4-1 Development of high uranium-density fuels for use in research reactors

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ABSTRACT

The uranium silicide U_3Si_2 possesses uranium density 11.3 gU/cm^3 with a congruent melting point of 1665°C, and is now successfully in use as a research reactor fuel. Another uranium silicide U_3Si and U_6Me-type uranium alloys (Me = Fe,Mn,Ni) have been chosen as new fuel materials because of the higher uranium densities 14.9 and 17.0 gU/cm^3, respectively.

Experiments were carried out to fabricate miniature aluminum-dispersion plate-type and aluminum-clad disk-type fuels by using the conventional picture-frame method and a hot-pressing technique, respectively.

These included the above-mentioned new fuel materials as well as U_3Si_2. Totally 14 miniplates with uranium densities from 4.0 to 6.3 gU/cm^3 of fuel meat were prepared together with 28 disk-type fuel containing structurally-modified U_3Si, and subjected to the neutron irradiation in JMTR (Japan Materials Testing Reactor). Some results of postirradiation examinations are presented.

Keywords: Silicide, Plate-type fuel, Disk-type fuel, Preparation, Alloy, Uranium density, Uranium, Aluminum, Manganese, Iron, Nickel, Irradiation

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1. INTRODUCTION

Because of apparently no reprocessing capability in the world, the amounts of spent fuel from research reactors should preferably be reduced. Higher neutron-fluxes will also be needed for those reactors in the future. Under these circumstances, the Department of Chemistry and Fuel Research implemented an R & D program of exploring the next generation fuel in 1988. Uranium silicides U_3Si_2, U_3Si and U_6Me-type uranium alloys (Me = Fe,Mn,Ni) have been chosen as target fuel materials because of the high uranium densities 11.3, 14.9 and 17.0 gU/cm^3, respectively.

The first irradiation capsule (designated 88F-2A) containing uranium silicides was produced in November 1989; the neutron irradiation in the JMTR started in May 1990 and terminated in May 1991. The second irradiation capsule (designated 89F-1A) containing uranium silicides and U_6Me-type fuels was made in July 1990. The irradiation was started in November 1990 and continued to the end of July 1992, aiming at higher burnup levels. Postirradiation examinations of the 88F capsule were completed in May 1993 while those of the 89F are in progress at the Hot Laboratory of Oarai Establishment, JAERI. This paper briefly describes how to prepare uranium silicides and U_6Me-type fuels. Details of equipment installed, method adopted and final products were described elsewhere [1].

2. PREPARATION OF MINIATURE PLATE-TYPE FUEL

Experiments were carried out to prepare miniature aluminum-dispersion plate-type fuel (miniplate); the process is based on the conventional picture-frame method. Fuel materials produced were U_3Si_2, U_3Si, $U_3(Si,Ge)$ and U_6Me (Me=Fe,Mn, Ni). A total of 14 miniplates with uranium densities from 4.0 to 6.3 gU/cm^3 was fabricated.

Miniplates were prepared according to the following steps:

(1) Argon arc-melting of U/Me metal and silicon tips
(2) Annealing of arc-melted button at 850 °C for 72 h or at 670 °C for 10 days
(3) Crushing and powdering of arc-melted and annealed button
(4) Sieving fuel powders to 4 classes of particle sizes ($<$ 150 μm)
(5) Weighing and blending fuel powders and aluminum powders
(6) Cold pressing of mixed powders to form fuel compact
(7) Assembling fuel compact, frame and cover made with 6061 Al alloy
(8) Peripheral welding of assembly
(9) 500°C hot-rolling and cold rolling (Fig. 1a)
(10) Mechanical bond test: bending (delamination) test of a sample trimmed from the rolled plate-end (Fig. 2)
(11) Sizing (partial shearing and polishing) to finished miniplate: 20mm x 30mm x 1.3~1.4mm thick (Fig. 1b)
(12) X-ray radiographic inspection to check fuel meat homogeneity and fuel-frame bond (Fig. 1)
(13) Immersion density measurement to calculate uranium density

Steps (3)-(6) were processed in argon-circulated glove boxes in which oxygen and moisture concentrations were controlled to less than ~30 and ~50 ppm, respectively.

