DEVELOPMENT OF U_6Fe-Al DISPERSIONS FOR THE USE OF LEU IN RESEARCH AND TEST REACTORS

S. Nazaré
Institut für Material- und Festkörperforschung
Kernforschungszentrum Karlsruhe
Postfach 3640
D-7500 Karlsruhe
Federal Republic of Germany

Introduction

For some time now, efforts are being made to develop fuel dispersions that would permit the use of low (∿ 20% 235-U) enriched uranium (LEU) instead of the currently used highly (∿ 93% 235-U) enriched uranium (HEU) in research and test reactors. Since penalties in the performance of the reactor have to be avoided, the 235-U content in the dispersion has at least to be retained at current levels. On account of their high U-densities, the major development effort has been focussed on the uranium silicides [U_3Si, $U_3Si(Al)$, and U_3Si_2-based dispersions [1,2,3]. With silicides as dispersants, it is possible to fabricate fuel element plates with U-densities in the dispersion of about 6.0 gU/cm^3.

In comparison to the silicides, the U_6Fe-phase offers several advantages namely:

- higher U-density (∿ 17.0 gU/cm^3);
- relative ease of formation compared to U_3Si;
- possible advantages with regard to reprocessing of the spent fuel due to the absence of silicon.

The studies outlined here were therefore performed with a view to investigating the preparation, reaction behaviour and dimensional stability after heat treatment of U_6Fe-Al dispersions.

Preparation of U_6Fe

The U-Fe phase diagram [4] shown in fig.1 reveals the occurence of two intermetallic compounds namely U_6Fe and UFe_2. The compound U_6Fe, which is formed by peritectic reaction at 1083 K has a tetragonal structure (a = 1028.9 pm, c = 523.2 pm).

Induction melting in an argon atmosphere (52 kPa) with alumina crucibles was used to prepare the compound from the elements. A slightly hyperstoichiometric composition (4.0 wt.% Fe) was chosen. The material was homogenized at 973 K for 24 hours in a vacuum using additionaly a zirconium getter.

A typical microstructure of the material thus obtained is shown in fig.2. It reveals essentially two phases U_6Fe being the matrix and UFe_2 the white inclusions.

Powder metallurgical processing of the U_6Fe-phase

For the preparation of the miniature fuel element plates it is necessary to comminute the molten material to a desired particle size. In the commercial fabrication of standard UAl_x and U_3O_8-Al dispersion fuel plates, it is common practice to use particle sizes for the dispersed phase in the range of 44-150 µm (74 wt.%) allowing only a limited quantity (25 wt.%) of finer powder (<44 µm). This requirement arose in the past from the concept, that under irradiation, the zones of fission fragment recoil should not overlap [5,6], so that a continuous undamaged metallic matrix is preserved for the dissipation and transport of fission heat. In addition, the ductility and thickness of the ductile matrix rim around the fissionable particles is thought to contribute towards the prevention of crack initiation and propagation from stresses that develop due to particle swelling. Although there is evidence [7] that at low volume contents of the dispersed phase (HEU, e.g. 38.8 vol.% UAl_x) the stability and swelling are not influenced by the particle size (size fraction <44 µm) but primarily by the porosity in the particles and in the dispersion, the specifications have been retained because of the proven stability of fuels with these characteristics. It should be mentioned, that in thermodynamically unstable dispersions, the reaction kinetics could be accelerated by using a large amount of fine powder. In addition the thermal conductivity of the dispersion might be decreased when the particle size is reduced, specially at high contents of the dispersed phase.

For the experiments outlined here a narrow particle size range of 63-90 µm was selected. Although this particle size range may neither be important, desirable nor economical in commercial fabricated fuel elements, it was retained with a view to keeping possible surface and interfacial phenomena unchanged and to compare the behaviour of the dispersion with that of the silicides.

A jaw crusher was used in the first step of the communition which was carried out in an argon filled glove box, followed by grinding in a WC-Co lined "shatterbox", with intermittent sieving. The communition behaviour of the U_6Fe-phase seems to be comparable to that of the U_3Si-phase. The chemical composition and the powder characteristics of the powder obtained along with those of the Al-matrix powder used are shown in tab.1.

