CONF-8410173--4

CONF-8410173--4

DE85 004037

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by

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September 1984

Submitted to the RERTR Program Office for oral presentation and publication in proceedings of international meeting to be held at Argonne National Laboratory on October 15-18, 1984.

*Work supported by the U.S. Department of Energy.

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ABSTRACT

Powder metallurgy dispersions of uranium silicides in an aluminum matrix have been developed by the international Reduced Enrichment for Research and Test Reactors program as a new generation of proliferation-resistant fuels. A major issue of concern is the compatibility of the fuel with the matrix material and the dimensional stability of this fuel type.

A total of 45 miniplate-type fuel plates were annealed at 400°C for up to 1981 hours. A data base for the thermal compatibility of unirradiated uranium silicide dispersed in aluminum was established. No modification tested of a standard fuel plate showed any significant reduction of the plate swelling.

The cause of the thermal growth of silicide fuel plates was determined to be a two-step process: 1) the reaction of the uranium silicide with aluminum to form $U(AlSi)_3$ and 2) the release of hydrogen and subsequent creep and pillowing of the fuel plate.

INTRODUCTION

The Reduced Enrichment for Research and Test Reactor (RERTR) program has demonstrated that powder metallurgy dispersions of uranium silicides in aluminum are potentially a new generation of proliferation-resistant fuels.[1] As with any composite material the stability of the components can affect the performance of the structure. This presentation is limited to a discussion of studies of the compatibility of uranium silicide fuel with its cladding and matrix for plate-type configurations.

The term compatibility refers to the effect of varied environments on dissimilar materials in contact with each other. Dimensional and geometric changes occur between uranium-silicide fuels and aluminum powders due to diffusion reactions.[2] Such changes, even in the absence of a neutron flux, always cause an increase in volume of the materials. That is, irrespective of the fuel (U_3Si, U_3Si_2, or U_3SiAl), the reaction products between the silicide and aluminum are less dense than the reactants. The result, invariably, is an increase in the plate thickness. Prolonged anneals have also resulted in "dishing" and "pillowing" of simulated miniplates. Inasmuch as similar dimensional and geometric changes have been observed to occur as a result of postirradiation examination (PIE) tests[3] (e.g., blister tests), it is desirable if not mandatory that the changes associated with thermally induced diffusion reactions be defined as reference values for plate changes related to the presence of a neutron flux. Such information has become even more important as dimensional changes in irradiated plates have been measured recently in plates taken to very high "burn-ups," of the order of 80% or more.[4]

For the reaction between U_3Si fuel particles and Al, the following reaction and volume change could be expected:

$$U_3Si + 8\ Al \rightarrow 3\ U(Al,Si)_3.^* \qquad (1)$$

The stoichiometric reaction would occur at ∿38 vol % U_3Si and 62 vol % Al, and the $\Delta V/V$ (volume change) would be ∿11%. If all the increase was constrained to a thickness increase of a standard 0.51-mm (0.020-in.)-thick[5] meat, the plate thickness would grow 0.056 mm (∿2.2 mils) [(i.e., from 1.27-1.33 mm (0.050 to 0.0522 in.) or from 1.52-1.58 mm (0.060 to 0.0622 in.)]. This assumes no porosity in the fuel zone of a finished plate (a 40-vol % U_3Si miniplate has ∿10-12% porosity) a complete reaction (i.e., equilibrium is attained), and no increase in either the width or the length of the fuel zone.

For the U_3Si_2 + Al reaction, equation (2) occurs:

$$U_3Si_2 + 7\ Al \rightarrow 3U(Al,Si)_3.^* \qquad (2)$$

Stoichiometrically, at 48 vol % U_3Si_2, 52 vol % Al, and no porosity in the fuel zone, the $\Delta V/V$ due to the reaction (at equilibrium) would be ∿7%, or about half that for U_3Si. Plate thickness increase for 0.51-mm (0.020-in.) meat would be 0.036 mm (∿0.001 in.).

For U_3SiAl (U + 3.5 wt % Si + 1.5 wt % Al), the reaction product would be principally $U(Al,Si)_3$.* Balancing the equation is more complicated because the fuel alloy particles before reaction contain U_3Si, U_3Si_2, and UAl_2 phases.

