1. Basic Research on High-Uranium Density Fuels for Research and Test Reactors

M. UGAJIN, A. ITOH, M. AKABORI

Department of Fuels and Materials Research
Tokai Research Establishment, JAERI
Tokai-mura, Naka-gun, Ibaraki-ken 319-11, Japan

ABSTRACT

High-uranium density fuels, uranium silicides (U_3Si_2, U_3Si) and U_6Me-type uranium alloys (Me=Fe,Mn, Ni), were prepared and examined metallurgically as low-enriched uranium (LEU) fuels for research and test reactors. Miniature aluminum-dispersion plate-type fuel (miniplate) and aluminum-clad disk-type fuel specimens were fabricated and subjected to the neutron irradiation in JMTR (Japan Materials Testing Reactor).
Fuel-aluminum compatibility tests were conducted to elucidate the extent of reaction and to identify reaction products. The relative stability of the fuels in an aluminum matrix was established at 350°C or above. Experiments were also performed to predict the chemical form of the solid fission-products in the uranium silicide (U_3Si_2) simulating a high burnup anticipated for reactor service.

1. INTRODUCTION

The Fuel Processing and Qualification Laboratory of the Department implemented a new program of exploring the next generation fuel for use in research and test reactors in April 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 for R & D because of their high-uranium densities: 11.3, 14.9 and 17.0 gU/cm^3, respectively.

The installation of equipment necessary for the preparation of miniature Al-dispersion plate-type fuel was completed at the end of March 1989. The first irradiation capsule (designated 88F-2A) containing uranium silicides was produced in November 1989 [1]; the neutron irradiation in JMTR started in May 1990 and terminated in May 1991. Postirradiation examinations of the 88F-2A capsule commenced at the beginning of October 1991.

The second irradiation capsule (designated 89F-1A) containing uranium silicides and U_6Me-type fuels was made in July 1990. The neutron irradiation was started also in JMTR in November 1990 and will continue to the end of November 1992, aiming at a burnup level higher than that achievable in the 88F-2A capsule.

This paper will deal with the experimental results so far obtained for uranium silicides and U_6Me-type fuels with emphasis on: (1) fuel preparation, (2) fuel-aluminum compatibility and (3) high-burnup simulation.

2. FUEL PREPARATION

2.1 MINIATURE PLATE-TYPE FUEL

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

Miniplates were fabricated according to the following steps:

(1) argon arc-melting of together charges of U metal and silicon tips
(2) annealing of arc-melted button at 850 °C for 72 h
(3) crushing and powdering of arc-melted and annealed button
(4) sieving fuel powders to 4 classes of particle sizes ($\leqq$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
(10) mechanical bond test: bending (delamination) test of a sample trimmed from rolled plate-end
(11) sizing (partial shearing and polishing) to finished miniplate: 20mm x 30mm x 1.3~1.4mm thick
(12) X-ray radiographic inspection to check fuel meat homogeneity and fuel-frame bond
(13) immersion density measurement

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.

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 to the U_3Si that is too ductile to be comminuted.

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

88F2A-01, 89F1A-01 = 100 wt% U_3Si_2
88F2A-02, 89F1A-02 = 80 wt% U_3Si_2 + 20 wt% USi
88F2A-03, 89F1A-03 = 99.5 wt% U_3Si_2 + 0.5 wt% Mo
88F2A-04, 89F1A-04 = 80 wt% U_3Si_2 + 20 wt% U_3Si
89F1A-05 = 100 wt% U_6Mn, 89F1A-06 = 100 wt% $U_6(Fe_{0.6}Mn_{0.4})$

The following miniplates are also included in the 89F-1A series capsule now under irradiation: $U_3(Si_{0.8}Ge_{0.2})$, $U_3(Si_{0.6}Ge_{0.4})$, $U_6(Fe_{0.4}Ni_{0.6})$ and U_6Ni.

Fig.1 shows a typical example[1,2] 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) range adopted for manufacturing the fuel compact are: 19% for ps$\leqq$45, 20% for 45$<$ps$\leqq$75, 21% for 75$<$ps$\leqq$106 and 40% for 106$<$ps$\leqq$150 μm.