Further processing of the powders to produce miniature Al-clad fuel plates (plate dimensions 220 • 40 • 1.3 mm^3; nominal meat dimensions 200 • 30 • 0.5 mm^3) was carried out using the well known state of the art picture frame technique. The microstructures of the fuel plates obtained with U-densities of 4.0, 6.0 and 7.0 gU/cm^3 are shown in fig.3. The homogeneity in the fuel meat and the dimensional tolerances are within the limits currently acceptable.

Compatibility of U_6Fe with the Al-matrix

The objectives of the investigations outlined here were primarily:

- to extend the present knowledge of the ternary U-Fe-Al system [8-10] to those regions that are of relevance for the fabrication and in-pile behaviour of the dispersions;
- to investigate the reaction behaviour of roll bonded dispersions particularly with regard to the dimensional changes that occur in the fuel plate as a consequence of the reaction.

Equilibrium investigations

The samples for the investigations of the reaction behaviour under equilibrium conditions were prepared by arc melting of the elements in an argon atmosphere (0.5 MPa). The initial compositions, were chosen to correspond to 20, 30, 40 and 50 vol.% U_6Fe-Al. Fig.4 shows the microstructure of the U_6Fe-Al samples in the as-cast state as well as after heat treatment at 873 K ($\sim$ 33 days). After heat treatment the sample corresponding to 20 vol.% U_6Fe-Al reveals three phases UAl_3, UAl_4 (with Fe in solution) and Al (fig.4b). The sample corresponding to 30 vol.% U_6Fe-Al (fig.4d) reveals practically a single phase of UAl_3(Fe). The sample with 40 vol.% U_6Fe-Al reveals two phases namely UAl_3(Fe) and UAl_2(Fe) and finally the sample with 50 vol.% reveals again two major phases UAl_2(Fe) and probably U_6Fe(Al). Although this limited data indicates that uranium aluminides are the main products of the reaction, additional experiments are necessary to determine the phase regions in the ternary sytem.

It must be mentioned in this context, that probably equilibrium conditions will not be generally attained during the normal operation conditions and life of the fuel. However, it is well known, that the diffusion reactions are enhanced in a neutron environment in UO_2-Al, UAl_x-Al and U_3O_8-Al fuels. Smaller particle sizes of the dispersed phase and surface energy effects could also significantly increase the reaction rate assisting the attainment of equilibrium. The phases formed under equilibrium conditions could be important during a LOCA.

Influence of the reaction of the dimensional stability of roll bonded miniature plates

In order to investigate the reaction behaviour of the U_6Fe-Al dispersions, small miniature plates with a pure Al-cladding were fabricated using standard procedures. Cylindrical pellets (10 mm Ø, 2.5 mm high) with 23 vol.% U_6Fe-Al were used. This corresponds to an U-density of 4.0 gU/cm^3 in the meat. The powders were degassed at 873 K in a vacuum <1 mPa for 4 hours. The particle size was within the sieve range of 63-90 microns. The miniplate dimensions were, 70•15•1.3 mm^3, the nominal meat-thickness was 0.5 mm; the nominal cladding thickness being 0.4 mm. The samples were heat treated in evacuated pyrex tubes for times up to about 2000 hours. Selected photographs of the samples after heat treatment are shown in fig.5. Apart from providing a qualitative picture of the volume increases, they reveal that the volume increase occurs only in the meat region of the fuel plate. Dimensional measurements carried out have shown that the plates are dimensionally stable up to temperatures of 423 K even after an anneal of 2000 hours. This behaviour

compares favourably with the one observed with the silicides. However, at 723 K large volume increases are observed due to reaction.

Standard X-ray diffraction procedures (Guinier), metallography and energy dispersive X-ray analysis were used to identify the reaction products. After the 2000 hours anneal, the major reaction product identified was UAl_3 probably with Fe in solution. It seems, that from the reaction kinetics viewpoint, the UAl_3 phase shows preferred growth over the other possible phases. Metallographic analysis was carried out on all annealed specimens. Vacuum infiltration was used to prevent breakout of the material during the standard grinding and polishing operations. Fig.6 is a set of micrographs that shows the microstructure after heat treatment. Practically no reaction is evident after heat treatment (2000 hours) at 523 K and 623 K (figs.6b and 6c). The main reaction product (UAl_3(Fe)) after heat treatment at 723 K appears white.

