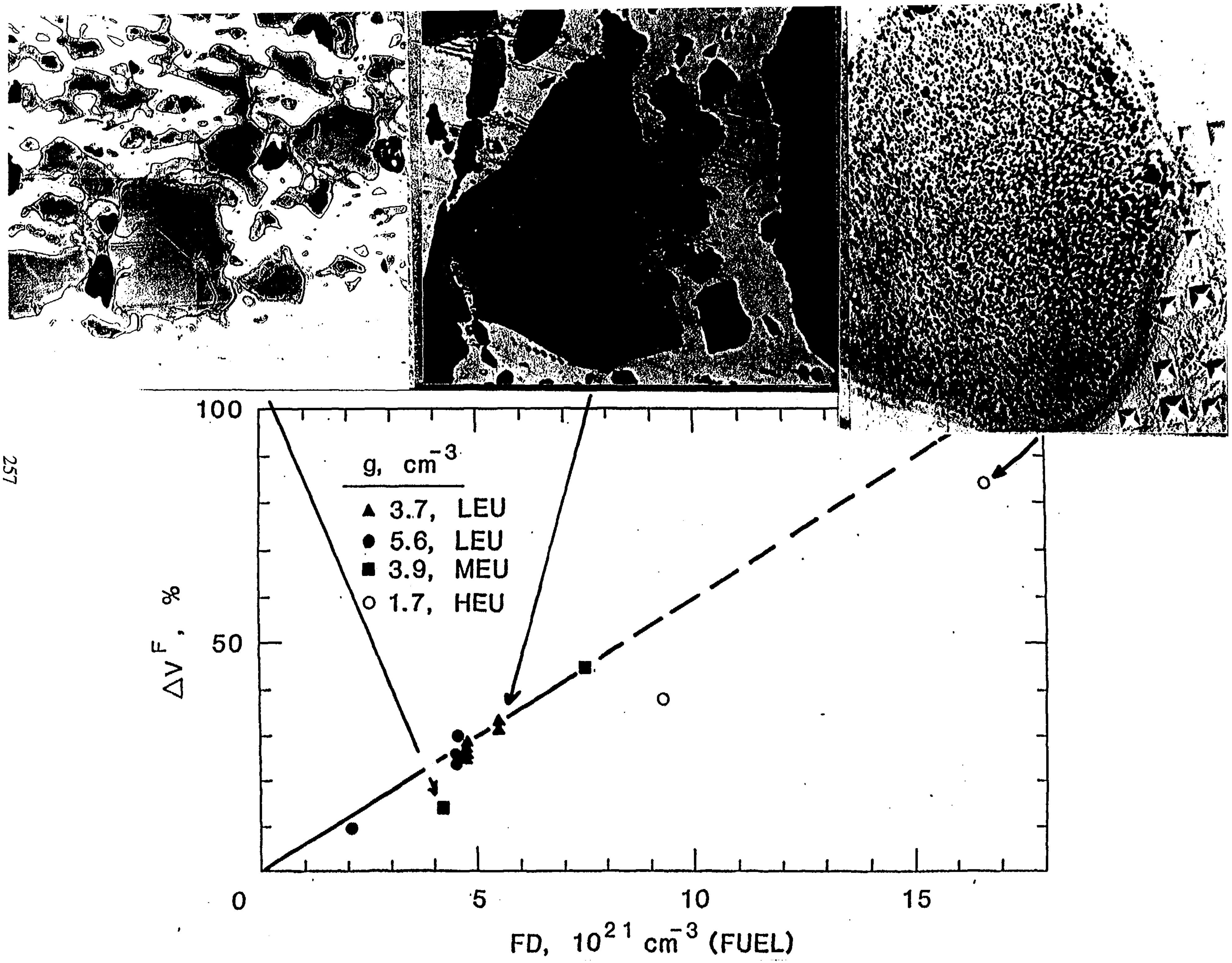
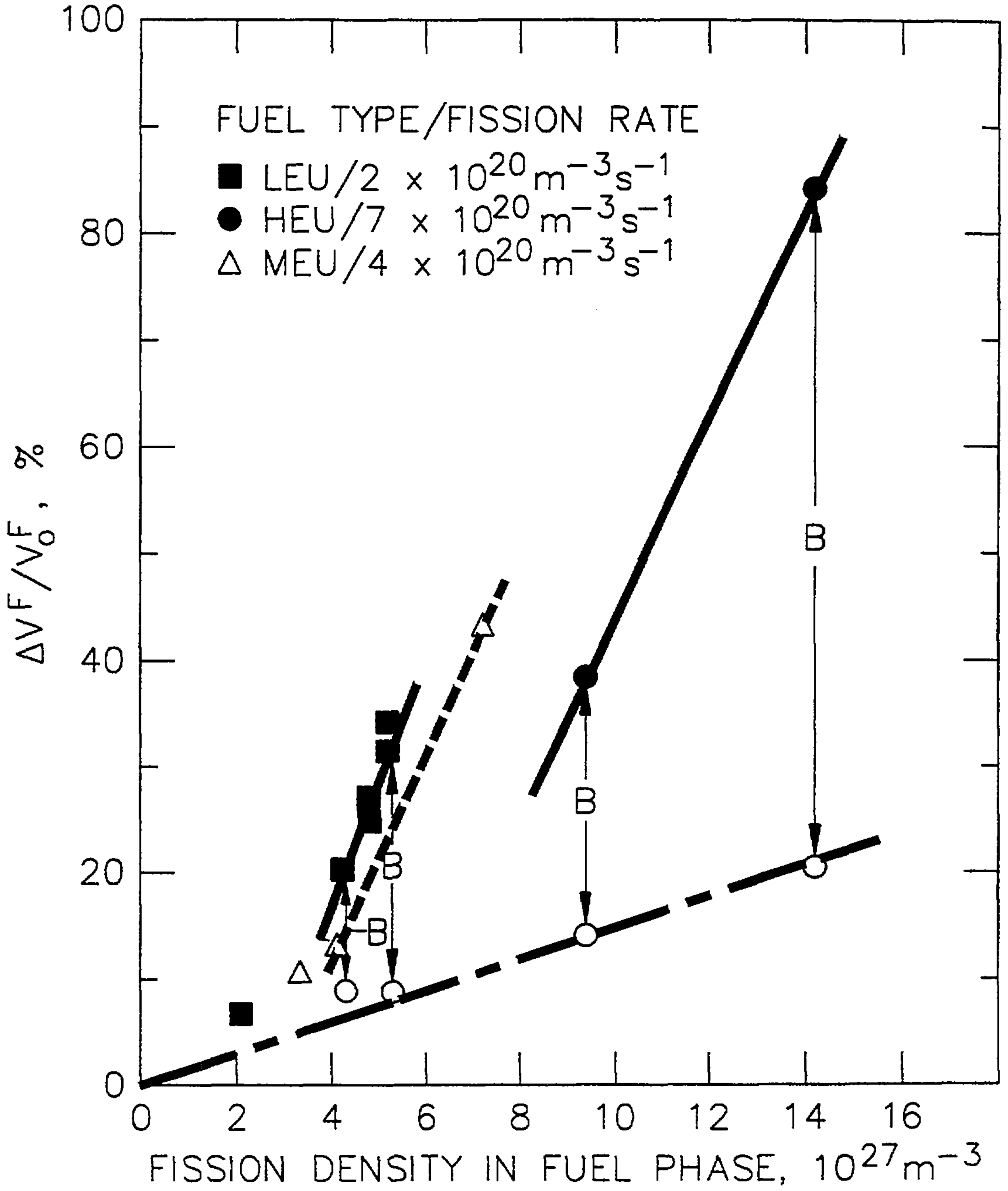