2.2 ALUMINUM-CLAD DISK-TYPE FUEL

U_3Si, U_3Si + U_3Si_2, U_3Si_2 and U_3Si_2 + USi were arc-melted and heat-treated at 850°C for 72 h. The annealed silicide 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. Fig.2 shows a typical cross section of the aluminum-clad disk-type silicide fuel. The 28 disks fabricated were irradiated in the capsules 88F-2A and 89F-1A.

3. FUEL - ALUMINUM COMPATIBILITY

Fuel-aluminum compatibility is a matter of concern not only in the irradiation performance but also in the fabrication of fuels involving elevated temperatures[3,4]. Tests were conducted to elucidate the extent of the reaction and to identify reaction products by EPMA and X-ray diffraction analysis as well as metallographic examination. The results showed that U_3Si_2 exhibits greater stability than U_3Si in contact with aluminum at temperatures $\geqq$ 350°C. Ge-added U_3Si showed essentially no improvement in the compatibility with aluminum. The rates of reaction at temperatures $<$ ~300°C have been demonstrated to be very low.

Fig.3 shows volume increases of fuel-aluminum dispersion compacts as a function of temperature and time of heating for uranium silicides and U_6Mn.

The relative stability of the fuels was thus established for aluminum at temperatures $\geqq$ 350°C as follows:

$$U_3Si \sim U_3(Si,Ge) < U_6Mn << U_3Si_2 \sim USi$$

As seen from the 450°C data points in Fig.3, U_6Mn is marginally more stable than U_3Si despite its lower peritectic temperature 726°C, compared with the peritectoid temperature of 925°C for U_3Si.
This may be due to the differences between U_3Si and U_6Mn in the reaction kinetics for aluminum; in the former, path for the preferential diffusion of aluminum is formed along the U_3Si grain boundaries.

As the reaction products, the compounds $U(Al,Si)_3$ and $(U,Me)Al_3$ were identified respectively, for uranium silicides and for U_6Me-type fuels. Fig.4 illustrates an EPMA image of reacted $U_6(Fe_{0.6}Mn_{0.4})$ particles in a miniplate shown next. Fig.5 shows a metallographic cross-section (perpendicular to the rolling direction) of an initially 1.38-mm $U_6(Fe_{0.6}Mn_{0.4})$-dispersion miniplate heated at 450°C for 120 h; a plate-thickness increase of about 30% is noted at the fuel-aluminum interaction zone.

The observed volume and thickness increases are considered to be attributed to the void formation due to the reaction and to the formation of less-dense $U(Al,Si)_3$ and $(U,Mn)Al_3$ (U/Mn atom ratio ~6). These compounds have a lower density of ~6.7 g/cm^3, compared with the respective theoretical density of 12.2 and 17.8 g/cm^3 for U_3Si_2 and U_6Mn.

To improve the compatibility of U_3Si and aluminum, the U_3Si-Al matrix interface was stabilized by the slight oxidation of the surface of U_3Si. The validity of this method was examined employing the disk-type fuel. The surface oxidation of the thin U_3Si plate 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+x} on the surface of the U_3Si plate. The compatibility test was carried out at 500°C for 20 h using a disk-type specimen in which the U_3Si plate had the oxide layer on one side only. As evident in Fig.6, no reaction was observed at the interface between the surface-oxidized U_3Si and the Al-cladding while the reaction zone of ~50 μm was observed on the other side of the U_3Si plate.

4. HIGH-BURNUP SIMULATION EXPERIMENT

The chemical form of solid fission-products is of particular importance since it may affect the fission product release under a hypothetical accidental condition and the fuel-aluminum reaction during irradiation especially at abnormally high temperatures. However, essentially no information was previously available for the silicide fuel.

The uranium silicide (U_3Si_2) with 19.7% ^{235}U was assumed here to have

been irradiated under the likely operating conditions of research reactor: thermal neutron flux = 1.0 x 10^{15} n/cm^2 • sec, irradiation time = 75 EFPD. Inventories of actinide and fission products were calculated using the ORIGEN code with input parameters THERM=0.7989, RES=0.18388 and FAST=1.5305. The burnup value, ~98% of ^{235}U, is considerably high and may be the maximum anticipated for this type of fuel in reactor service.