It is interesting to note the large voids that are formed around the reacting particles. These could be an indication of a Kirkendall type of porosity which arises due to large differences in the partial diffusion coefficients of the reacting species. This type of porosity could contribute to the large swelling that occurs in the specimens which is much greather than the one predicted by merely considering the differences in the specific volumes of the theoretically dense reaction products and reactants. Similar volume increases have also been observed in the past with UO_2-Al dispersions.

It is possible that gases that are released as a result of the reaction contribute towards the large swelling observed when the internal pressure causes creep in the Al-matrix and cladding. In such a case, the swelling observed would be dependent on the high temperature strength of the cladding.

Summary and conclusions

From the fabrication and the reaction behaviour outlined here, U_6Fe dispersions could be an interesting alternative to the silicides that are considered to be the prime candidates for high U-density LEU dispersion fuels. The high U-density of the compound should allow fabrication of fuel plates with densities up to 7.0 gU/cm^3. A satisfactory irradiation performance of the silicides now being tested would however make the U_6Fe-Al combination an interesting but academic alternative.

References

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Chemical composition (wt.%)	U_6Fe	Al
Al	< 0.1	99.5
C	0.07	—
Fe	3.9	0.13
H_2	—	0.011
N_2	0.01	—
O_2	0.15	0.29
Si	—	0.07
Particle size (μm)	63-90	18.6 (mean)
Particle shape ⟼ 10μm		

Table 1: Chemical composition and powder characteristics of U_6Fe and Al.

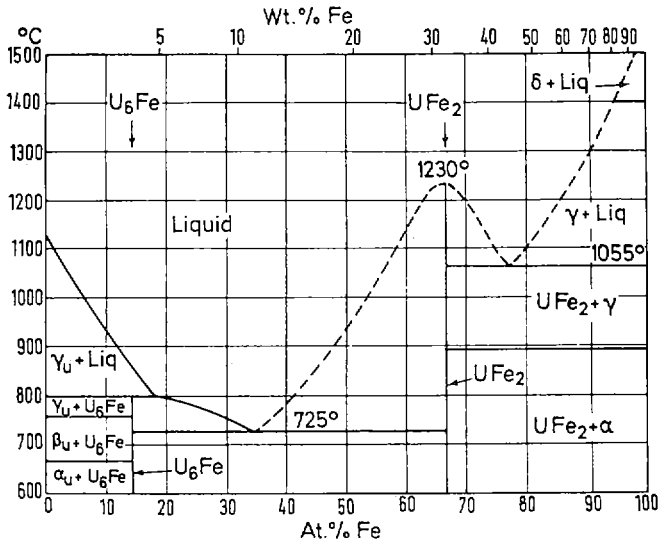


Fig.1: The U-Fe phase diagramm [4].