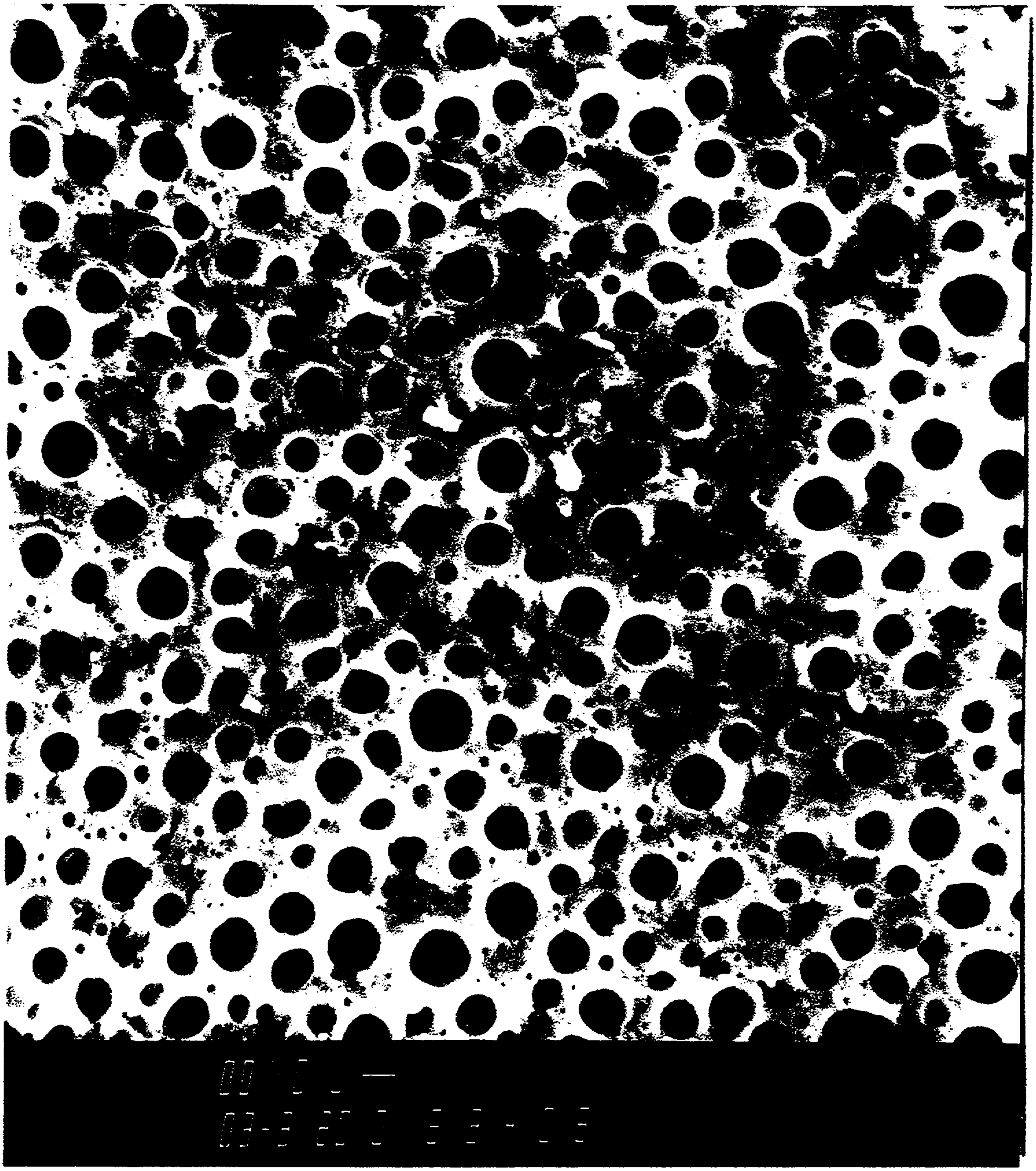