Fig. 3 shows a typical example of a metallographic cross-section of a 1.4-mm-thick miniplate; the fuel meat consists of U_3Si_2-Al with uranium density of 4.69 gU/cm^3 and void volume fraction of 13 %. Weight fractions of each U_3Si_2 particle-size (ps) adopted for manufacturing the fuel compact are: 19% for ps $<$ 45, 20% for 45 $\leqq$ ps $<$ 75, 21% for 75 $\leqq$ ps $<$ 106 and 40% for 106 $\leqq$ ps $<$ 150 μm.

U_6Mn and $U_6(Fe_{0.6}Mn_{0.4})$ are friable materials; powders can easily be produced from the alloy castings by the use of jaw crusher and agate mortar/pestle. This is the greater advantage of U_6Me-type fuels over the U_3Si that is too ductile to be comminuted. Fig. 4 shows EPMA analyses before and after heating a 1.38-mm-thick miniplate with $U_6(Fe_{0.6}Mn_{0.4})$ particles dispersed in Al.

Table 1 lists important parameters of miniplates fabricated. The fuel compositions of each plate are as follows:

88F-01, 89F-01 = 100 wt% U_3Si_2
88F-02, 89F-02 = 80 wt% U_3Si_2 + 20 wt% USi
88F-03, 89F-03 = 99.5 wt% U_3Si_2 + 0.5 wt% Mo
88F-04, 89F-04 = 80 wt% U_3Si_2 + 20 wt% U_3Si
89F-05 = 100 wt% U_6Mn,
89F-06 = 100 wt% $U_6(Fe_{0.6}Mn_{0.4})$

For the 89F capsule, miniplates containing the following alloys were also prepared:
U_3Si, $U_3(Si_{0.8}Ge_{0.2})$, $U_3(Si_{0.6}Ge_{0.4})$, $U_6(Fe_{0.4}Ni_{0.6})$ and U_6Ni, in which thin alloy specimens cold-pressed and embedded in the aluminum were used as the compacts for the miniplate preparation.

3. PREPARATION OF ALUMINUM-CLAD DISK-TYPE FUEL

U_3Si and U_3Si_2 + U_3Si were arc-melted and heat-treated at 850°C for 72 h. The annealed buttons were cut into a thin-plate form; each plate was then clad with two aluminum disks by hot-pressing at about 25 MPa and at 400° ~ 450 °C for 30 min in vacuum. Totally, 28 disks were fabricated and irradiated in the capsules 88F and 89F. To improve high-temperature compatibility of U_3Si and aluminum, oxidation of the surface of U_3Si plates was performed at 500° ~ 800°C under a low oxygen partial pressure in a vacuum < 0.1 Pa.

This treatment yielded the dense thin layer of UO_2 + U_3Si_2 on the surface of the U_3Si plate. The surface-oxidized U_3Si showed improvement of its compatibility with aluminum at temperatures as high as ~300°C under irradiation.

4. IRRADIATION BEHAVIOR [2]

Fuels thus prepared were irradiated in the sealed irradiation capsule in the He atmosphere. Fig. 5 shows a cross section of a U_6Mn miniplate irradiated for 216 EFPD (effective full power days) to a burnup of 53%. The surface temperature was 175 °C, which is considerably high compared to the normal irradiation temperatures of ~130 °C. U_6Me alloys did not show any excessive fuel swelling causing the plate failure even at such a relatively high irradiation temperature.

In the U_3Si disk-type fuel, the growth of fission-gas bubbles is clearly shown to be enhanced under the unrestraint condition (Fig. 6).

5. SUMMARY

Preparation experiments were carried out to fabricate miniature Al-dispersion plate-type fuel and Al-clad disk-type fuel by using the powder-metallurgical picture-frame method and hot-pressing technique, respectively. Laboratory-scale technique was established for the preparation of miniplates. Inspection showed that both miniplates and disk-type fuel were prepared to meet the requirements of specification for the reactor irradiation tests.