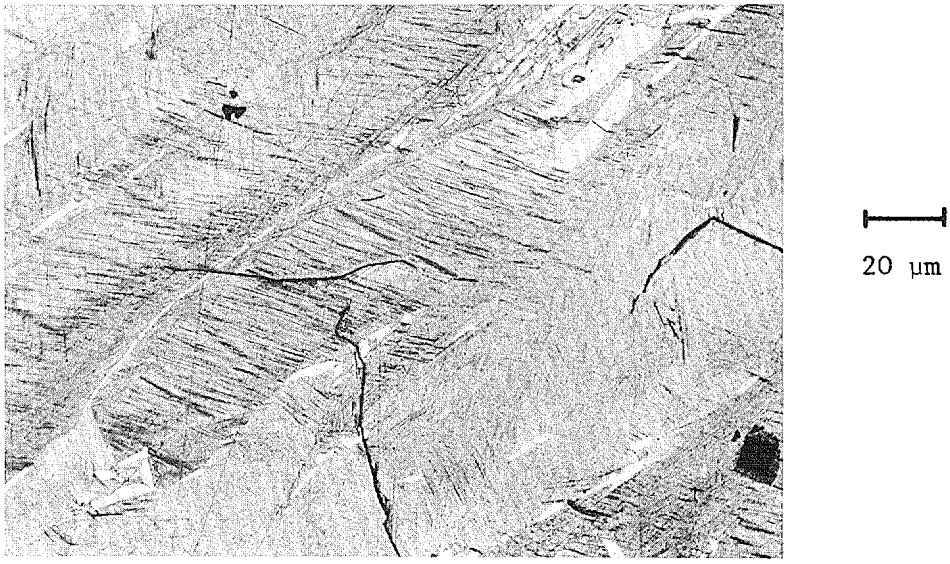


Fig.2: Microstructure of U_6Fe after heat treatment.