Fig. 1. Swelling of various LEU intermetallic fuel compounds as a function of accumulated fissions in fuel.


g, cm^{-3}
▲ 3.7, LEU
● 5.6, LEU
■ 3.9, MEU
○ 1.7, HEU
ΔV^F, %
100
50
0
5
10
15
FD, 10^{21} cm^{-3} (FUEL)



FUEL TYPE/FISSION RATE
■ LEU/2 x $10^{20} m^{-3} s^{-1}$
● HEU/7 x $10^{20} m^{-3} s^{-1}$
△ MEU/4 x $10^{20} m^{-3} s^{-1}$
B
B
B
B
$\Delta V^F/V_o^F$, %
0
20
40
60
80
100
0
2
4
6
8
10
12
14
16
FISSION DENSITY IN FUEL PHASE, $10^{27} m^{-3}$



Bubble morphology in HEU, U_3Si_2@ ~70% Bu, ORR (SEM)

SWELLING BEHAVIOR DURING IRRADIATION (CONT'D)

- Further investigation revealed two problems:
  - Neutron diffraction studies of fuel irradiated in the IPNS at ANL indicated that both U_3Si and U_3Si_2 became amorphous at very low doses.
  - Our swelling model could not reproduce the bubble distribution seen in U_3Si_2.

SWELLING BEHAVIOR DURING IRRADIATION (CONT'D)

- The solution was to postulate the formation of subgrains in U_3Si_2 at the fission density of the knee of the swelling curve. These could be formed by subdivision of grains of crystalline material or by recrystallization of amorphized material.
  - ○ Subgrains had been found in UO_2 and U_4O_9, with similarities to U_3Si_2 in the fission gas bubble morphology.
  - ○ Ion simulation experiments gave evidence of grain refinement.
  - ○ We have now found evidence of subgrains in U_3Si_2 irradiated in the HFIR.

