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Letter to the Editors

Irradiation behavior of high uranium-density alloys in the plate fuels

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Abstract

The U_6Mn and $U_6Fe_{0.6}Mn_{0.4}$ alloys were irradiated to ~54% of the initially contained 19.6% ^{235}U at around 190°C for 216 days. The volume swelling of the miniplates with such fuel dispersions in an Al matrix was reduced to the previously reported U_6Fe plate. Irradiation tests using $U_6No_{0.6}Fe_{0.4}$ and $U_3Si_{0.8}Ge_{0.2}$ proved that the cladding restraint is more effective to suppress the gas-bubble growth in the foil-type than in the dispersion-type plates. The fuel-aluminium reaction was also investigated. © 1998 Elsevier Science B.V.

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1. Introduction

The uranium silicide U_3Si_2 with a U-density of 11.3 g U/cm^3 is used as a low-enriched uranium plate-type fuel for research reactors. The U_3Si_2 particles are dispersed in an Al matrix to form fuel meat; by a roll-bonding process, the fuel meat is clad with an Al alloy to produce a plate (~1.3 mm-thick). In the future reactors, generation of higher neutron-fluxes will be needed. Also, the spent fuel arising should preferably be reduced for the limited storage capacity. Therefore, the U_6Me alloys (Me = Fe, Mn, Ni or their combinations) have been chosen as candidate fuels because of their higher U-density 17.0 g U/cm^3. We fabricated miniature plates (miniplates) which were irradiated in the Japan Materials Testing Reactor (JMTR). This paper describes irradiation behavior of U_6Me and U_3 (Si, Ge), with emphasis on the swelling due to fission-gas (Xe + Kr) bubbles and on the reaction with the Al matrix. Irradiation data [1] on U_6Me is available only for U_6Fe at the realistic burnup levels of reactor operation that was obtained by Argonne National Laboratory (ANL).

2. Preparation of miniplates

Miniplates containing U_6Mn or $U_6Fe_{0.6}Mn_{0.4}$ were prepared by the powder-metallurgical picture-frame method [2]:

(1) Ar arc-melting of U and the elements of Me (Si and Ge tips for $U_3Si_{0.8}Ge_{0.2}$).

(2) Annealing of as-melted buttons at 670°C for 10 days to obtain bct U_6Me, of which the existence was confirmed by X-ray diffraction.

(3) Crushing and powdering of annealed buttons.

(4) Sieving fuel powders to the four classes of particle sizes.

(5) Weighing and mixing the fuel and Al powders.

(6) Cold pressing of the mixed powders to form fuel compact.

(7) Assembling fuel compact, frame and cover made with Al–1.0 wt% Mg–0.6 wt% Si alloy (A16061).

(8) Peripheral welding of the assembly.

(9) 500°C hot-rolling followed by cold-rolling.

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Table 1
Thickness increases of irradiated U_6Me–Al dispersion miniplates

Alloy	U density (g U/cm^3)	Porosity (vol.%)	Burnup (% ^{235}U)	Irrad. temp. (°C)	Thickness increase (%)
U_6Mn	6.3	13	53	180	18
$U_6Fe_{0.6}Mn_{0.4}$	6.0	16	55	190	12
U_6Fe [1]	7.5	14	~ 46	110	~ 70

(10) Sizing (partial shearing and polishing of the rolled plates).

The sizes of the finished miniplates were: 20 mm wide, 30 mm long and ~ 1.3 mm thick. Weight fractions of each fuel particle-size (ps) in making compacts were: 19% for $ps < 45$, 20% for $45 \leq ps < 75$, 21% for $75 \leq ps < 106$ and 40% for $106 \leq ps < 150$ μm. The U_6Me alloys are friable; powders are easily produced from the buttons using a jaw crusher and agate mortar/pestle. This is an advantage of U_6Me over the U_3Si-based alloy that is too ductile to be comminuted. The U-density and void-volume fraction (porosity) of these dispersion-type miniplates were calculated from the data obtained by immersion density measurements [3].