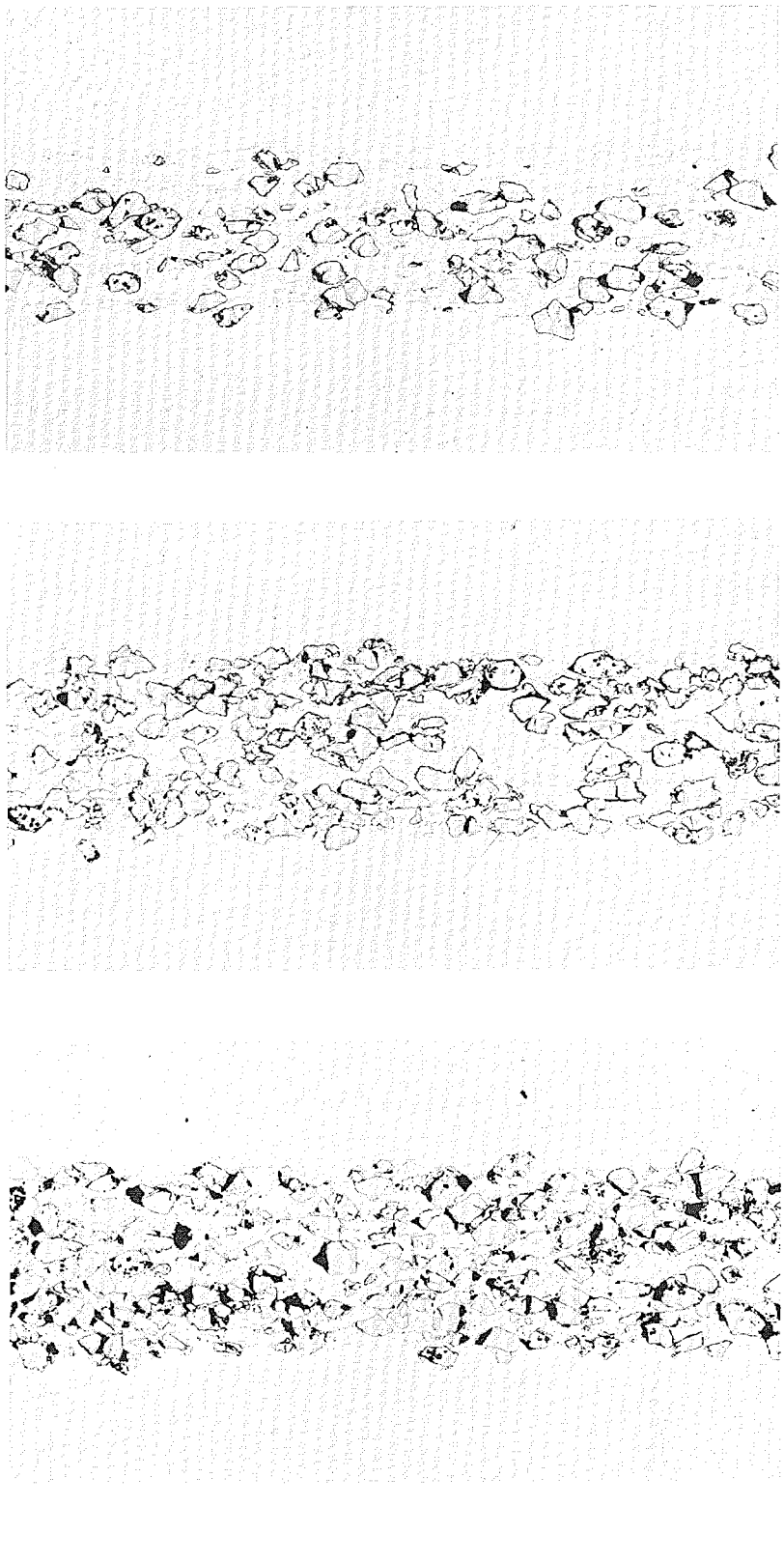


Fig.3: Microstructures of U_6Fe-Al miniature fuel plates.