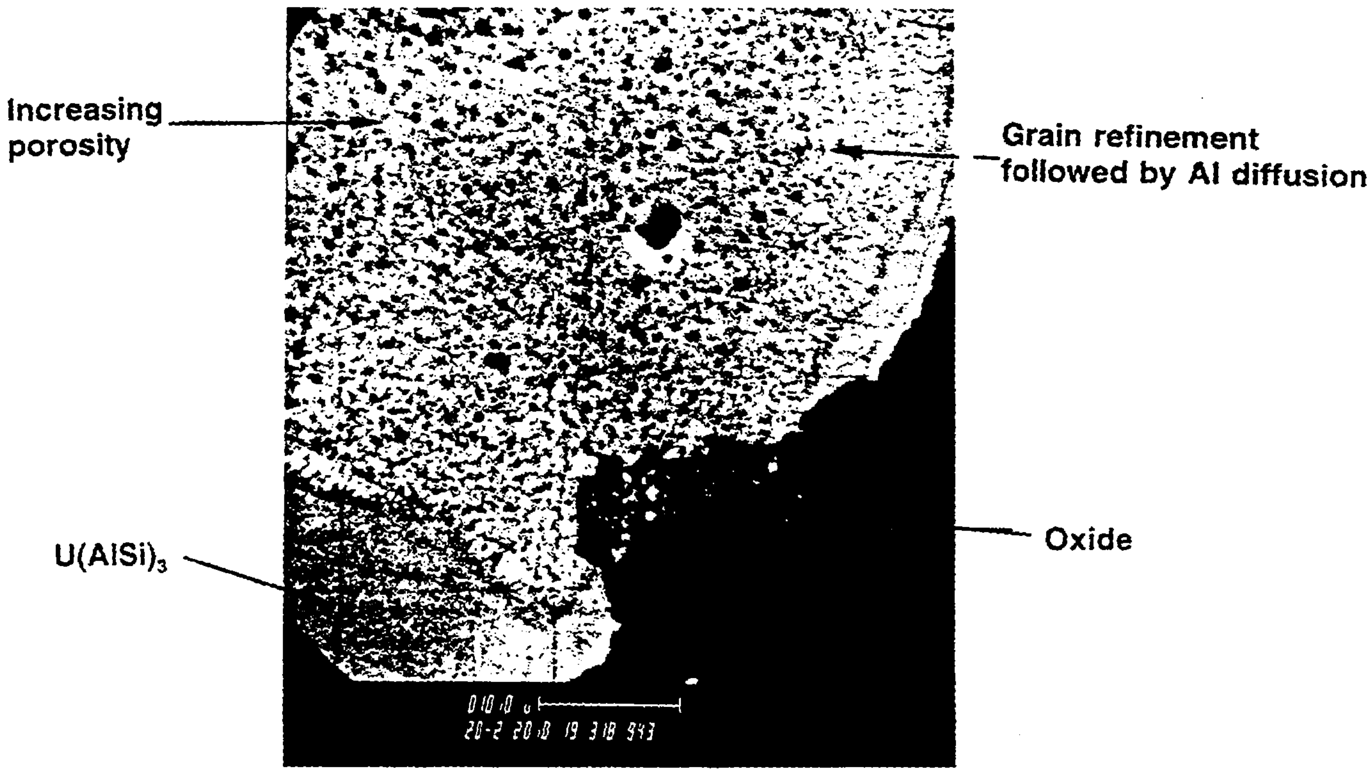
SWELLING BEHAVIOR DURING IRRADIATION (CONT'D)

- ANS irradiation conditions of temperature, fission rate, and fission density were well beyond those in the ORR irradiations.
- Examination of samples irradiated in the HFIR target region has provided further insights into the behavior of U_3Si_2.
- We postulate a multi-stage process:
  - The outer portion of the U_3Si_2 particle reacts with Al to form a compound with a UAl_3-type structure, $U(SiAl)_3$. This material has the high stability characteristic of UAl_3; no fission gas bubbles are seen.

SWELLING BEHAVIOR DURING IRRADIATION (CONT'D)

- ○ Inside this zone subgrains form in the unreacted U_3Si_2 when the fission density reaches the knee of the swelling curve, and typical swelling ensues.
- ○ The center of many fuel particles, however, show bubbles characteristic of the unstable, amorphous materials. We postulate that depletion of the U has led to the formation of an unstable phase in high-burnup HEU fuel particles. We think that this phase may be the peritectoid USi_2, analogous to U_3Si. Break-away swelling is restrained, however, by the outer $U(SiAl)_3$ shell.