Three simulated fuel samples were prepared by arc melting the together charges of elements according to the calculated inventories, and were heat-treated at 900-1100°C for 4-7 days. The following 13 elements were simulated in this work:

U, Si, Mo, Ru, Rh, Pd, Nd, Ce, La, Pr, Y, Zr, Sr.

Other rare earth, alkaline earth and actinide elements were taken into consideration by adding their equivalent quantities to the charges using the elements listed above. Phases present in the samples were identified by EPMA and X-ray diffractometer.

On the basis of the present investigation, the chemical form of the solid fission-products can tentatively be classified into the following four categories:

(A) Matrix phase U_3Si_2:

The major constituents of the matrix phase are uranium and silicon; it consists of tetragonal U_3Si_2. The solubility limits of other elements were found to be very small to the U_3Si_2 phase; e.g., the Mo solubility is less than ~0.05 wt%.

(B) U_5MoSi_4-type precipitate:

Mo, Ru and Rh form a solid solution based on this new compound; its crystal structure has not yet been defined but found very close to that of U_3Si_2. It should be noted that palladium added as one of the platinum group elements does not coexist with Ru and Rh.

(C) RESi-type (RE=rare-earth elements) precipitate:

Rare-earth elements (Nd, Ce, La, Pr and Y) form this type of silicide. Palladium and part of the uranium enter into the lattice to form the solid solution (RE,Pd,U)Si.

Fig.7 shows EPMA data indicating the existence of the RESi-type precipitates in the matrix phase of U_3Si_2.

(D) Free metal

Strontium and barium appear to remain as metallic state without forming silicide or uranium compound. Attempts using $SrSi_2$ instead of strontium metal as starting material failed to form any type of strontium silicide.