Uranium nitride and carbide have higher uranium density, i.e., 13.53 and 12.97 gU/cm^3, respectively. Considering its better irradiation performance, UN may represent a potentiality as a fuel for this type of reactor.

References

[1] M. Ugajin, A. Itoh, M. Akabori, " Preparation of U-Si/U-Me (Me = Fe, Ni, Mn) aluminum-dispersion plate type fuel (miniplates) for capsule irradiation ", (in Japanese): JAERI-M 93-121 (June 1993)

[2] M. Ugajin, M. Akabori, A. Itoh, K. Kawamata, Y. Tayama, Y. Nakakura, "Irradiation behavior of high uranium-density fuels": presented at the 1995 annual meeting of the Atomic Energy Society of Japan, March 28-30, 1995 (Tokyo)

Table 1 List of miniplates fabricated for the capsule irradiation in JMTR

MINIPLATE	PLATE WT. (g)	PLATE VOL. (cm^3)	TOTAL U WT. (g)	U DENSITY (gU/cm^3)	VOID (%)	FUEL VOL. (%)
88F2A-01	2.2508	0.7907	0.1348	4.38	5.94	38.76
88F2A-02	2.2748	0.8023	0.1247	4.37	5.28	39.83
88F2A-03	2.2447	0.7941	0.1125	4.66	2.82	41.67
88F2A-04	2.2669	0.7989	0.1279	4.66	7.78	39.25
89F1A-01	2.2204	0.7808	0.1408	4.07	14.37	36.00
89F1A-02	2.2518	0.7917	0.1389	4.03	11.03	36.79
89F1A-03	2.2223	0.7789	0.1454	4.15	11.42	37.17
89F1A-04	2.2641	0.7930	0.1515	4.39	13.26	37.03
89F1A-05	2.2784	0.7767	0.2105	6.31	12.84	36.79
89F1A-06	2.3152	0.7929	0.2057	5.98	15.59	34.88

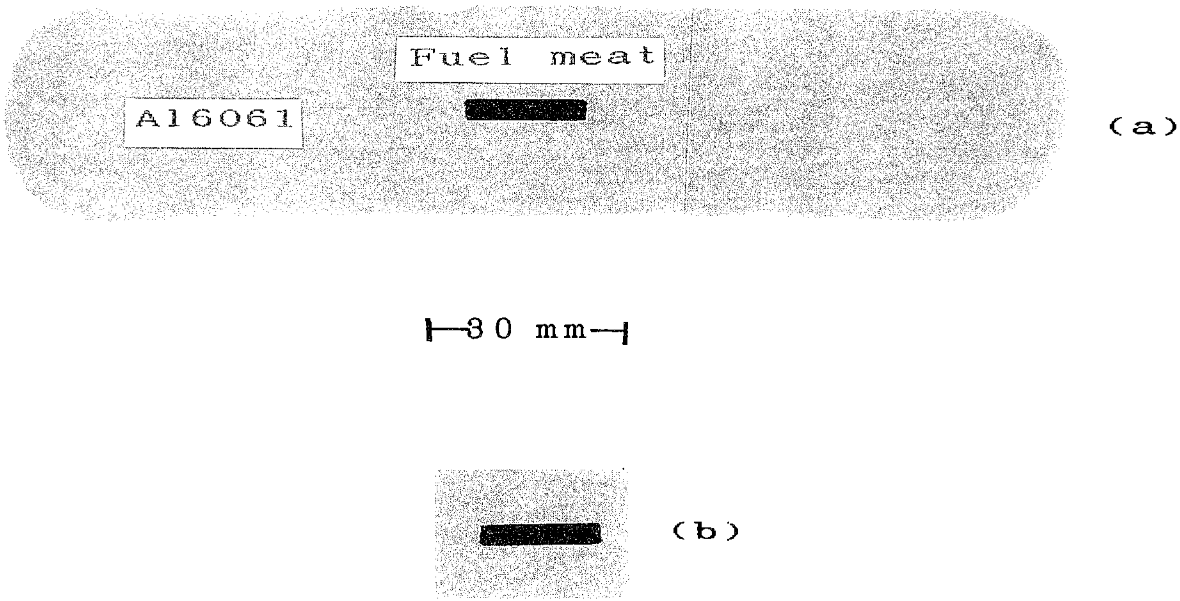

Fig. 1 X-ray radiography of welded and rolled assembly
(a) as-rolled
(b) peripherally sheared and polished to a finished miniplate