The foil-type miniplates containing $U_6Ni_{0.6}Fe_{0.4}$ and $U_3Si_{0.8}Ge_{0.2}$ were also prepared. Instead of using the processes (3)–(6), the annealed buttons were cut into the foils (0.2–0.3 mm thick), embedded in the Al powder and then cold-pressed to obtain compacts.

3. Irradiation tests

The dispersion- and foil-type miniplates were irradiated in a He-sealed capsule for 216 days. Burnups were calculated from the thermal neutron fluxes $0.9–1.1 \times 10^{14}$ n/cm^2 s (< 0.68 eV) utilizing the ORIGEN code [4] modified for the present work, and found 53–57% of the initially contained 19.6% ^{235}U. Temperatures measured at surfaces of the miniplates were 180–190°C, and maintained almost constant during irradiation by controlling the He gas pressure outside the capsule. The fuel temperatures were estimated to be ~ 20°C higher than the surface ones at least in the early stage of irradiation. Post-irradiation examinations included plate thickness/volume measurements, X-ray radiography, optical metallography, micro-gamma scanning and EPMA/SEM analyses.

4. Results and discussion

Table 1 lists thickness increases of irradiated U_6Me–Al dispersion miniplates. Table 2 lists approximate maximum sizes of fission-product gas bubbles in the dispersion- and foil-type U_6Me and in the foil-type $U_3Si_{0.8}Ge_{0.2}$. These tables include the ANL data on the U_6Fe–Al dispersion miniplate [1]. It is seen that the increase of the plate thickness is primarily attributed to the fission gas-bubble swelling of the uranium alloys. Thickness of the plate increases with increasing size of the fission gas-bubble.

4.1. Thickness increase of U_6Me–Al dispersion miniplates

The U_6Mn and $U_6Fe_{0.6}Mn_{0.4}$ miniplates, respectively with 6.3 and 6.0 g U/cm^3 and a nominal fuel-volume loading of 42%, behaved well at burnups of 53–55%. Neither extreme swelling nor delamination of the plates was observed at irradiation temperatures higher than the ANL plate. Thicknesses of these miniplates before irradiation were 1.31 and 1.34 mm, respectively. The thickness increases due to irradiation were uniform along the rolling direction of the plates (Fig. 1a, Fig. 2a), and were 240 and

Table 2
Approximate maximum sizes of fission-product gas bubbles in U_6Me and $U_3Si_{0.8}Ge_{0.2}$ irradiated in miniplates for 216 days

Alloy	Fuel type	Burnup[a] (% ^{235}U)	Irrad. temp. (°C)	Max. bubble size (μm)
U_6Mn	dispersion	53	180	85
$U_6Fe_{0.6}Mn_{0.4}$	dispersion	55	190	75
U_6Fe [1]	dispersion	~ 46	110	140
$U_3Si_{0.8}Ge_{0.2}$	foil	57	190	2
$U_6Ni_{0.6}Fe_{0.4}$	foil	54	180	9[b]

[a] Fission densities: $U_6Mn = 3.7$, $U_6Fe_{0.6}Mn_{0.4} = 3.9$, U_6Fe [1] = ~ 3.3 (irradiated for 127 days), $U_3Si_{0.8}Ge_{0.2} = 3.5$, $U_6Ni_{0.6}Fe_{0.4} = 3.8 \times 10^{21}$ fissions/cm^3.
[b] Thickness increase of foil-type $U_6Ni_{0.6}Fe_{0.4}$ miniplate = ~ 7%.