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

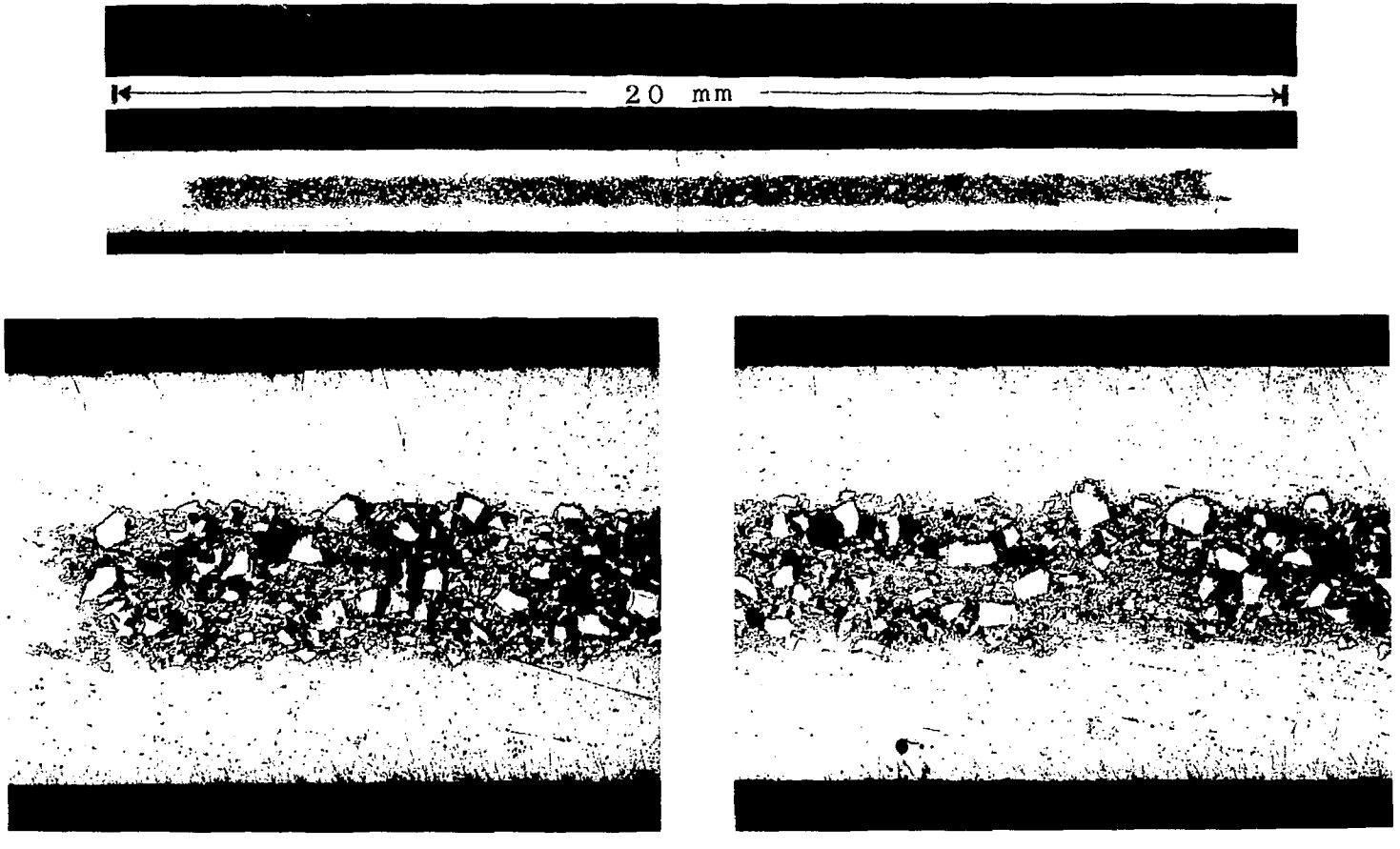

Fig.1 Cross-section of 1.40-mm-thick miniplate with fuel meat of U_3Si_2 and Al, clad with 6061Al alloy (Bottom:Partly enlarged of the top)

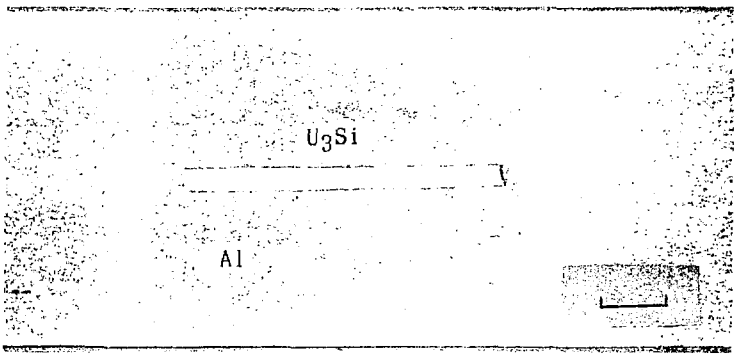

Fig.2 Cross section of aluminum-clad disk-type silicide fuel

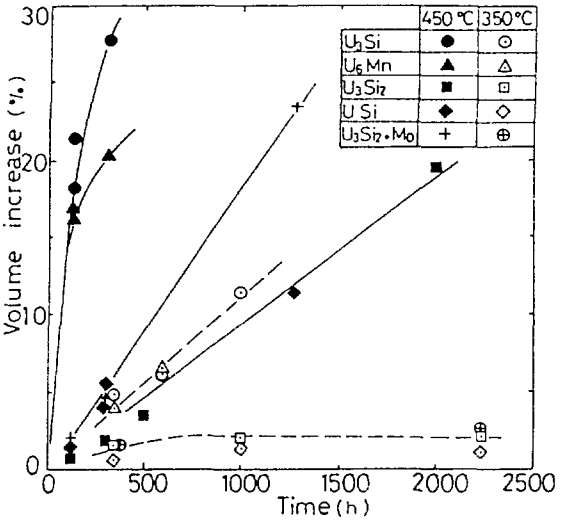

Fig.3 Volume increases of fuel compacts comprising aluminum and uranium silicides or U_6Mn due to their reaction at 350° and 450°C