Optical **500X**



SEM **2000X**

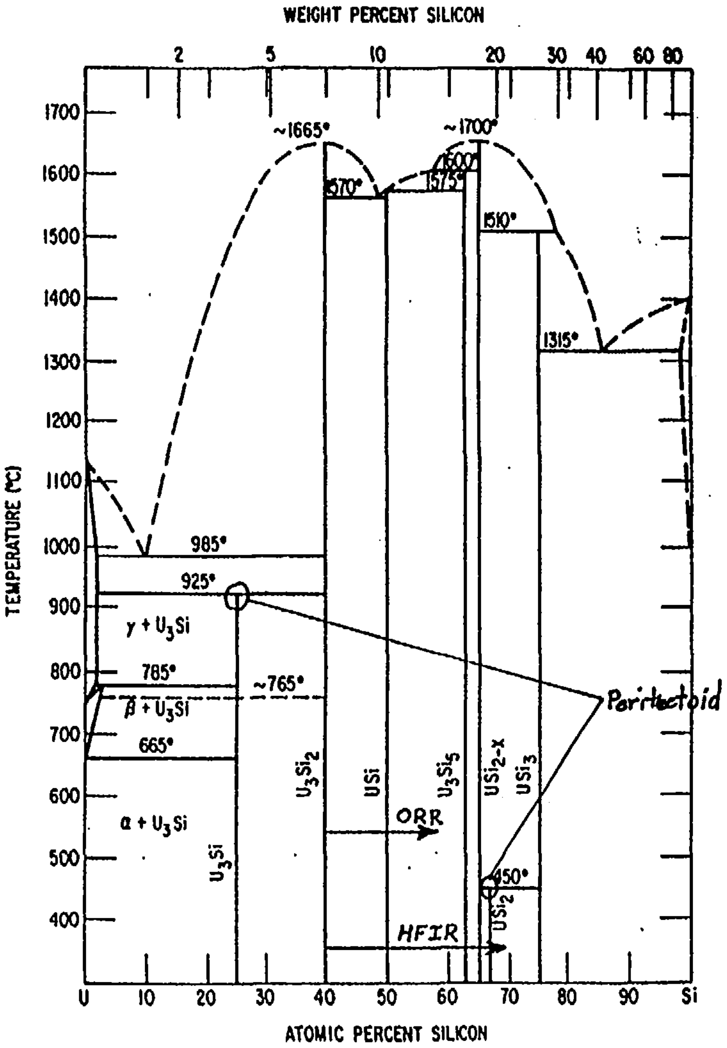