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*$U(Al,Si)_3$ is a notation for a singular phase based on UAl_3 wherein some of the Al atoms have been replaced by Si atoms. Equations (1) and (2) would appear to be "unbalanced" chemically. However, inasmuch as UAl_3 and USi_3 apparently form a continuous solid solution field, the number of Si atoms in $U(Al,Si)_3$ will vary according to where one is in the two-phase [Al + $U(Al,Si)_3$] field or the one-phase $U(Al,Si)_3$ field. The density of $U(Al,Si)_3$ varies as a function of composition.[5]

However, the volume change would be less than that for U_3Si and more than for U_3Si_2. So the ΔV/V figure would be <11% and >7% -- or between 0.036-mm (0.001-in.) and 0.051-mm (0.002-in.) growth in plate thickness. Again, this is based on no porosity, a complete reaction, and all growth limited to a thickness increase for 0.51-mm (0.020-in.) meat.

The reactions noted above are diffusion controlled and therefore exponentially dependent on temperature. The rates of reaction at temperatures of 300°C and less have been demonstrated to be very low in the absence of a neutron flux.[2]

The net result of this introduction is to conclude that severe growth and/or blistering and/or pillowing of fuel plates observed in reactor and PIE tests should not occur under research reactor conditions as a result of diffusion reactions only. The purpose of the studies described here was to investigate the cause of the growth of silicide fuel plates and to establish a data base which will be useful for PIE and modeling studies as well as for understanding the behavior of dispersed phase silicide fuels.

MATERIALS AND PROCEDURES

Simulated miniplates were fabricated by standard procedures[6] using cylindrical compacts [19.05-mm (0.750-in.) diameter] which produced fuel zones nominally elliptical in shape. Plates were sheared (not machined) to a size of approximately 50.8 x 152.4 x 1.52 mm (2.0 x 6 x 0.060 in.). All plates were tested in the as-cold-rolled condition with ∿15% cold work. The test matrix described below and summarized in Table 1 was fabricated.

Three fuels were used in this study: U_3Si (U + 4.0 wt % Si), U_3Si_2 (U + 7.5 wt % Si), and U_3SiAl (U + 3.5 wt % Si + 1.5 wt % Al) depleted-uranium alloy powders only. Most compacts were made with 85% (-100 + 325) mesh (150 - 45 μm) + 15% (-325) mesh powder. The standard compact porosity was ∿14%.

The clad for all plates was 6061 Al. For most plates, degassed Alcan MD101 Al powder (99.3 wt % free metallic Al by analysis) was used as the matrix material. However, a few selected compacts were prepared using degassed Alcoa No. 718 powder. The 718 powder is nominally Al + 12 wt % Si + 0.5 wt % Fe (12.2 wt % Si + 0.4 wt % Fe by analysis). The selection of this powder matrix was based on a report in the literature stating that the reaction between Al and U may be reduced by as much as a factor of five if an Al + Si alloy replaces Al.[7]

In the interest of economy, only two levels of fuel-alloy concentration were selected: 32 and 45 vol % for each alloy. This converts to U loadings of from 3.4 g/cm^3 (32 vol % U_3Si_2) to 6.5 g/cm^3 (45 vol % U_3Si).

Reference and control samples were fabricated and treated in a manner identical to the "regular" test samples. These include:

Table 1. Test Matrix of Simulated Fuel Plates

Identification	Matrix	Fuel	Volume Percent Fuel in Fuel Zone	Notes
CS-6061-16 to 24	None	None	-	6061 rolled stock
CS-MD101-25 to 28	MD101	None	-	Not degassed Al
CS-MD101-29 to 32	MD101	None	-	Degassed Al
CS-Al718-33 to 36	718Al	None	-	Degassed 718Al
CS-WAl-37 to 40	MD101	W	32	Degassed Al, no W fines
CS-T-41 to 48	MD101	U_3SiAl	32	Degassed Al
CS-T-49 to 56	MD101	U_3SiAl	45	Degassed Al
CS-T-57 to 60	MD101	U_3SiAl	32	Degassed Al, no U_3SiAl fines
CS-T-61 to 64	MD101	U_3SiAl	45	Degassed Al, no U_3SiAl fines
CS-T-65 to 68	718Al	U_3SiAl	32	Degassed 718Al
CS-T-69 to 72	718Al	U_3SiAl	45	Degassed 718Al
CS-T-73 to 76	MD101	U_3SiAl	32	Low compact porosity
CS-T-77 to 80	MD101	U_3SiAl	45	Low compact porosity
CS-T-81 to 84	MD101	U_3SiAl	32	Degassed compact
CS-T-85 to 88	MD101	U_3SiAl	45	Degassed compact
CS-B-89 to 94	MD101	U_3Si	32	Degassed Al
CS-B-95 to 100	MD101	U_3Si	45	Degassed Al
CS-X-105 to 110	MD101	U_3Si_2	32	Degassed Al
CS-X-111 to 112	718Al	U_3Si_2	32	Degassed 718Al
CS-X-113 to 118	MD101	U_3Si_2	45	Degassed Al
CS-X-119 to 120	718Al	U_3Si_2	45	Degassed 718Al
CS-B-126 to 127	718Al	U_3Si	32	Degassed 718Al
CS-B-128 to 129	718Al	U_3Si	45	Degassed 718Al
CS-6061-130	None	None	-	6061 Rolled Stock
CS-B-131 to 132	Mg	U_3Si	45	Degassed Mg
CS-X-133 to 134	Mg	U_3Si_2	45	Degassed Mg
CS-F-135 to 136	Mg	U_6Fe	45	Degassed Mg
CS-Mg-138 to 139	Mg	None	-	Degassed Mg
CS-B-152	Mg	U_3Si	45	Degassed Mg
CS-X-153	Mg	U_3Si_2	45	Degassed Mg
CS-F-154	Mg	U_6Fe	45	Degassed Mg
CS-Mg-155	Mg	None	-	Degassed Mg

a) No compact at all, that is, three bonded layers of 6061.

b) A pure MD101 Al compact used as the "fuel."

c) A compact of Aloca 718 powder (Al + Si) used as the "fuel."

d) A compact with W + Al used as the "fuel". (This category was chosen to represent a combination of Al together with a high-density metal wherein a reaction at the test temperatures should be minimal.)

e) Several samples prepared with vacuum degassed compacts [1-2 h at 200°C and 1.3 Pa (10^{-2} torr)].

f) A compact with all -100 + 325 mesh (150-44 μm) powder (no fines).

g) A compact with less than 5% porosity.

h) Magnesium powder used as the matrix.

Samples were annealed at 400 ± 5°C for incremental times up to 1981 hours. Samples were periodically withdrawn from the electrically heated air furnaces, measurements made, and the specimens returned to the furnace for an additional time which was added to the already accumulated exposure.

After each heat treatment the appearance, weight, thickness, in and out of fuel zone, and immersion density were determined and recorded. Periodically, the samples were "Oakite" cleaned to remove the aluminum oxide film. Immersion density measurements permitted very accurate calculations of fuel zone volumes and porosities of the fuel zone.

It may be noted that the matrix in Table 1 is heavily skewed to the U_3SiAl fuel. U_3SiAl was the leading fuel because of previous irradiation tests in rod elements and its excellent corrosion resistance. However, after the postirradiation examinations, U_3SiAl was found to be unacceptable as a fuel at high burnups in plate-type elements.[4] By the time these results were presented, the compatibility study was well under way. The investigation was nevertheless continued with U_3SiAl in the belief that variations resulting in a substantial reduction in thermal growth for the U_3SiAl fuel would probably apply also to U_3Si and U_3Si_2.

Selected U_3Si, U_3Si_2, and U_3SiAl plates were also chosen to be "pillowed." Pillowing is defined as an extreme change in thickness of a fuel plate, usually an increase of the fuel zone volume by a factor of two or more. Fuel plates were pillowed by heating for ∿200 h at 500°C.