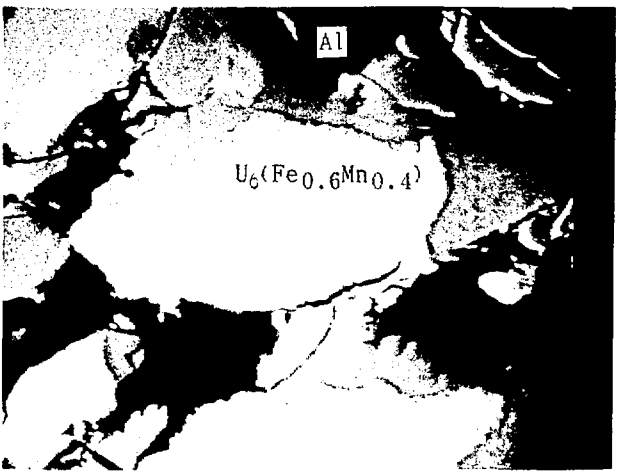

Fig.4 EPMA backscatter image of $U_6(Fe_{0.6}Mn_{0.4})$ particles reacted with aluminum in a miniplate same as in Fig.5

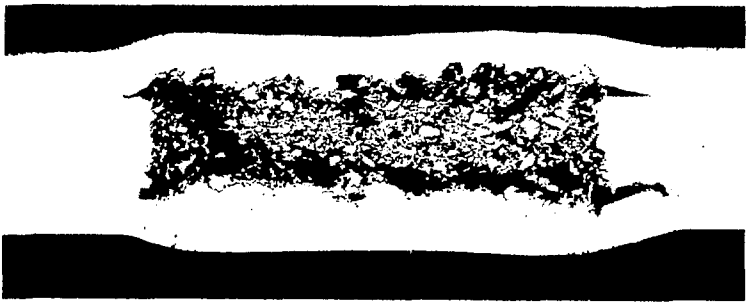

Fig.5 Cross-section of initially 1.38-mm-thick miniplate with fuel meat $U_6(Fe_{0.6}Mn_{0.4})$-Al after heating at 450°C for 120 h (see also Fig.4)

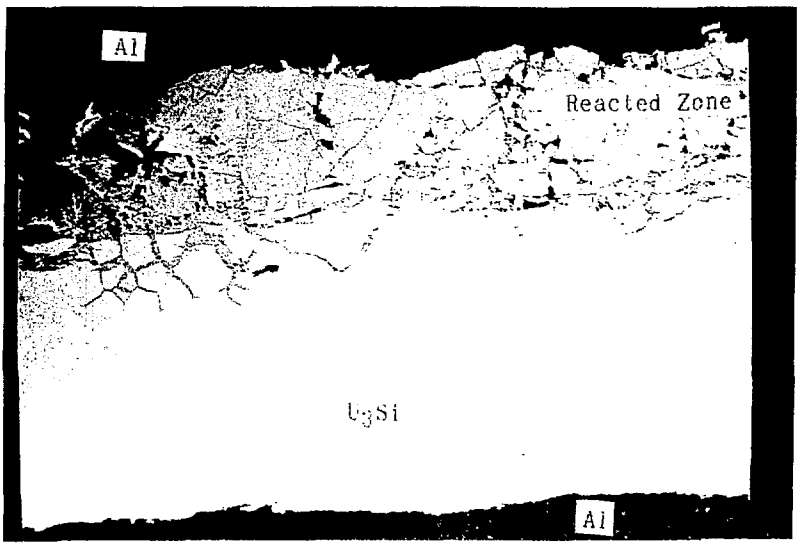

Fig.6 Cross section of aluminum-clad disk-type U_3Si (Lower side: surface-oxidized) after heating at 500°C for 20 h (EPMA backscatter image)