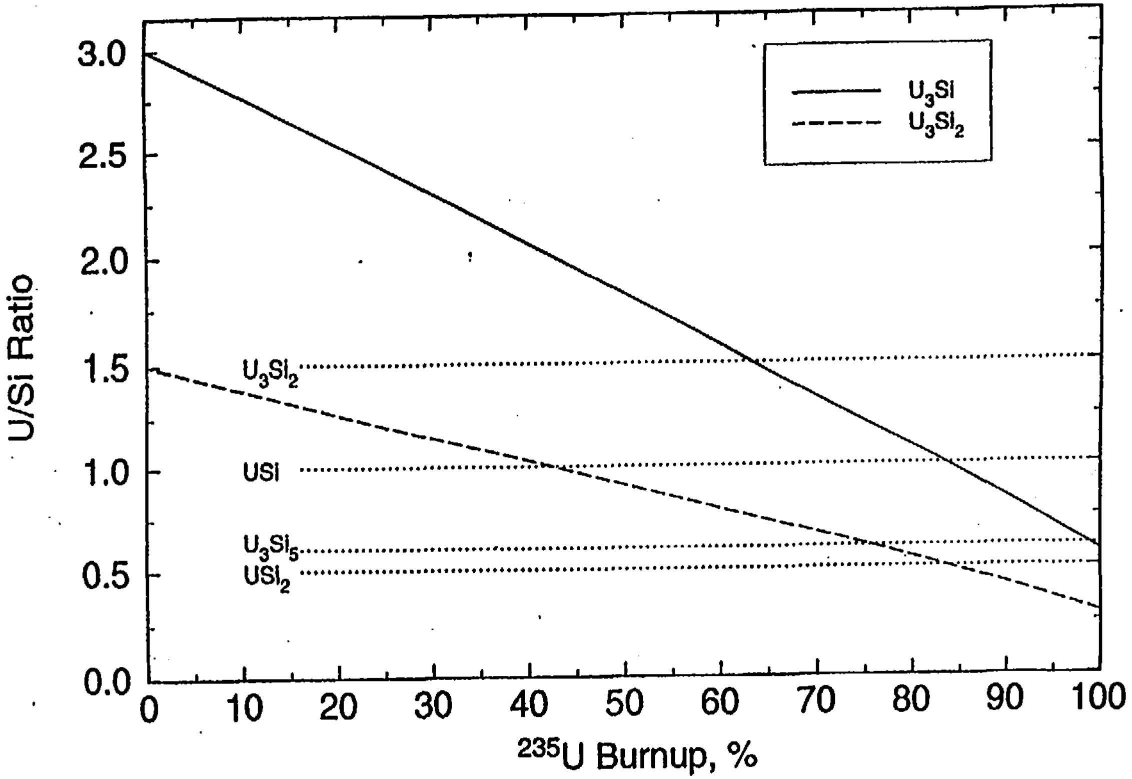
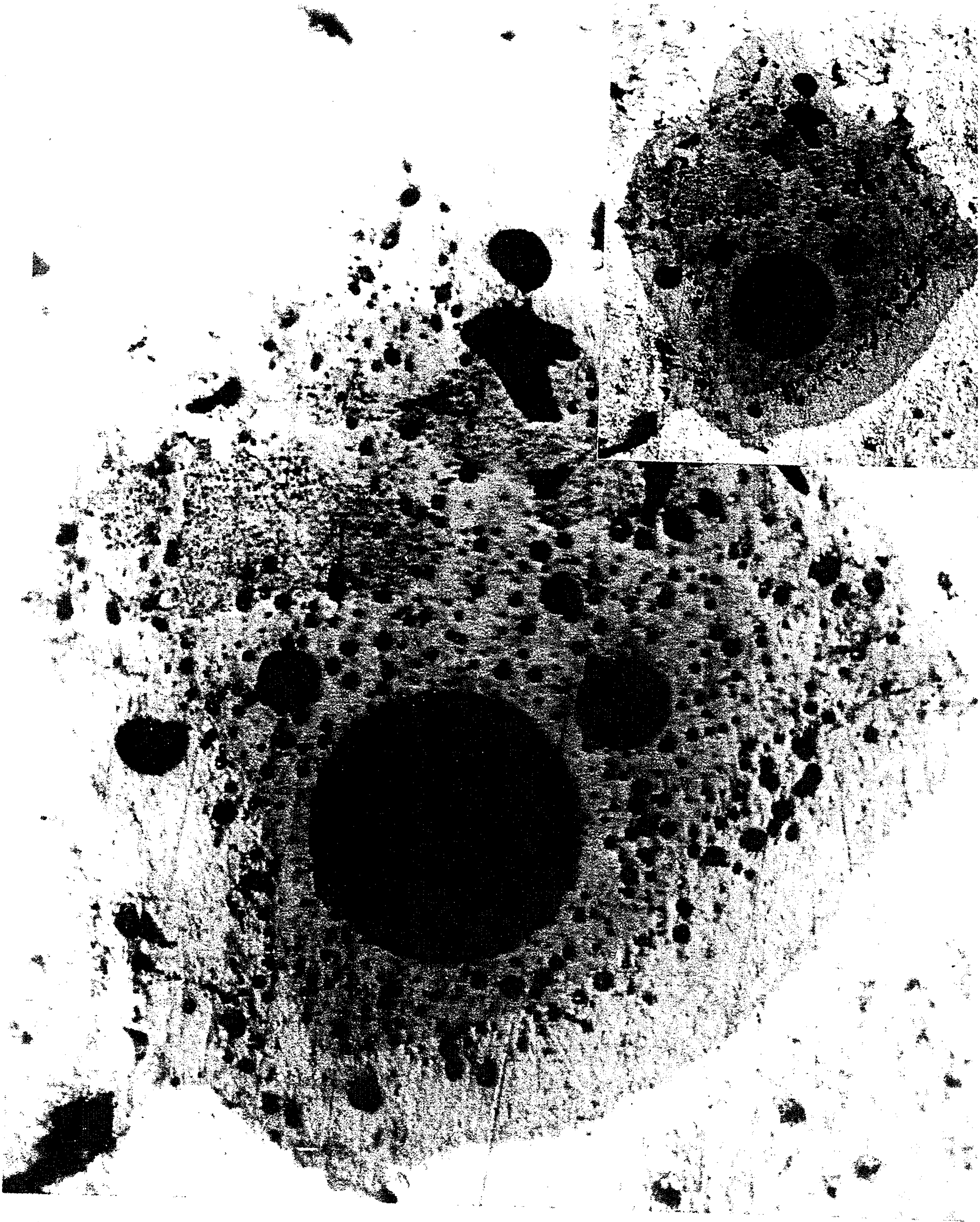