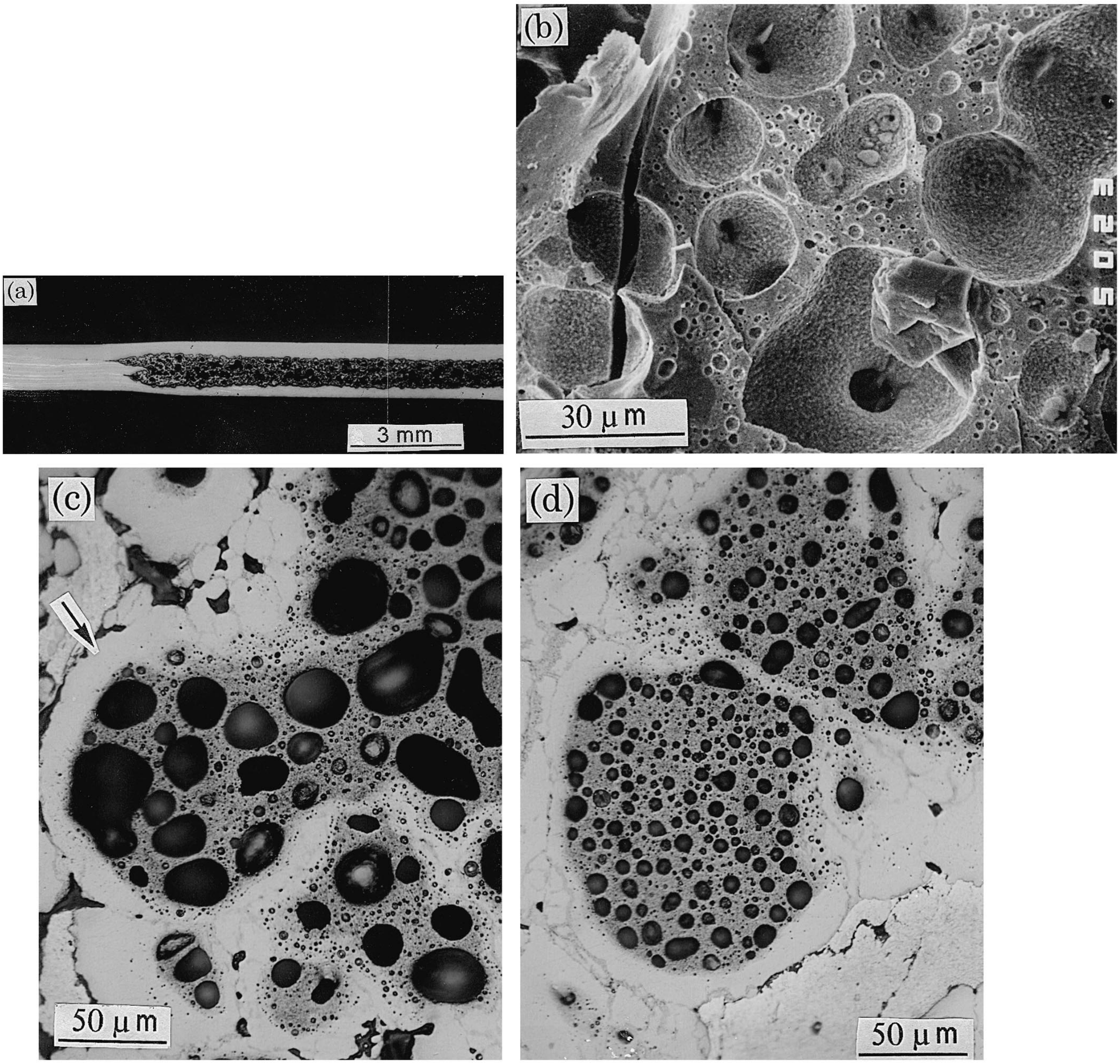

Fig. 1. U_6Mn–Al dispersion miniplate irradiated to 53% at 180°C for 216 days: (a) fuel meat clad with A16061 (partly), (b) SEM image of the fracture surface showing the fission-gas bubbles built up in U_6Mn and the tunnels in the large bubbles interconnecting originally two bubbles, (c) plastic flow of the fuel, accompanied by combination of the particles, (d) corner rounding and spheroidization of the fuel particles. The U_6Mn–Al reaction layers is indicated by an arrow.

160 µm, respectively. These correspond to 18 and 12% of the initial plate thickness (cf. ~ 70% in the ANL's U_6Fe plate). Relatively large gas-bubbles grow by the coalescence with others (Fig. 1b), and by the accommodation of small bubbles (Fig. 2b).