RESULTS AND DISCUSSION

A total of 45 of the fuel plates from Table 1 were chosen for annealing at 400°C. The annealing times and the volume percent increase in the fuel zones are presented in Table 2. The results for the fuel zone volume changes

Table 2. Compatibility Study of Fuel Zone Volume Changes as a Function of Annealing Time

Fuel Alloy	Vol %	Matrix	Compact Condition	Fuel Zone Volume - As Fabricated, cm^3	Percent Fuel Zone Volume Increase After Annealing at 400°C											
					2 h	4 h	10 h	26 h	50 h	100 h	196 h	428 h	733 h	975 h	1310 h	1981 h
U_3Si_2	32	MD101	Normal	0.8726	0.90	0.89	0.77	0.85	0.97	0.96	1.51	2.18	4.62	7.11	10.83	16.83
	32	718A1	Normal	0.9200	1.35	1.47	3.42	8.26	13.00	22.33	30.51	a	a	a	a	a
	45	MD101	Normal	0.9337	0.45	0.55	0.61	0.95	1.15	1.72	3.53	6.97	13.48	19.65	27.68	38.98
	45	718A1	Normal	1.0059	1.45	1.83	4.28	12.33	21.02	41.82	57.78	a	a	a	a	a
U_3Si	32	MD101	Normal	0.8898	1.49	1.38	2.08	3.19	4.22	6.82	12.91	26.01	41.21	51.39	61.84	73.61
	32	718A1	Normal	0.9427	1.69	2.08	3.31	5.99	8.35	12.55	16.96	a	a	a	a	a
	45	MD101	Normal	0.9604	2.21	2.18	2.95	4.35	6.04	9.64	17.43	32.58	51.42	64.88	76.36	88.08
	45	718A1	Normal	1.0223	2.10	2.76	4.24	6.96	9.41	13.78	18.66	a	a	a	a	a
U_3SiAl	32	MD101	Normal	0.8864	-	1.10	1.49	2.55	3.97	7.09	14.69	30.03	46.29	55.52	63.68	71.92
	32	718A1	Normal	0.9430	-	2.75	4.16	7.06	9.85	14.80	19.89	a	a	a	a	a
	32	MD101	No Fines	0.8774	-	0.74	1.12	1.79	2.56	4.74	9.14	18.89	33.62	43.05	53.16	65.58
	32	MD101	Low Porosity	0.9927	-	1.04	1.35	2.43	3.58	6.48	12.12	23.76	38.01	45.45	52.86	68.71
	32	MD101	Degassed	0.8861	-	1.37	1.94	2.99	4.17	7.18	13.33	25.77	42.68	52.38	61.74	72.47
	45	MD101	Normal	0.9474	-	1.63	2.10	2.07	4.30	7.16	14.05	27.01	43.24	56.20	70.60	81.72
	45	718A1	Normal	1.0241	-	3.45	5.43	8.91	12.28	17.95	24.55	a	a	a	a	a
	45	MD101	No Fines	0.9405	-	1.66	2.04	3.16	4.50	7.77	13.61	24.47	41.26	54.91	68.96	89.28
	45	MD101	Low Porosity	1.0407	-	1.99	2.82	4.39	6.47	10.60	18.11	31.07	48.80	61.11	71.74	85.95
	45	MD101	Degassed	0.9539	-	2.52	3.37	5.37	7.49	12.01	19.89	33.99	55.00	68.14	79.79	93.04
None	-	718A1	Normal	0.8405	-	0.66	0.80	0.83	0.86	0.85	1.09	a	a	a	a	a
None	-	MD101	Normal	0.8389	-	0.68	0.85	1.00	1.14	1.31	1.47	0.98	1.27	1.04	1.18	0.97
W	30	MD101	No Fines	0.8634	-	0.58	0.60	0.72	0.57	0.71	0.95	0.56	0.52	0.25	0.31	0.47
Number of plates used for each value				2	1	2	2	2	2	2	2	2	2	2	2	2

[a] Annealing stopped after 196 hours.

as a function of annealing time at 400°C for the regular (85% -100 + 325 mesh fuel, 15% -325 mesh fuel and balance Al matrix + porosity) fuel plates are shown in Fig. 1. Two trends are clear. First, plates containing U_3Si_2 swell at a lower rate than those containing U_3Si or U_3SiAl; second, a 45 vol % fuel plate will have a greater change of the fuel zone volume than a 32 vol % fuel plate.

The results presented here are in agreement with the trends shown by Nazaré[8] for annealing treatments at 350°C. However, they do not agree with Nazaré's data for 450°C anneals. The differences could be due to the higher temperature, compositional differences in the fuels, differences in loadings, or differences in the range of fuel particle sizes used.