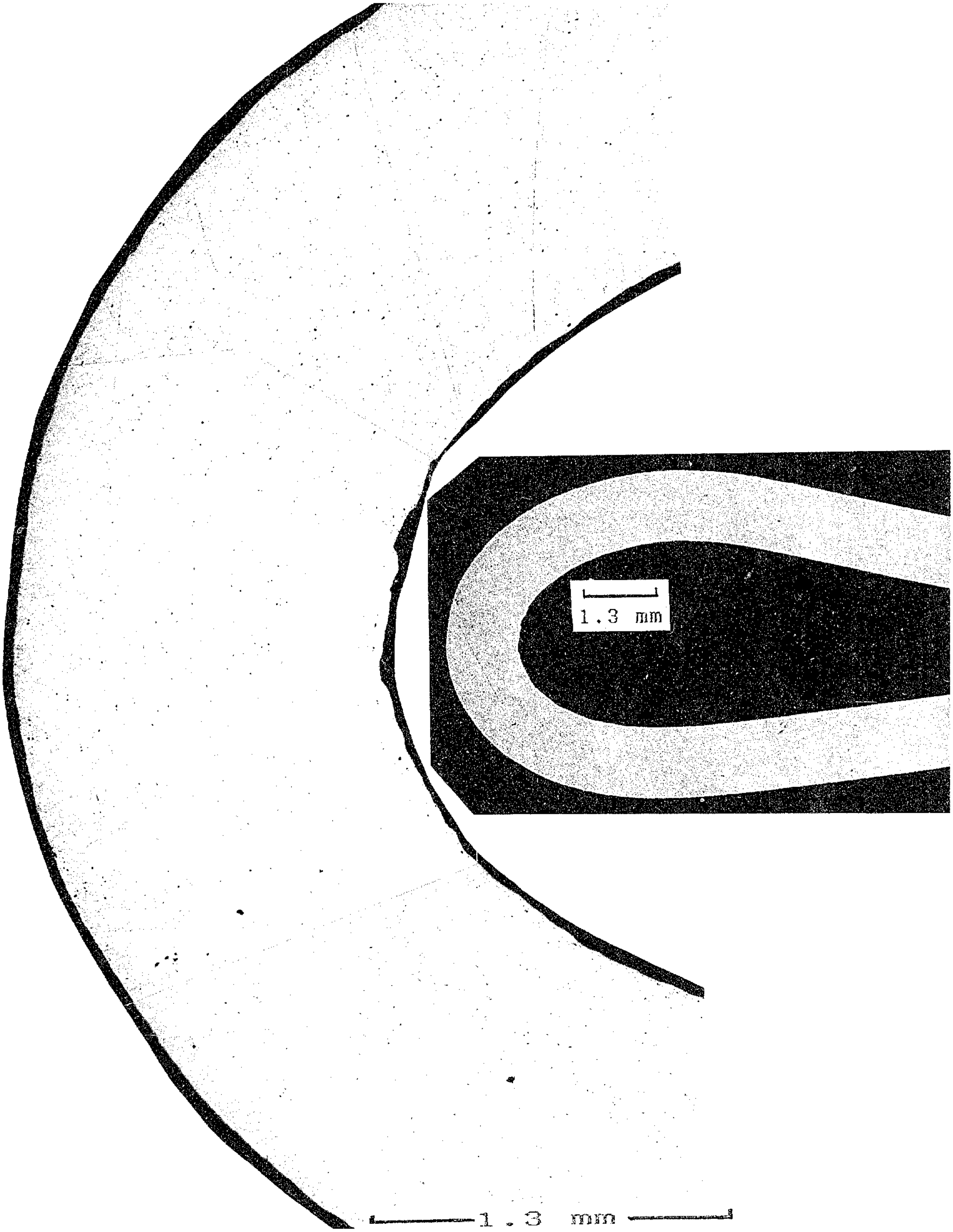

Fig. 2 Delamination test of a sample trimmed from the rolled plate-end

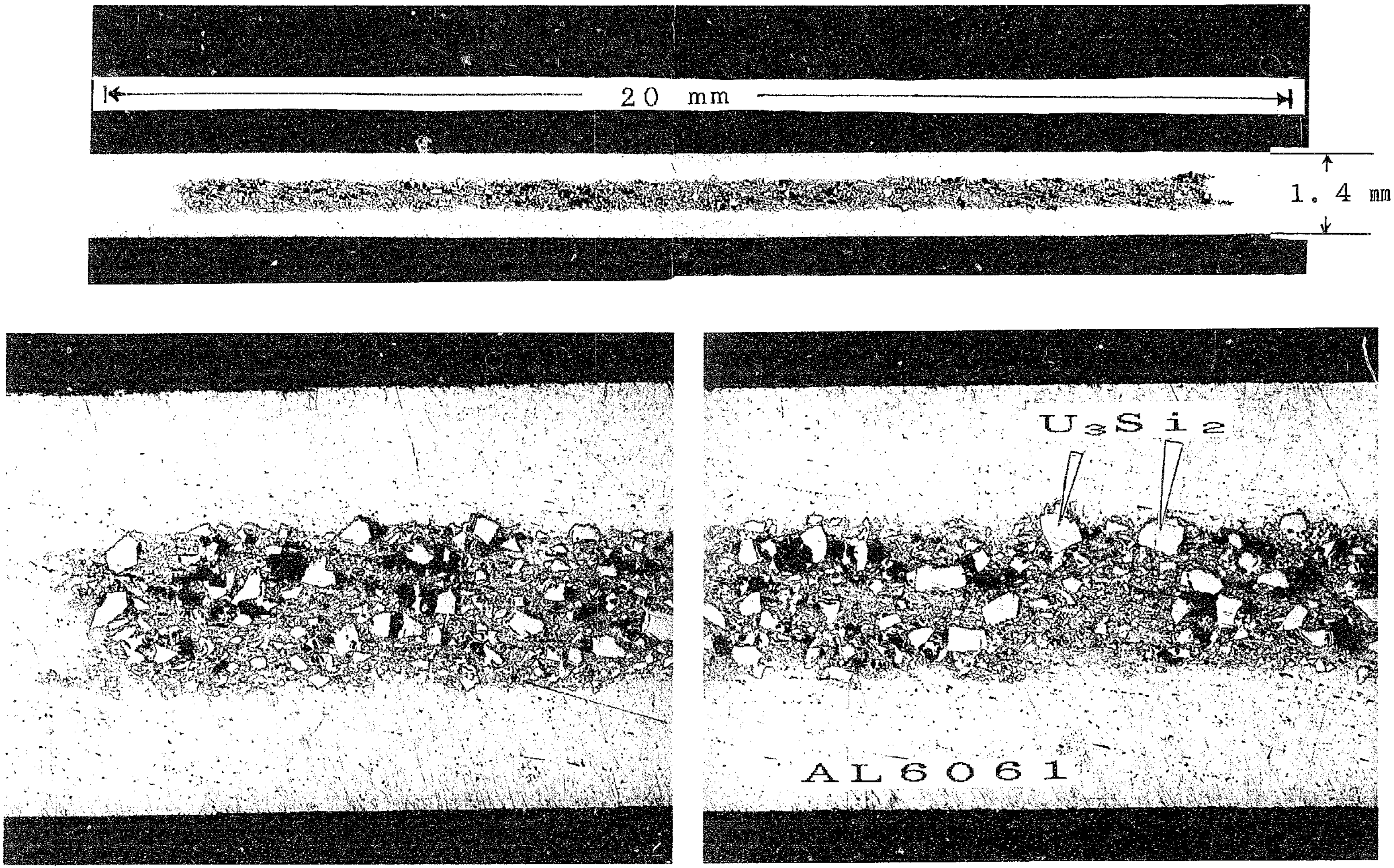

Fig. 3 Metallographic cross-section (rolling direction) of 1.40-mm-thick miniplate with fuel meat of U_3Si_2 and Al, clad with Al6061