In the U_6Fe miniplate irradiated for 127 days, excessive fission gas-bubbles were formed at burnups of only about 20% of the 19.8% ^{235}U [1]. Plate failure occurred at ~ 40% burnup by fission gas-driven pillowing. The U_6Fe plate had a higher U-density, but similar as-fabricated porosity (Table 1). As can be seen from Fig. 1c, the U_6Mn particle broke its peripheral reaction layer and combined an adjacent particle with concomitant gas-bubble linking. This implies that the higher U-density, i.e., the higher fuel-particle density may enhance the plate swelling due to the bubble coarsening. In the U_3Si_2–Al dispersion plate, a higher fission rate tends to reduce fission-gas-bubble swelling at a given burnup [5]. If it is true for the U_6Me-type dispersion plate, the effect of the higher U-density on the gas-bubble swelling in the ANL plate will be offset by

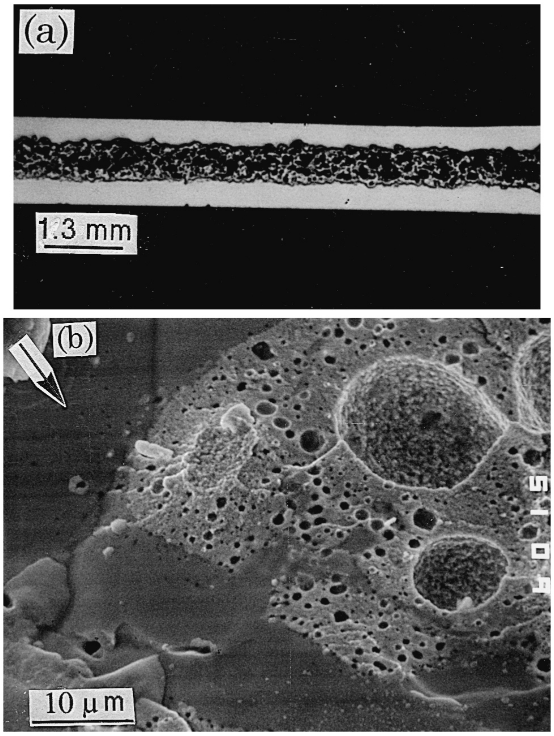

Fig. 2. $U_6Fe_{0.6}Mn_{0.4}$–Al dispersion miniplate irradiated to 55% at 190°C for 216 days: (a) fuel meat clad with A16061 (central part), (b) SEM image of the fracture surface of the $U_6Fe_{0.6}Mn_{0.4}$ particle: fission-gas bubbles forming the porous $U_6Fe_{0.6}Mn_{0.4}$ and the gas-bubble-free peripheral reaction layer (indicated by an arrow).

its higher fission rate (a factor of ~ 1.5 compared to ours). Thus, comparison of the dimensional stability may be valid between the ANL and our plates. The fission gas-bubbles observed in the U_6Fe particles are much larger than those in the present study. The maximum sizes of the gas bubbles in U_6Mn and $U_6Fe_{0.6}Mn_{0.4}$ were approximately 85 and 75 μm, respectively, whereas those of the U_6Fe were at least ~ 140 μm [1,6].

Hofman [6] estimated enthalpies for the crystalline-to-amorphous transformation of U_6Mn, U_6Fe, U_6Ni and U_6Co, and indicated that U_6Mn is the highest in value suggesting that this compound is the most stable against amorphization. The improved dimensional stability of U_6Mn and $U_6Fe_{0.6}Mn_{0.4}$ compared with U_6Fe is perhaps due to this crystal structure stability of U_6Mn, which could lead to the less fission-gas mobility and the lower rate of the fuel flow due to plasticity.

4.2. Gas-bubble swelling in the dispersion- and foil-type plates

The morphology of unirradiated U_6Mn particles in a dispersion-type plate showed a rectangular shape (Fig. 3), and did not change even after heating at 500°C for 40 h. However, after irradiation of 3.7×10^{21} fissions/cm^3, corner rounding and spheroidization of the U_6Mn particles were observed to minimize their surface energy (Fig. 1d). The irradiation temperature near 180°C is much lower than 726°C where crystalline U_6Mn melts peritectically [7]. Accordingly, such softening as well as plastic flow driven by bubble overpressure (Fig. 1c) will be evidence that might be a possible sign of amorphization. Amorphization of U_6Me has already been shown by other workers. Panteleev et al. [8] reported that U_6Mn, U_6Fe and U_6Ni undergo the radiation-induced crystalline-to-amorphous transformation after irradiation of $2 \times 10^{17-18}$ fissions/cm^3 at 40°C. Bloch [9] found the occurrence of amorphization of U_6Fe after only 2.3×10^{17} fissions/cm^3 at temperatures to 300°C.