The use of 718 Al as the matrix produced a twentyfold increase in the fuel zone volume growth rate after 196 h at 400°C for the 32 vol % U_3Si_2. The increase in volume for U_3Si and U_3SiAl was only 30% higher than a normal 30 vol.% plate. Metallographic examinations of the plates with the 718 Al matrix showed an increased reaction zone compared to similarly annealed MD101 Al matrix plates. Another possible cause of the increased swelling was the higher fabricated porosity in the 718 matrix fuel plates. Higher porosity produces a mechanically weaker fuel plate.

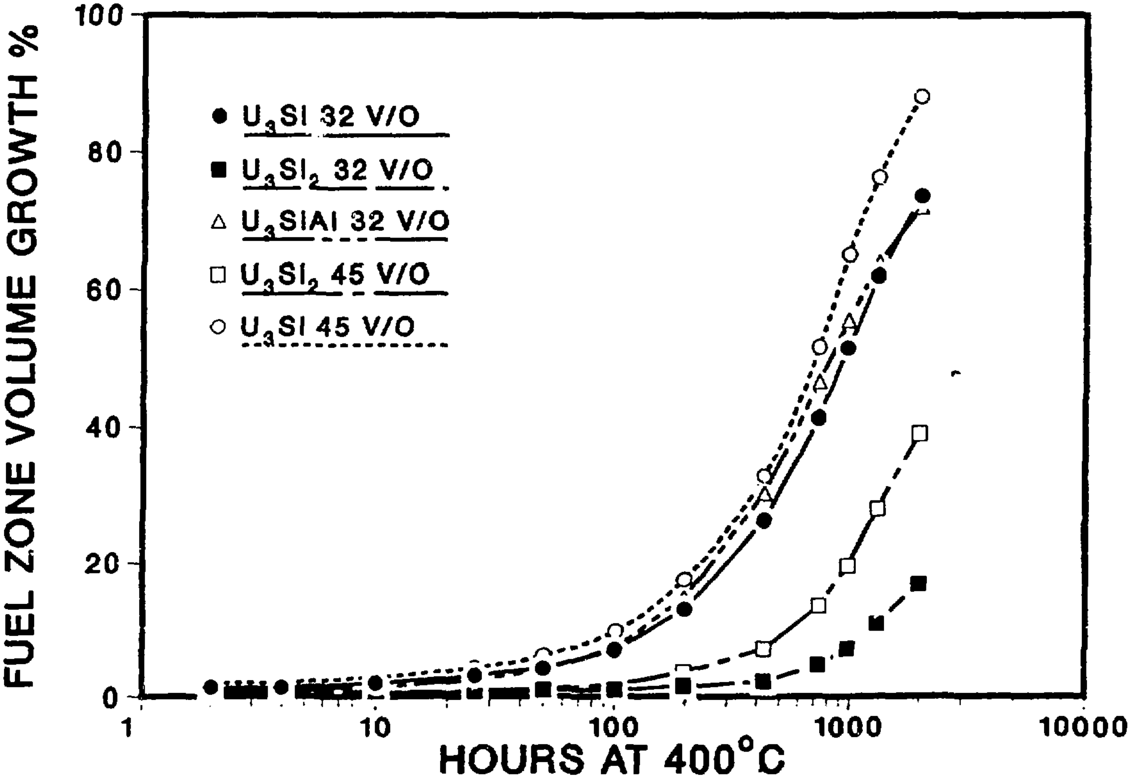

Fig. 1. Volume Percent Growth in the Fuel Zone for Uranium Silicide Fuels at 400°C.

Other variables, such as no fines, low compact porosity, and degassed compacts led to inconclusive results. The use of no fuel fines substantially reduced the swelling at 32 vol % but had little effect at 45 vol %. This could be due to a saturation in the surface area of the fuel so that the process becomes diffusion limited by the Al.

The use of a fuel compact with lower initial porosity (<5% versus 14% for normal compacts) did yield fuel plates with lower as-fabricated porosity, but no conclusive trend was seen in the thermal growth of these samples at 400°C.

The use of degassed compacts also showed no consistent trends. The authors believe that the overall thermal growth of a plate produced with a well-prepared (clean) compact will not be reduced by vacuum degassing.