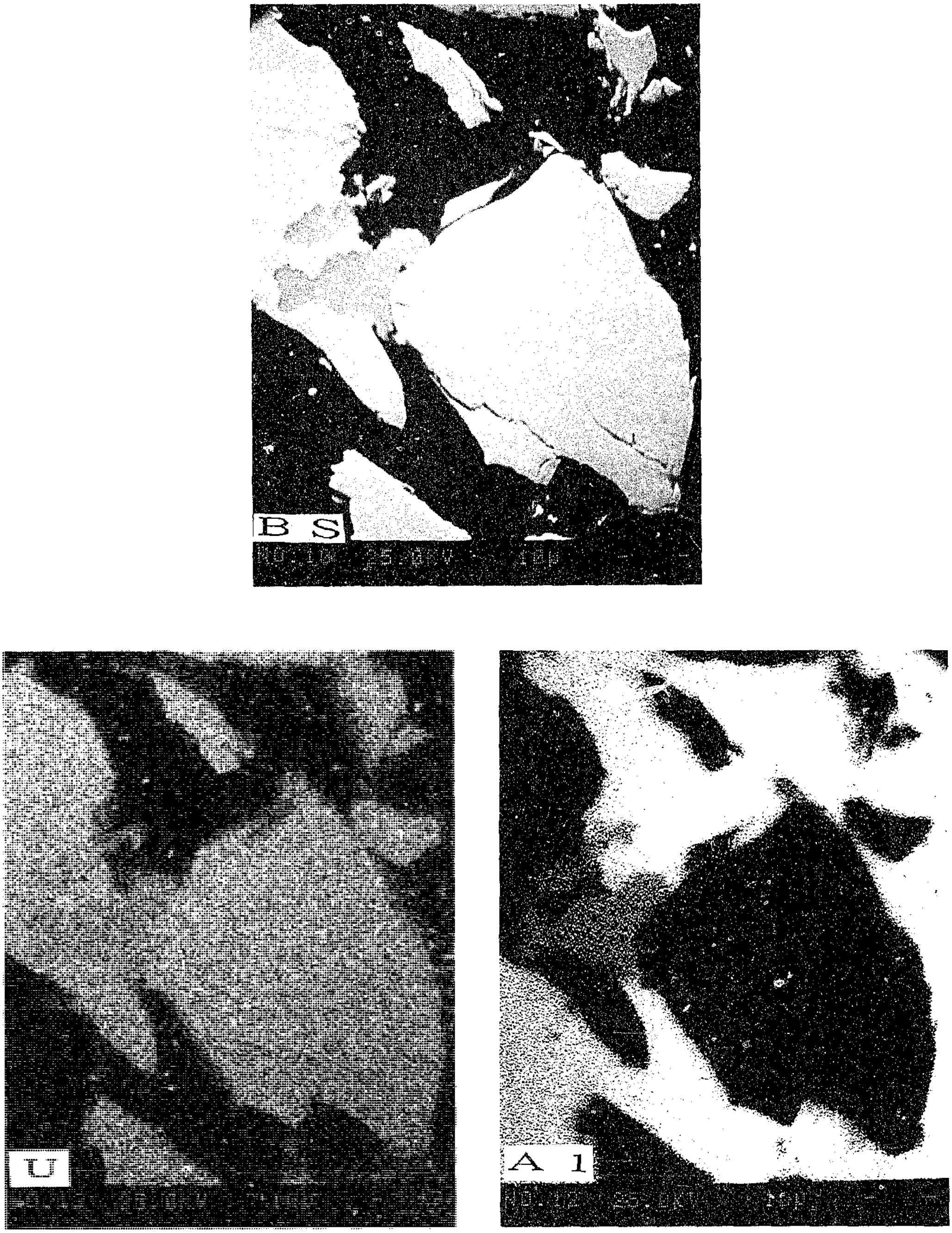

Fig. 4a EPMA analysis of cross section of 1.38-mm-thick miniplate with fuel meat $U_6(Fe_{0.6}Mn_{0.4})$-Al (BS = Back-scattered electron image)