The foil-type $U_6Ni_{0.6}Fe_{0.4}$ miniplate was irradiated to 54% at 180°C for 216 days (Fig. 4). The maximum bubble size is ~ 9 μm, and the plate thickness increase is about 7% (normalized to the dispersion-type $U_6Fe_{0.6}Mn_{0.4}$ plate). These values are considerably small compared to those of the dispersion-type U_6Mn and $U_6Fe_{0.6}Mn_{0.4}$ that were irradiated under similar conditions (see Tables 1 and 2). This foil-type miniplate was prepared from a compact with the fuel foil embedded in the Al matrix, and had as-fabricated porosity of ~ 3% (cf. 16% in the dispersion-type

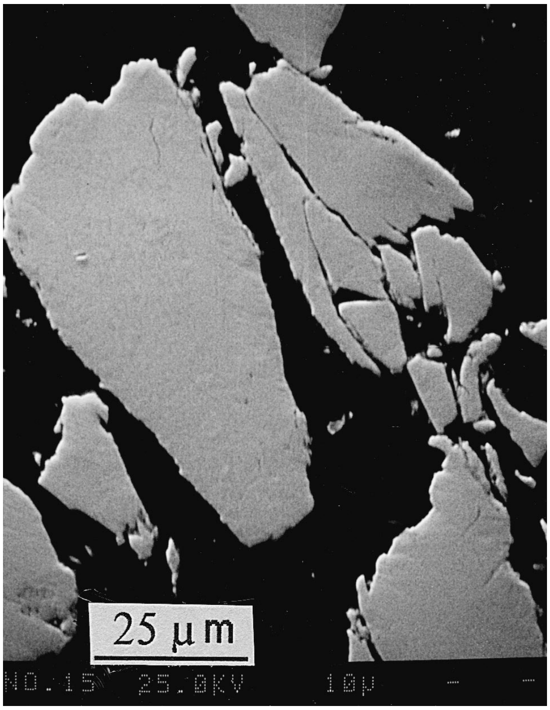

Fig. 3. Back-scattered electron image of the fuel meat in unirradiated U_6Mn–Al dispersion miniplate (black area = Al matrix).

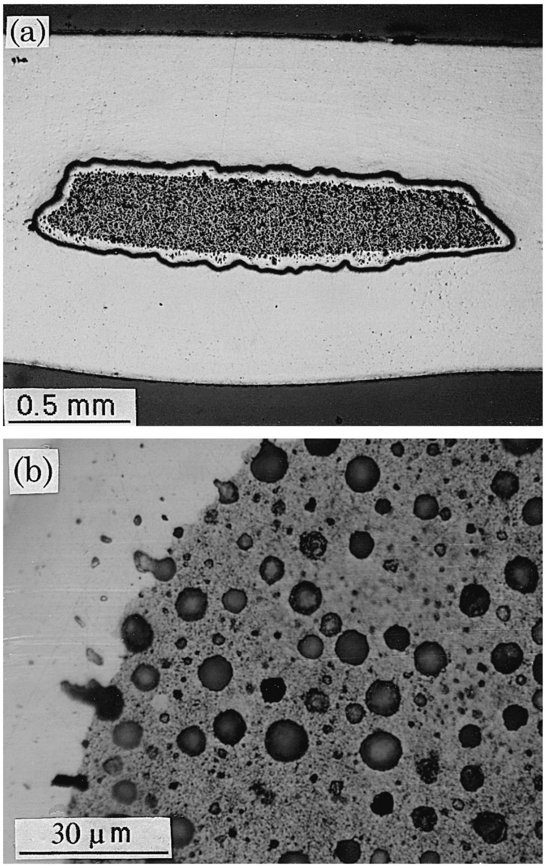

Fig. 4. Foil-type $U_6Ni_{0.6}Fe_{0.4}$ miniplate irradiated to 54% at 180°C for 216 days: (a) A16061-clad fuel foil in the Al matrix, (b) optical metallography showing fission-gas bubbles in $U_6Ni_{0.6}Fe_{0.4}$ much smaller than those in the dispersion-type.