FIG. 4. CHANGE IN U/Si RATIO VS. ^{235}U BURNUP AS 93%-ENRICHED U_3Si AND U_3Si_2 ARE BURNED





Bubble morphology and Al interaction @ ~85% Bu, 250°C, HFIR

THERMAL CONDUCTIVITY CALCULATIONS

THERMAL CONDUCTIVITY MODEL

- Model developed and implemented in the Dispersion Analysis Research Tool (DART) code by J. Rest, ANL.
- Thermal conductivity is a function of fuel and porosity volume fractions, which are calculated based on a mechanistic model of fuel particle swelling.
    - Fuel particle swelling model has evolved. Calculations now based on multiphase model of fuel swelling which fits both ORR and HFIR data.

THERMAL CONDUCTIVITY MODEL (CONTINUED)

Model Basis

- Spherical fuel particles are mixed into matrix aluminum.
- As-fabricated porosity is mixed into fuel meat.
- Irradiation-induced porosity is mixed into fuel particles.
- Aluminum matrix is assumed to be the continuous medium.
- Successive application of classical mixing formula gives equation for thermal conductivity as a function of fuel and porosity fractions.
- Fuel meat volume is assumed to remain constant as fuel particles swell--porosity is first closed then aluminum removed from meat.

THERMAL CONDUCTIVITY MODEL (CONTINUED)

Thermal Conductivity Equation

$$k_e^m = k_{al}\left[z_1 + z_2 F_v^{2/3} + z_3\left(k_e^g / k_{al}\right) F_v^{1/3}\left(1 - F_p^{2/3}\right) + z_4 F_p^{2/3} + z_5\left(F_v F_p\right)^{2/3} \right]$$

where

k_e^m, k_{al}, and k_e^g are the thermal conductivity of the fuel meat, matrix aluminum, and U_3Si_2 particles containing irradiation-induced porosity;

F_v and F_p are the fuel and as-fabricated porosity volume fractions;

and

z_1 - z_5 are geometric constants.

THERMAL CONDUCTIVITY MODEL (CONTINUED)

- The physical limits:
  - $k_e^m = k_{al}$ for $F_p = 0, F_v = 0$
  - $k_{em} = 0$ for $F_p = 1$
  - $k_{em} = k_{al}$ for $F_v = 1, k_{eg} = k_{al}$
  - $k_{em} = 0$ for $F_v = 1, k_{eg} = 0$

  imply that $z_1 = +1$, $z_2 = -1$, $z_3 = +1$, $z_4 = -1$

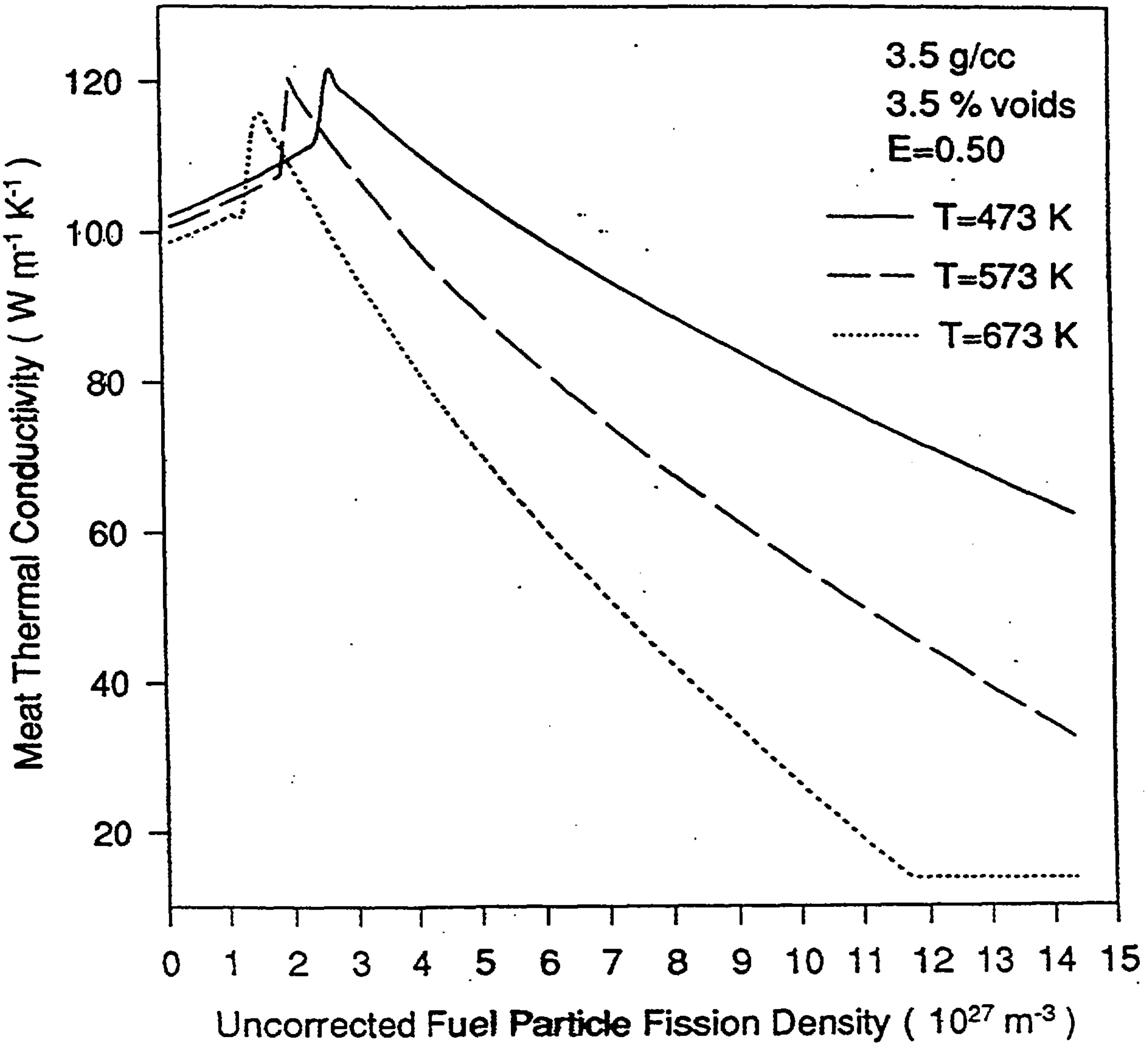
- z_5 is determined by fit to thermal conductivity data for unirradiated fuel to correct for idealized geometry assumption:
  - $z_5 = -0.3275$