Mg-MATRIX FUEL PLATES

An assembly of eight compatibility study plates (CS-131-139) was hot and cold rolled using degassed Mg powder as the fuel zone matrix. This concept was implemented with the intent of reducing the diffusion of Al into U_3Si by eliminating an Al matrix while preserving the Al base 6061 clad. Magnesium was chosen because of its low cross-section capture area for thermal neutrons and the fact that it is commercially available. The Mg-matrix plates were fabricated following procedures nominally identical to those employed for previous compatibility study samples except that the hot rolling temperature was lowered. There are two eutectics in the Al-Mg system; the lower temperature eutectic is at 437°C. In order to preclude melting, hot rolling was conducted at 385°C. The lower temperature did in fact prevent melting, but it also resulted in a non-bonded product. Hot rolling, blister annealing (also at 385°C), and cold rolling went without incident. Nevertheless, when the 1.52-mm (0.060-in.)-thick assembly was sheared into individual plates, they uniformly came apart, sometimes with no effort; in some locations the pieces of 6061 could easily be peeled apart.

A second attempt to produce Mg-matrix compatibility study plates was conducted. Two modifications of the standard practice were to increase the amount of 6061 etched away in the chemical cleaning process prior to assembly and to increase the hot rolling temperature to ∿415°C. This assembly (CS-152-155) also resulted in non-bonded plates, although there were some bonded areas.

ORIGIN OF PLATE SWELLING

In order to address the question of what causes pillowing in plates annealed for prolonged periods at temperatures >300°C, a simulated miniplate with ∿31 vol % U_3Si in the fuel zone (MD101 matrix) was chosen and heated at 500°C for a sufficient time (∿110 h) to cause pillowing. It was then cold rolled to the original thickness and reheated to 500°C. The reheated plate pillowed again after ∿6 h, suggesting that trapped gases are responsible for the fuel zone growth. If the original growth was the simple manifestation of a diffusion reaction forming less dense phases, then it should have been possible to flatten the fuel zone by cold rolling and to keep it flat (non-pillowed) even though it was reheated. Also, even a complete reaction could not account for the >75% growth in the volume of the fuel zone.

One clue to the source of this gas came from one set of the control plates. A 30-vol % W (no fines) plate (CS-40) showed no signs of swelling after nearly 2000 h at 400°C. Since there is almost no solid solubility of Al in W and the diffusion of Al in W is estimated to be very low at this temperature,[9] no detectable reaction occurred. This situation implies that gas must be released by a reaction between the fuel and the matrix.

SOURCES OF GAS

Samples of matrix powders MD101 and 718 were analyzed for H, O, and H_2O. H and O were determined using a Leco RH-1 hydrogen determinator and a Leco RO16 oxygen determinator; H_2O was estimated from weight loss measurements after drying weighed samples at 110°C in a vacuum oven for 2 h. The results are presented in Table 3.

Table 3. Gas Analysis of MD101 Al and 718 Al Powders

	H	O	H_2O	
Powder Type[a]	ppm by weight			Condition
ALCAN MD101	100	3290	56	As received
ALCAN MD101	55	1910	10	Outgassed ∿12 months before analysis
ALCAN MD101	35	3070	14	Outgassed ∿1 month before analysis
ALCOA 718	72	480	20	As received
ALCOA 718	35	420	63	Outgassed ∿1 month before analysis

[a] MD101 is minimum 99.3 wt % free metallic Al. 718 is nominally Al + 12 wt % Si + 0.5 wt % Fe.

Using the average content of 0.0045 wt % for H and 0.0012 wt % for H_2O, the possible volume of hydrogen gas from both sources at standard temperature and pressure (STP) was calculated to be ∿28.2 cm^3 H_2/compact.

GAS ANALYSIS OF PILLOWED PLATES

To confirm the presence and species of gas in pillowed plates, six plates were heated for 193-243 h at 500°C. The plate identities are given in Table 4. All six plates pillowed to some degree. After pillowing, samples CS51, CS99, and CS114 had microcracks in the cladding to the fuel zone and were not analyzed for gas content. At ~40X magnification CS44 and CS91 showed evidence of possible surface cracks. CS107 appeared to have no surface cracks. Only CS44, CS91 and CS107 were analyzed for gas content.