$U_6Fe_{0.6}Mn_{0.4}$ plate). On the other hand, the foil-type $U_3Si_{0.8}Ge_{0.2}$ miniplate (as-fabricated porosity = ~ 3%) was also irradiated to 3.5×10^{21} fissions/cm^3 at 190°C (Fig. 5). Its swelling was estimated from the foil thickness before and after irradiation, and found to be approximately 15% neglecting the thickness increase due to the reaction with Al. This value of the $U_3Si_{0.8}Ge_{0.2}$ swelling is small (~ 1/2), compared to that of the U_3Si in the U_3Si–Al dispersion plate with higher as-fabricated porosity (7–10%) but at the same fission density [1].

The improvement of the dimensional stability of $U_6Ni_{0.6}Fe_{0.4}$ and $U_3Si_{0.8}Ge_{0.2}$ irradiated in the foil-type plates will owe to the narrow void space within the fuel meat and to the resultant effectiveness of the cladding restraint. This is consistent with the results of our recent study on irradiated U_3Si [10]. Namely, if compressive stresses and restraint due to the cladding are maintained during irradiation, these forces are effective to suppress the gas bubble growth, and therefore, the fuel swelling. On the contrary, the gas bubbles in highly irradiated U_3Si are allowed to grow by gas-atom collection and their coalescence, providing the local restraint force imposed to the fuel matrix is weakened. Such unrestraint condition is created by the gap or void space to which the direction the gas bubbles can expand. Thus, it may be concluded that the foil-type fuel having intrinsically small void space (i.e., low as-fabricated porosity) exhibits a dimensional stability superior to the dispersion-type.

4.3. Fuel–Al reaction

From ex-reactor experiments, it is assumed that the U_6Me–Al reaction under irradiation takes place according to

$$U_6Me + 21Al \rightarrow 7\left(U_{6/7}Me_{1/7}\right)Al_3.$$

The post-irradiation examination revealed that the mean layer thicknesses of the reactions of $U_6Ni_{0.6}Fe_{0.4}$ and $U_3Si_{0.8}Ge_{0.2}$ with Al are, respectively, 37 and 25 μm under similar irradiation conditions (Table 2). The reaction product (U,Me)Al_3, mainly consisted by UAl_3, has a lower density (~ 6.8 g/cm^3) compared with ~ 17.8 and 15.8 g/cm^3 for the U_6Me and U_3(Si,Ge) densities, respectively. It is estimated that the reactions will accompany theoretical volume increases of approximately 5 and 11%, respectively. Thus, contribution of the reaction to the plate thickness increase is almost the same between the U_6Me and the U_3Si-based alloys. SEM fractography of the $U_6Fe_{0.6}Mn_{0.4}$ particle (Fig. 2b) shows that the reaction product is gas-bubble free, although the fission density is less than a half of that of the fuel matrix.

5. Conclusions

The U_6Me-type intermetallic compounds, U_6Mn and $U_6Fe_{0.6}Mn_{0.4}$, were irradiated to burnups of ~ 54% of the initially contained 19.6% ^{235}U at around 190°C for 216 days. The dimensional stability of the miniplates with such fuel dispersions in the Al matrix was improved compared to the previously reported U_6Fe plate irradiated under similar conditions. The volume swelling of U_6Me results from the high fuel plasticity due to the possible amorphization that enables the rapid fission-gas-bubble growth and interlinking. Irradiation tests using $U_6Ni_{0.6}Fe_{0.4}$ and $U_3Si_{0.8}Ge_{0.2}$ proved that the cladding restraint is more effective to suppress the gas-bubble growth in the foil-type than in the dispersion-type plates. This will owe to the intrinsically smaller void space in the former. Contribution of the fuel–aluminum reaction to the plate thickness in-