THERMAL CONDUCTIVITY MODEL (CONTINUED)

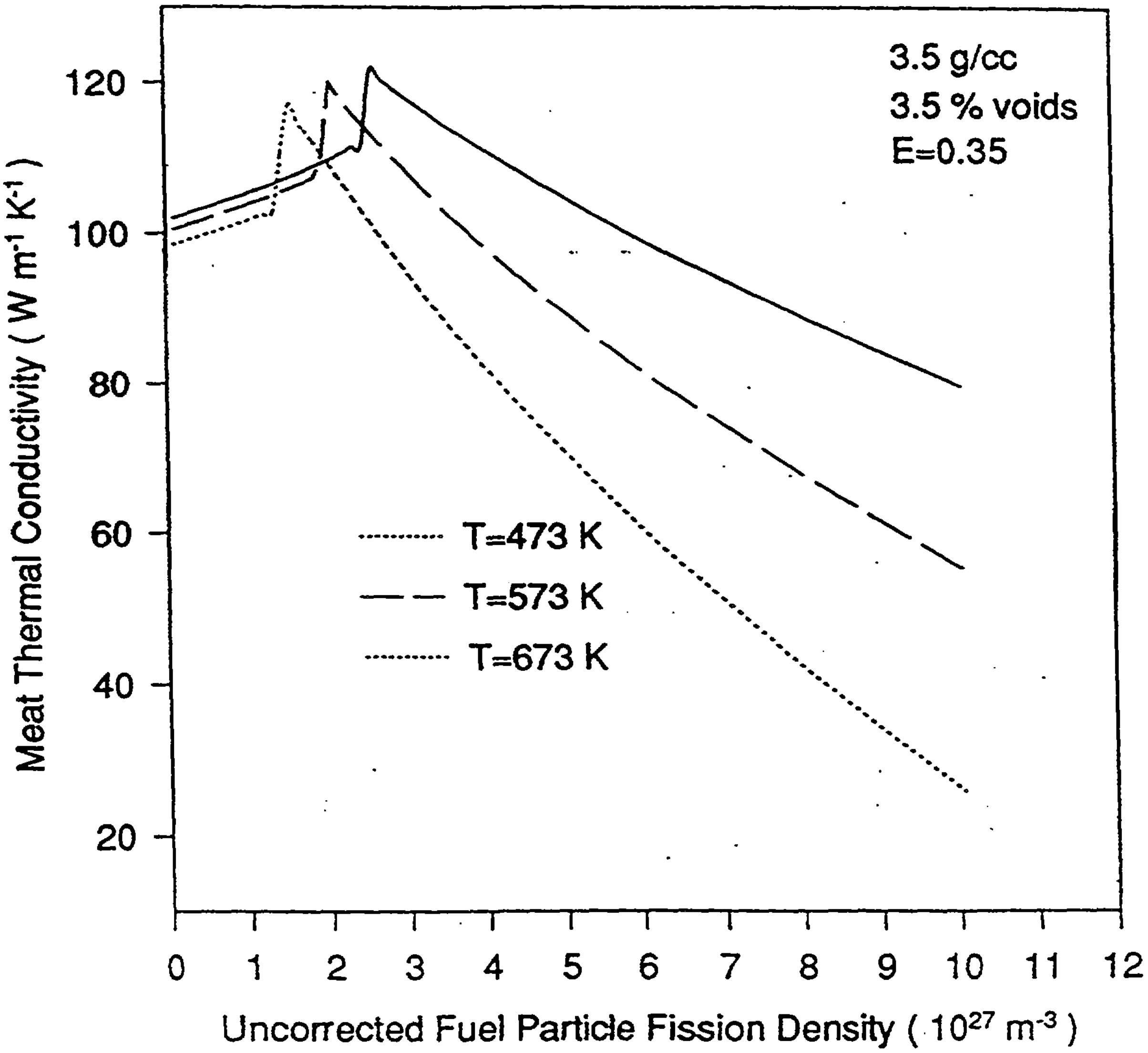
Model Limitations

- Treats only single size of fuel particle.
- Invalid when aluminum matrix is discontinuous, i.e., for less than 35 to 40 vol% aluminum.
- Does not account for any degradation of aluminum thermal conductivity due to irradiation.
- Accuracy of the results depends on the accuracy of the thermal conductivity and fuel swelling models and data. Fuel swelling depends on fission rate, total fissions, and temperature.

MULTINODE DART CALCULATIONS



MULTINODE DART CALCULATIONS



MULTINODE DART CALCULATIONS