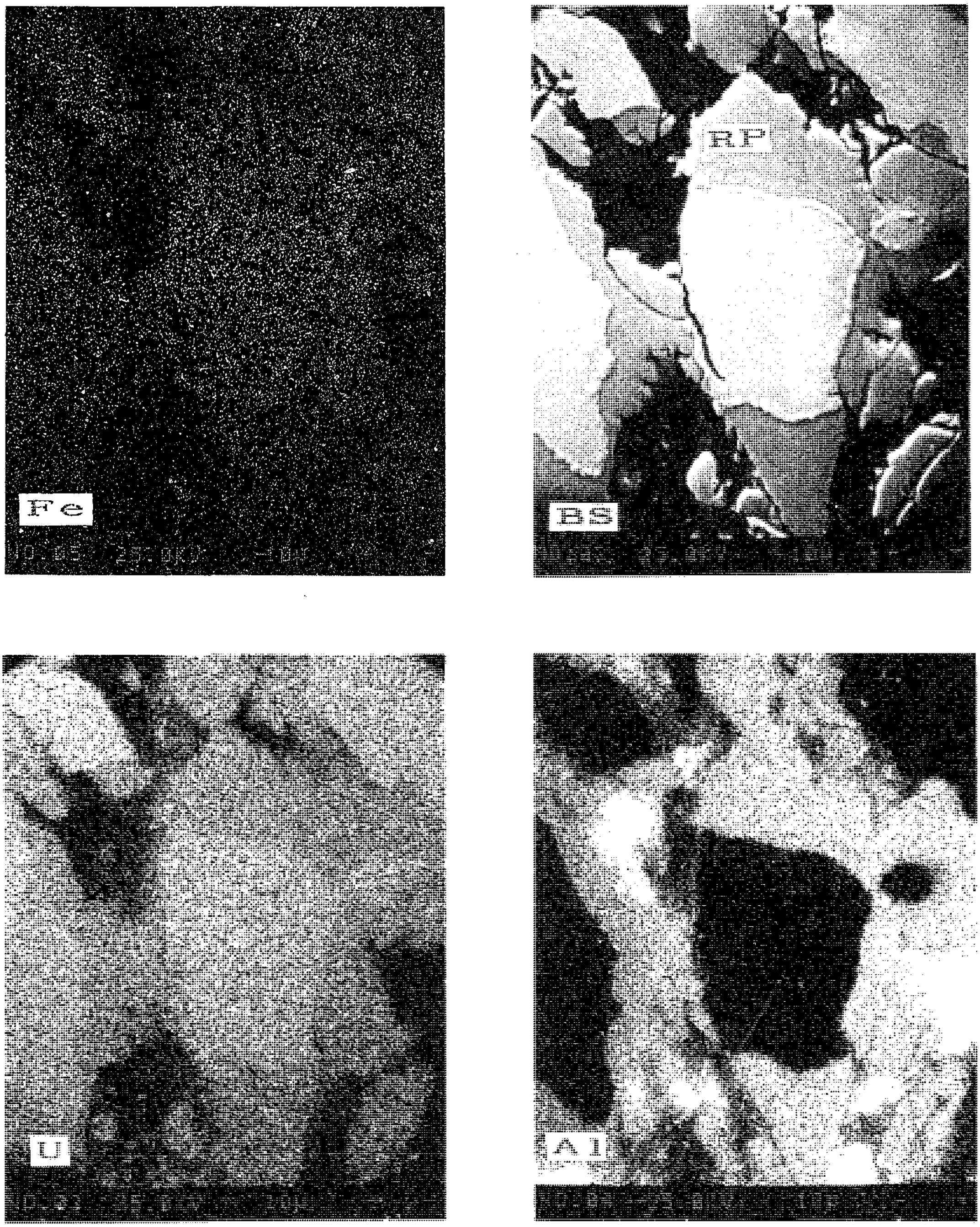

Fig. 4b EPMA analysis of the same miniplate as in Fig. 4a, heated at 450°C for 120 h.
RP = fuel - Al reaction product: $(U,Me)Al_3$