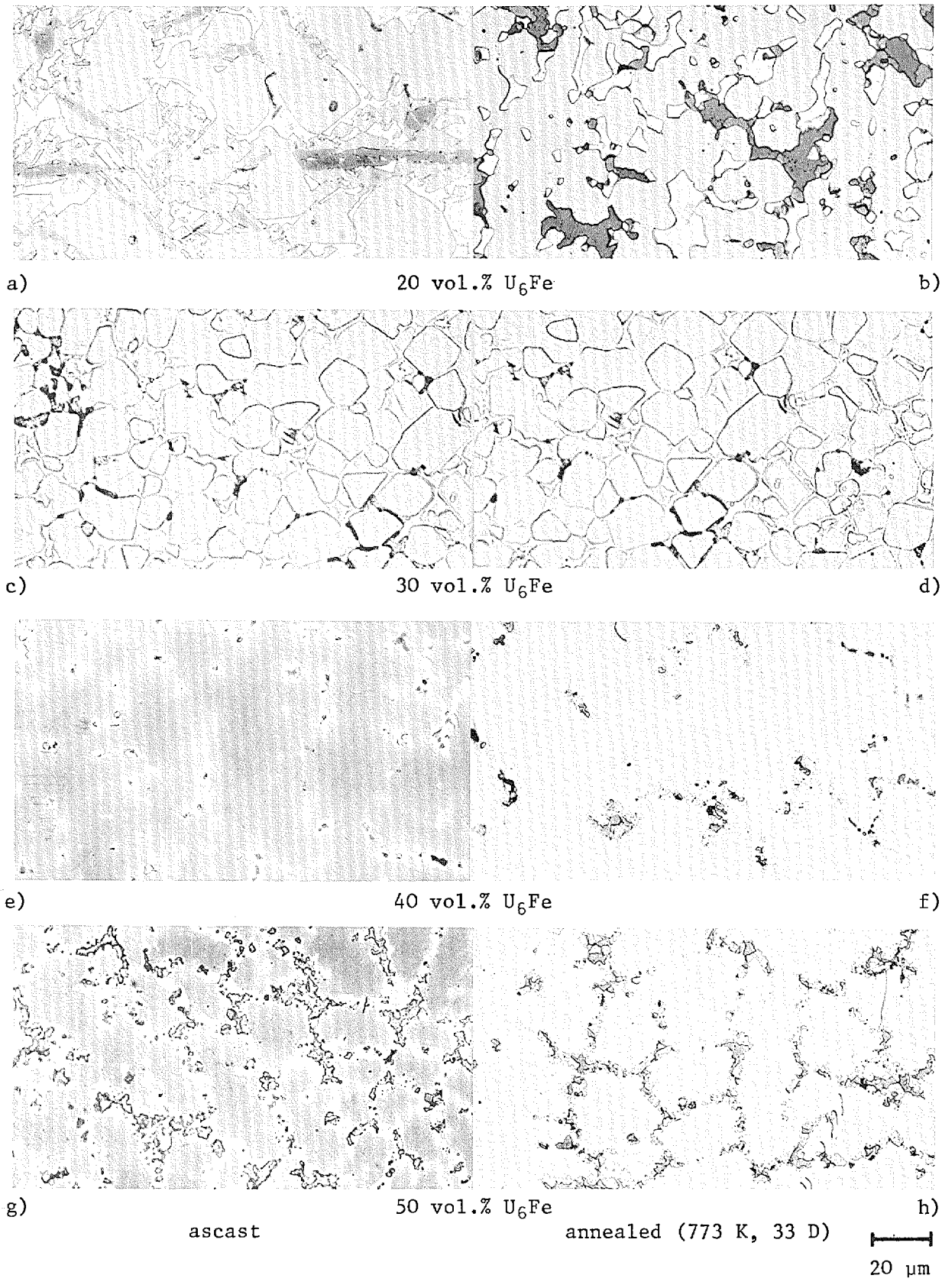


Fig.4: Reactionsin the U-Fe-Al system.