A brass pipe tee with a brass disk base 47.6 x 6.4 mm (1-7/8 in. diameter x 1/4 in.) with a 12.7-mm (1/2-in.) diameter hole in the center was attached to each "pillowed" plate with a quick-setting epoxy. A drill bit for puncturing the plate was inserted into the crossbar of the tee and a ball joint was attached to the leg of the tee. A 50.8 x 50.8 x 6.4-mm (2 x 2 x 1/4-in.) brass plate was epoxied to the side of the sample plate opposite the drill to ensure the integrity of the sample in case both sides of the sample plate were punctured.

After the device was secured to a sample plate, the assembly was checked for He leaks and connected to a mass spectrometer (MS) Model CEC 21-620. The air between the MS inlet and the sample plate was evacuated; pumping was continued until surface outgassing was minimal. Sample plate CS44 was pumped on for about 8.5 h, CS91 and CS107 for more than 17 h each.

The stopcocks to the MS pumps were closed after a static vacuum was obtained. The sample plate was punctured by manually rotating the Teflon-gasketed drill; the contained gas was allowed to expand into the MS manifold, where its pressure was measured on micromanometer. The spectrum from mass 2 to mass 100 of the released gas was recorded, and the percentages of the observed components were calculated; the results are given in Table 5.

By using the calculated fuel zone volumes and applying the gas law, the pressure within plate CS107 at 500°C was determined to be 1.114 kg/cm^2. This is slightly above normal atmospheric pressure (1.033 kg/cm^2), a situation that is required since a pillow would not form if the internal pressure was less than atmospheric. By using 1.114 kg/cm^2 as the pressure within any pillowed plate at 500°C, the fuel zone volumes for CS44 and CS91 were calculated for room temperature. The results are shown in Table 6.

The large differences between the calculated and measured values of the fuel zone volumes suggest that CS44 and CS91 both developed areas of the clad thin enough to allow some of the gas to diffuse out. This gas was most likely H_2, since 99.7% of the gas in CS107 is H_2 and the diffusion rate in Al for hydrogen is several orders of magnitude greater than that for oxygen or nitrogen. Once the H_2 had escaped and the H_2 partial pressures on both sides of the clad became equal, there would be no driving force to enlarge the diffusion path. The larger gas molecules would be trapped inside and outside the fuel plate. The low values measured for N_2 and O_2 and the small volume of gas present (a partial vacuum was measured) in CS44 and CS91 support this theory.

Table 4. Fuel Plates Deliberately Pillowed at 500°C

Plate No.	Fuel	Volume Percent Fuel in Meat	Anneal Time (h)	Surface Rating after Anneal
CS-44	U_3SiAl	32	193	Possible cracks
CS-51	U_3SiAl	45	193	Visible cracks
CS-91	U_3Si	32	193	Possible cracks
CS-99	U_3Si	45	193	Visible cracks
CS-107	U_3Si_2	32	193	No visible cracks
CS-114	U_3Si_2	45	243	Visible cracks

Table 5. Analysis of Gases Present in Pillowed Fuel Plates

Gas	Gas Content (μl)		
	CS-44 U_3SiAl	CS-91 U_3Si	CS-107 U_3Si_2
H_2	8.0	4.9	1196.4
CH_4	0.21	0.21	1.68
H_2O	a	a	0.36
N_2, CO	0.18	0.036	≤ 0.12
O_2	≤ 0.056	0.036	0.036
Ar	5.5	12.8	0.84
CO_2	<0.028	<0.036	<0.036
Other	ND[b]	ND[b]	ND[c]
Total reduced to STP	14.0	18.0	1200

[a] Very small amount of gas precluded reliable determination of H_2O.

[b] No other species detected; sample size limits detection to 0.2% or greater.

[c] As little as 20 ppm of C_2H_2, H_2S, or CS_2 should be detectable in this sample.

Table 6. Measured and Calculated Volumes of the Fuel Zone for Pillowed Plates

Plate No.	Fuel Zone Volume Original	Fuel Zone Volume after Anneal	ΔV at 21°C Measured by Archimedes Principle	Measured Volume of Gas at 21°C $1.033\ kg/cm^2$	Calculated Volume of Gas at 21°C and $1.114\ kg/cm^2$
	cm^3				
CS-44	0.8873	2.956	2.069	0.014	0.849
CS-51	0.9493	-	Not Measured	-	-
CS-91	0.8954	5.172	4.277	0.018	1.754
CS-99	0.9606	-	Not Measured	-	-
CS-107	0.8741	3.799	2.925	1.2	1.2
CS-114	0.9355	-	Not Measured	-	-

The conclusion is that the values for CS107 are probably most representative of the "true" gas content and that the amount of H_2 measured is less than 5% of that potentially available from the Al powder. Thus the major component of the gas is H_2, with a small amount of H_2O, N_2, O_2, and CO_2 from air entrapped in the original compact, Ar of unknown origin, and CH_4 from the possible reaction of C from the lubricant with hydrogen.