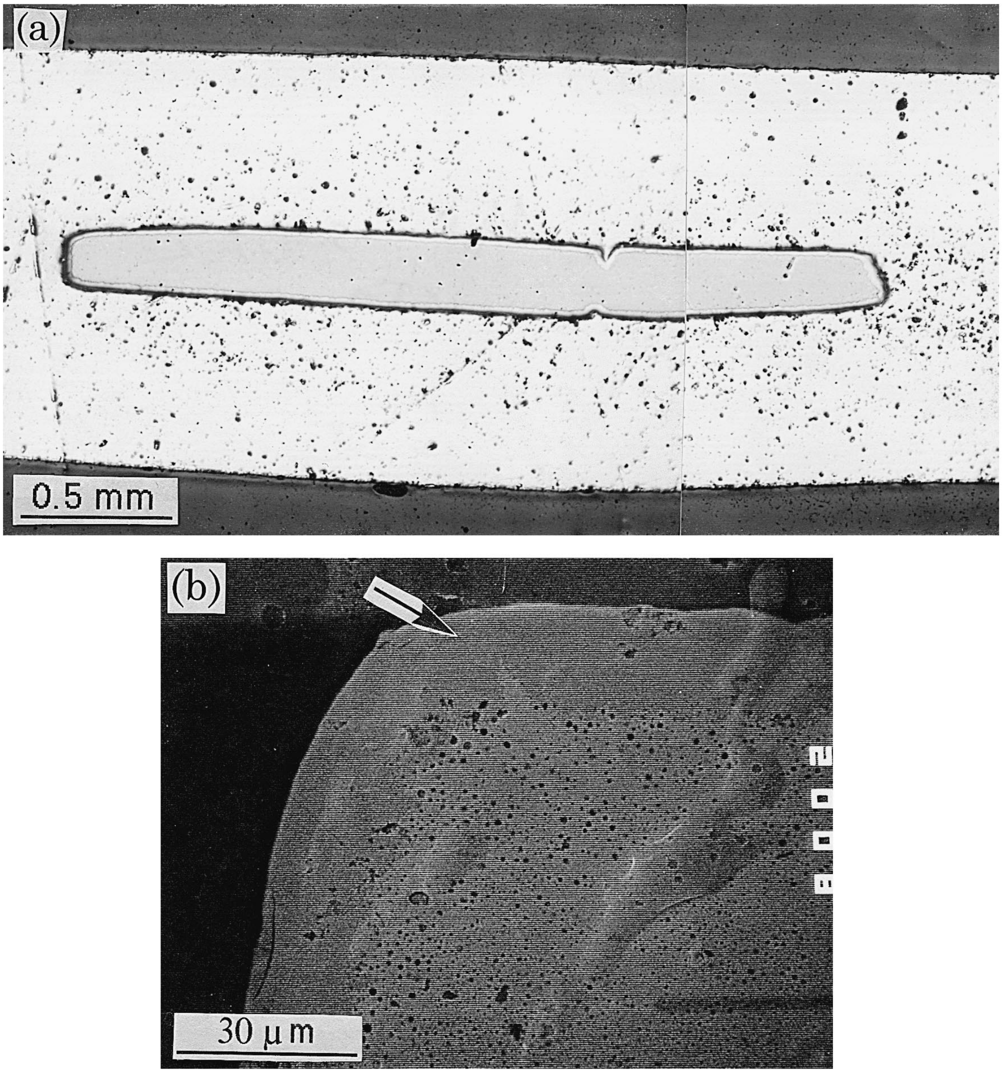

Fig. 5. Foil-type $U_3Si_{0.8}Ge_{0.2}$ miniplate irradiated to 57% at 190°C for 216 days: (a) A16061-clad fuel foil in the Al matrix, (b) SEM image showing small fission-gas bubbles in $U_3Si_{0.8}Ge_{0.2}$ and the gas-bubble-free peripheral reaction layer (indicated by an arrow).

crease is almost equal between the U_6Me and the U_3Si-based alloys.

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