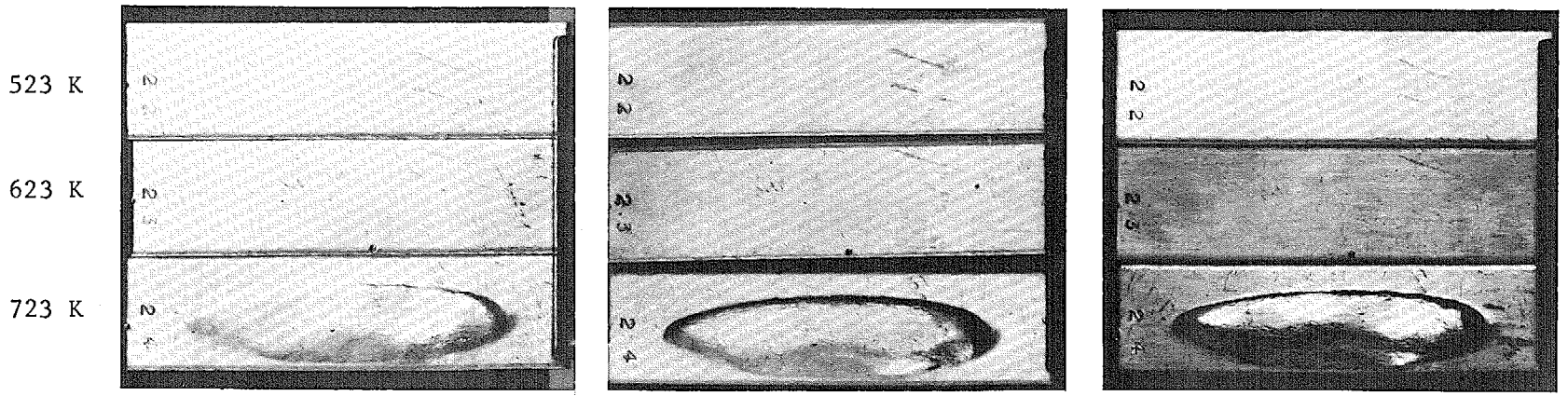
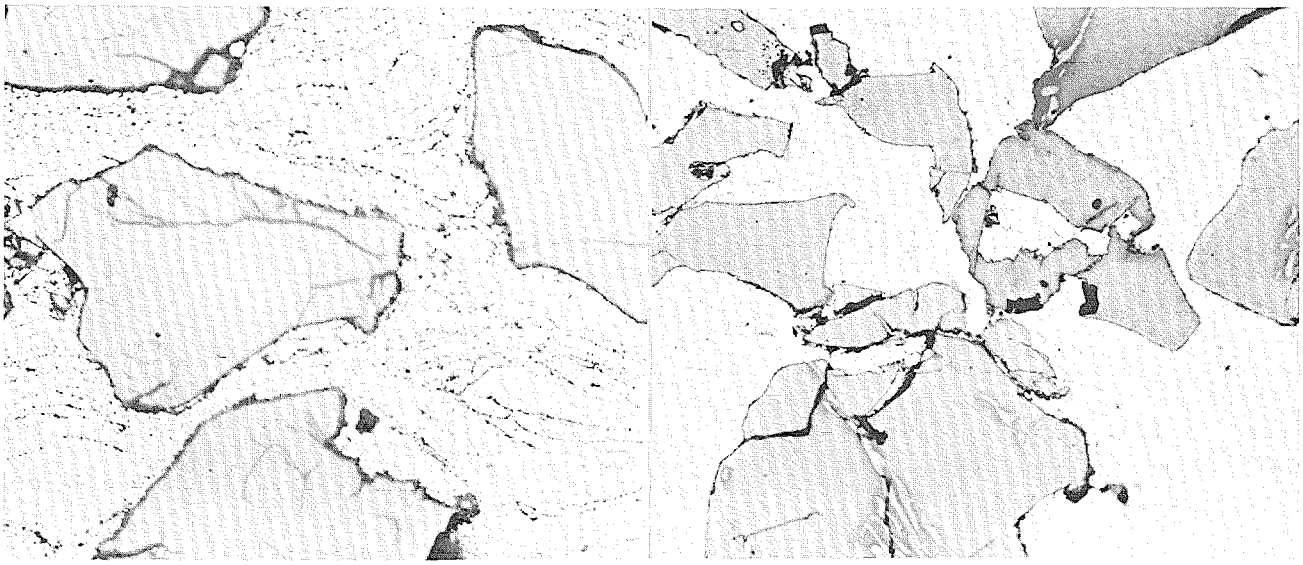
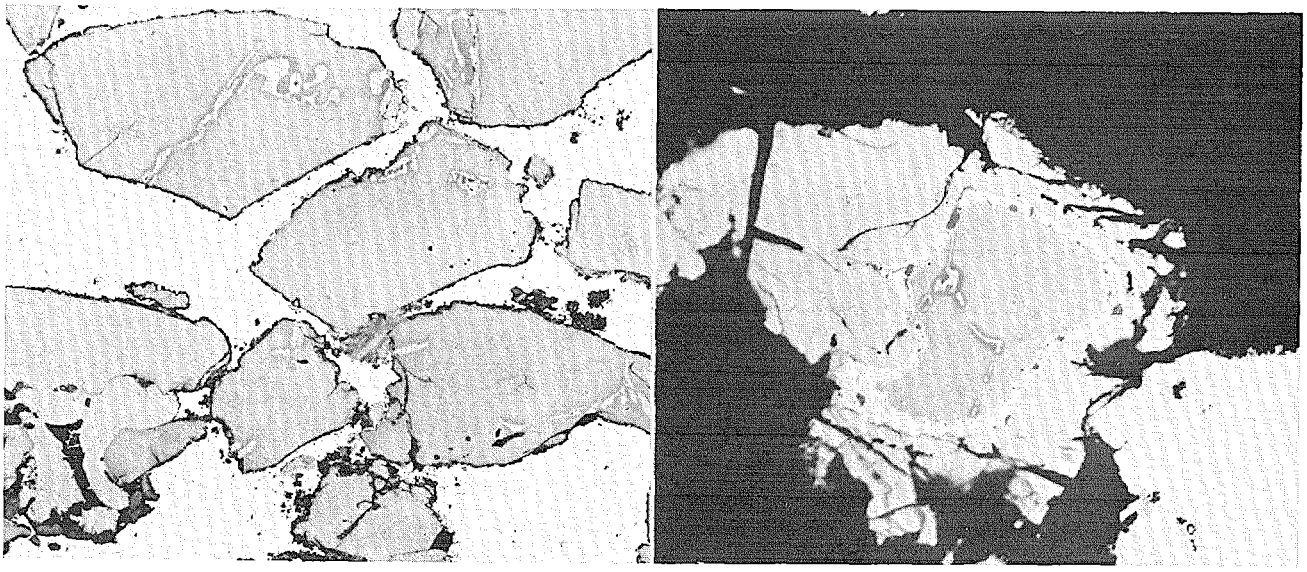


Fig.5: Swelling of U Fe-Al minitature fuel plates after heat treatment (4.0 gU cm^{-3}).



a) after rolling 523 K/2000 h b)



c) 623 K/2000 h 723 K/2000 h d)

20 µm

Fig.6: Microstructure of U_6Fe-Al fuel plates after heat treatment.