PROPOSED THEORY OF MINIPLATE SWELLING

It is proposed that the swelling of the miniplates begins with diffusion of matrix Al into the areas of contact with the fuel. Diffusion is greatest along the grain boundaries, and the resultant product is $U(Al,Si)_3$. The effect is to separate the particles as shown in Fig. 2 for U_3Si and Fig. 3 for U_3SiAl. U_3Si_2 does not react this way; a zone of reacted material encircles the U_3Si_2 fuel, forming a "kernel"-like structure (Fig. 4). The reason for this difference could be that the U_3Si_2 has fewer internal grain boundaries. U_3Si and U_3SiAl powders probably have more internal grain boundaries, resulting from the excessive "cold" work in powder production, which allow faster diffusion of the Al into the fuel. This in part would explain why the U_3Si_2 swells at a lower rate than U_3Si or U_3SiAl.

As the Al reacts with the fuel, the fuel zone volume increases due to the differences in densities and to pores produced by the Kirkendall effect. If the Al was reacting with any adsorbed H_2O, then the plate containing W-Al in the "fuel zone" should have pillowed. Since the W-Al plate did not swell, the source of the H_2 must be the hydrogen that is in solid solution in the MD101 Al powder. During the reaction this H_2 is released and fills the plate voids. When the internal pressure exceeds the restraint of the cladding, it begins to creep and finally pillows.

The presence of hydrogen in CS44, CS91, and CS107 (Table 5) supports this theory.

SUMMARY AND CONCLUSIONS

A data base for the thermal compatibility at 400°C of unirradiated uranium silicide dispersed in aluminum has been established. The cause of the growth of silicide fuel plates was determined to be a two-step process: 1) the reaction of the uranium silicide with aluminum to form a less dense product, $U(Al,Si)_3$ and 2) the release of hydrogen as a result of the reaction and the subsequent creep and pillowing of the fuel plate when the internal pressure within the fuel zone exceeds atmospheric pressure.

Fig. 2. U_3SiAl Particle Showing Reaction after 24 h at 500°C.

Fig. 3. U_3Si Particle Showing Reaction after 24 h at 500°C.

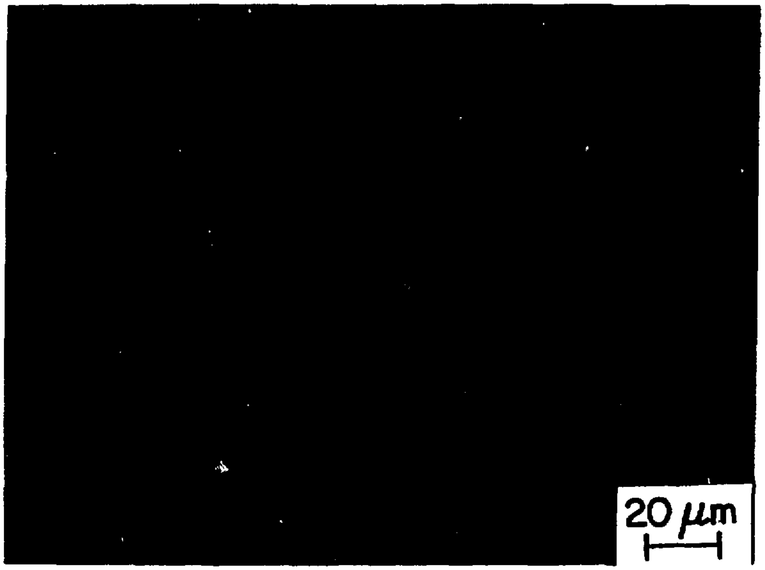

Fig. 4. U_3Si_2 Particle Showing Reaction after 24 h at 500°C.

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