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EFFECTS OF IRRADIATION ON SOME URANIUM-PLUTONIUM ALLOYS

by

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ABSTRACT

Irradiations were made on a number of uranium-plutonium alloy specimens made from both cast and extruded materials. The cast alloys included alloys of uranium with 3.7, 6.6, and 13.0 w/o plutonium, and the extruded alloys included alloys of uranium with 9.5, 14.1, and 18.7 w/o plutonium. One-half of the extruded specimens were given a heat treatment consisting of heating to 645°C and cooling to and holding at 500°C for one hour in an attempt to remove the preferred orientation that was anticipated from extrusion. The specimens were irradiated to burnups ranging up to 0.84 a/o (7100 MWD/T) with central temperatures ranging up to 490°C. The cast specimens were all found to have developed severe surface roughening as a result of the irradiation they received, presumably because of excessively large grain sizes present before irradiation. Identically fabricated unalloyed uranium specimens showed similar behavior. The as-extruded alloy specimens maintained good surface smoothness under irradiation, but showed elongations which were dependent on plutonium content. For example, in samples with 0.4 a/o burnup, a 14.1 w/o plutonium alloy specimen elongated 96%, whereas an 18.7 w/o plutonium alloy specimen elongated only 8.4%. The heat-treated extruded specimens did not elongate anisotropically, indicating that the heat treatment used was effective in randomizing the grain orientation. However, the heat-treated specimens developed excessive surface roughening, apparently because the heat treatment caused an undesirably large grain size.

INTRODUCTION

The development of fast breeder reactors appears to be indispensable if the world's reserves of U^{238} are to be utilized for the production of nuclear energy. This situation results from the fact that thermal reactors can be expected to have breeding ratios somewhat less than 1.0 where U^{238} is used as fertile material.(1) On the other hand, a suitably designed fast

reactor with a U^{238} blanket can be expected to have a breeding ratio of about 1.2 with U^{235} fuel or a ratio of about 1.5 if Pu^{239} is used as fuel.(1) Thus, a fast breeder utilizing the Pu^{239} - U^{238} cycle might supply sufficient fuel to maintain operation of several thermal power reactors, depending upon their uranium conversion ratios and power levels.

The successful development of a fast breeder utilizing the Pu^{239} - U^{238} cycle depends to a large extent upon the ability of the fuel material, which would normally contain both Pu^{239} and U^{238}, to adequately resist irradiation damage during fuel burnups ranging upward from 1 a/o while at irradiation temperature of the order of 650°C or higher.

The simplest fuel compositions would of course be binary alloys of uranium and plutonium. A few irradiations of such alloys have been made on small nickel-plated pins, 1/16 in. in diameter and 3/16 in. long, cast to size.(2) The small specimen size was dictated by the lack of a means of cooling larger specimens during irradiation. In these tests, uranium-10 w/o plutonium and uranium-15 w/o plutonium alloys were examined after burnups ranging up to 0.35 a/o. It was found that the pins suffered little damage, although the amount of restraint contributed by the nickel cladding was not known.

The availability of the MTR, with its inherent water cooling of fuel specimen capsules, has more recently enabled further irradiations to be made on uranium-plutonium alloys, using specimens of a larger size and without protective plating. The present report describes the results of irradiation tests on these specimens, which included alloys of uranium with 3.7, 6.6, 9.5, 13.0, 14.1 and 18.7 w/o plutonium. A few unalloyed uranium specimens were also irradiated for purposes of comparison. Photographs of the irradiated specimens are presented, along with data on dimensional and weight changes resulting from irradiation.

After the work described in the present report was completed, a series of irradiations were made on uranium-plutonium-molybdenum and uranium-plutonium-fissium alloys. Results from these irradiations have already been reported.(3)

SPECIMEN MATERIALS AND EXPERIMENTAL PROCEDURE

Two groups of specimens were made for the irradiation experiments. The first group consisted of specimens made by casting into a copper mold. The nominal size of each casting was 0.140 in. diameter x 1.00 in. long. After casting, the specimens were annealed at 500°C for one hour and then machined to a nominal size of 0.125 in. diameter by 0.750 in. long. An exception was specimen AA-240, which was machined to a length of 0.440 in. Compositions of the cast specimens included uranium plus 0, 3.7, 6.6, and 13.0 w/o plutonium.