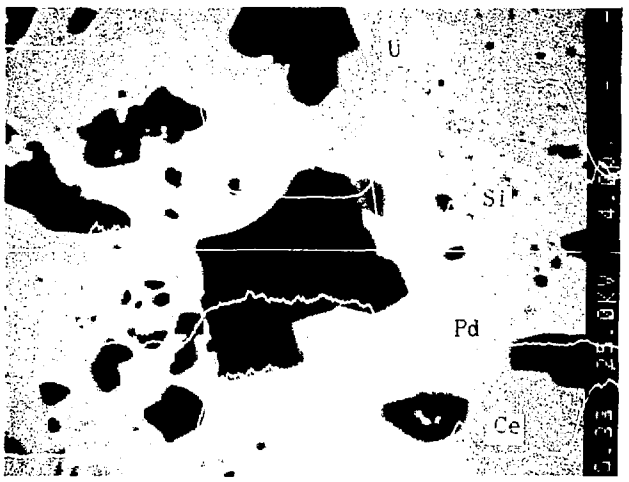

Fig.7 EPMA backscatter image and line-scan analysis showing the existence of the (RE,Pd,U)Si phase (RE=Ce,La,Nd,Y) in the matrix of U_3Si_2

5. SUMMARY AND CONCLUSIONS

Preparation experiments were carried out to fabricate miniature aluminum-dispersion plate-type fuel and aluminum-clad disk-type fuel by using the conventional picture-frame method and hot-pressing technique, respectively. Fuel materials produced were U_3Si_2, U_3Si, USi and U_6Me (Me=Fe,Mn, Ni). Totally 14 miniplates with uranium densities from 4.0 to 6.3 gU/cm^3 and 28 disk-type fuel specimens containing structurally-modified U_3Si were prepared and subjected to the neutron irradiation in JMTR. Postirradiation examinations just started for the first domestic fuel specimens will provide a basis for better understanding of the behavior of high-uranium density fuels.

Fuel-aluminum compatibility tests demonstrated that U_3Si_2 exhibits greater stability than U_3Si in contact with aluminum at or above 350°C. It was also shown that the rates of reaction at temperatures less than ~300°C are very low. U_6Mn is marginally more stable than U_3Si in the aluminum matrix. $U(Al,Si)_3$ and $(U,Me)Al_3$ were identified as the reaction products, respectively, for uranium silicides and for U_6Me-type fuels. The observed volume and thickness increases are considered to be attributed to the void formation due to the reaction and to the formation of these less-dense compounds. Ge-added U_3Si indicated no improvement in compatibility, while the surface-oxidized U_3Si showed no evidence of the reaction with aluminum at a temperature as high as 500°C.

High-burnup simulation experiments were also performed to predict the fission-products behavior in uranium silicide (U_3Si_2) irradiated to approximately 98% burnup of the ~20% ^{235}U. Emphasis of the study was placed on the chemical form of the solid fission-products comprising zirconium, molybdenum, rare-earth elements, alkaline-earth metals and elements of the platinum group. It was revealed that this new type of fuel shows the unique nature in the fission-products chemistry. Further study will be needed to confirm the present results and to obtain more detailed information on this multicomponent system.

ACKNOWLEDGEMENTS

The present authors are grateful to Dr. T. Kondo and Dr. M. Handa, respectively, Director and Deputy Director of the Department of Fuels and Materials Research for their encouragement.

REFERENCES

(1) M.Ugajin, A. Itoh, M. Akabori, K.Ikawa,"Preparation of silicide fuel for capsule irradiation" : presented at the 1989 fall meeting of the Atomic Energy Society of Japan, Oct.17-19, 1989 (Tokai-mura, Japan) Abstract No. K57 (in Japanese)

(2) M.Ugajin, "Basic program to compute uranium density and void volume fraction in laboratory-scale uranium silicide aluminum dispersion plate-type fuel", JAERI-M 91-069 (May, 1991)

(3) N.Ooka, K.Kanaya, Y.Etou, H.Yamashita, "Experiments on compatibility of U_3Si_2 and Aluminum at high temperature in out-pile", Keikinzoku yousetsu 339,28(1990)8 (in Japanese)

(4) T.C.Wiencek,R.F.Domagala and H.R. Thresh, "Thermal compatibility studies of unirradiated uranium silicide dispersed in aluminum", Nucl. Technol. 71(1985)608