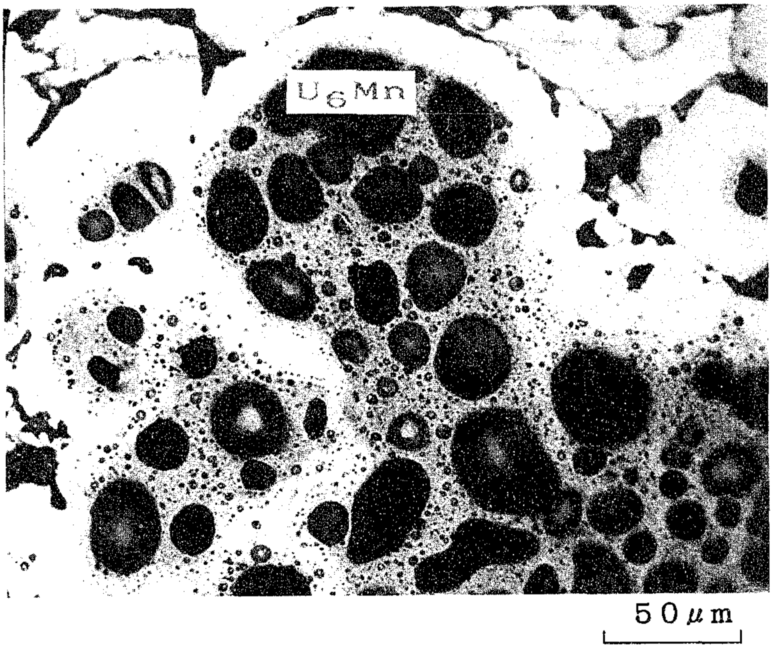

Fig. 5 Microstructure of Al-dispersion U_6Mn miniplate, irradiated to 53 % at 175 °C for 216 days

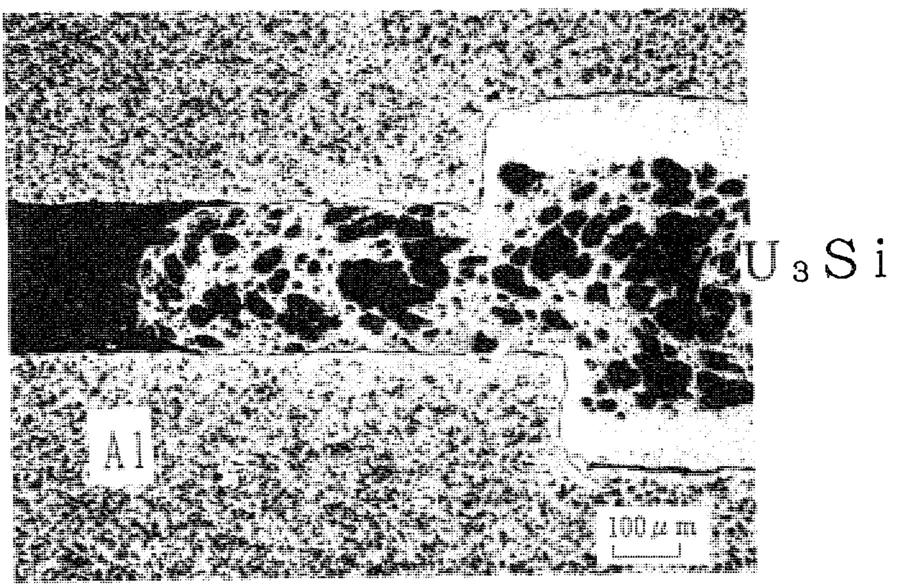

Fig. 6 Microstructure of U_3Si disk-type fuel, irradiated to 62 % at ~280 °C for 216 days