The material for the second group of specimens was fabricated by casting to 3/8-in. diameter in a copper mold. The castings were then inverse extruded at temperatures from 450° to 525°C to a diameter of 0.185 in. The specimens were jacketed in copper as a lubricant and required pressures in the range of 80,000 to 120,000 psi for extrusion.

Alpha-extruded uranium shows a high degree of preferred orientation as evidenced by a low longitudinal expansion coefficient and a correspondingly high radial expansion coefficient. Dilatometric studies of the extruded uranium-plutonium alloys that were irradiated indicated that they not only had a high degree of texture, but that the degree and type of texture was greatly influenced by the plutonium content. The longitudinal expansion coefficient increased with increasing plutonium content and the radial coefficient decreased. There was some evidence that at above 10 w/o plutonium the radial coefficient became smaller than the longitudinal coefficient.

It appeared desirable, therefore, to attempt to remove, by suitable heat treatment, the preferred orientation in at least some of the extruded alloy specimens. Irradiation studies of other uranium-base alloys at Argonne National Laboratory have shown the effectiveness of isothermally transforming the alloys prior to irradiation.[4,5] Reference to the uranium-plutonium phase diagram,[6] shown in Figure 1, indicated that a temperature of 500°C would permit the alloys to transform completely from any retained beta or gamma. Therefore, one-half of the specimens were then given a heat treatment consisting of heating to 645°C, cooling to and holding at 500°C for one hour, and cooling to room temperature. Both the as-extruded and the heat-treated specimens were then machined to a nominal size of 0.120 in. diameter x 0.750 in. long. Compositions of the extruded specimens included uranium plus 0, 9.5, 14.1, and 18.7 w/o plutonium.

After machining to size, measurements were made on all specimens for dimensions, weight, and hardness. Densities were determined by loss of weight in 1, 2, 3-tribromopropane. Pre-irradiation hardness and density values are shown in Table I. A typical specimen, before irradiation, is shown in Figure 2.

The specimens were then placed in zirconium irradiation capsules containing NaK for heat transfer purposes.[7] An assembled capsule is shown diagrammatically in Figure 3. A small wire of an aluminum-cobalt-manganese alloy was also included in each capsule to permit determination of the thermal flux incident on each capsule. Irradiation procedures and thermal flux determinations using capsules of this type have been previously described.[8] In calculating the atom burnup in the specimens, appropriate corrections were made to the thermal flux measurements to compensate for flux depression near and within the fuel specimen.

EXPERIMENTAL RESULTS AND DISCUSSION

Tables II and III summarize the conditions under which the specimens were irradiated. Also shown in the tables are dimensional and weight changes resulting from irradiation. Photographs of the irradiated specimens are shown in Figures 4 through 11.

Cast Material

As shown in Figures 4 through 7, all the cast specimens showed extensive surface roughening as a result of irradiation. Apparently, the amount of surface roughening was not directly related to the amount of plutonium in the alloys. Thus, even the unalloyed uranium specimens showed severe roughening, although they were irradiated to comparatively lower burnups.

The type of surface damage exhibited by the cast specimens is characteristic of coarse-grained material. For example, similar surface roughening was noted in a group of uranium-chromium alloy castings known to be coarse-grained.(9) Since the surfaces of the alloyed specimens were as badly damaged as those of the unalloyed specimens, it may be concluded that the additions of plutonium to uranium did not effect a reduction in grain size.

The changes in length and diameter shown in Table II are considered to be due almost entirely to surface roughening. As might be expected in cast material, the dimensional changes resulting from irradiation appeared to be essentially isotropic. The large weight losses noted in some of the specimens are attributed to incomplete recovery of specimen fragments from irradiation capsules, rather than to corrosion by NaK.

One specimen, No. AA-236, obviously increased considerably in volume. This specimen was calculated to have been irradiated at 440°C, the highest temperature of the group. It is believed that the large volume increase in this specimen resulted from the mechanism known as "swelling." This phenomenon is believed to be due to the nucleation and growth at elevated temperatures of bubbles of volatile fission products.(10) It may be noted that uranium specimens used in another investigation(11) were observed to begin swelling at the nearly identical temperature of 430°C.

Although it is not known why the present cast specimens were much more severely damaged by irradiation than the small pins described in reference 2, possible explanations for the greater stability of the small pins are that the smaller size of the pins would presumably have resulted in a faster cooling rate with a correspondingly finer grain size; also, the nickel plating on the pins may have given some degree of restraint.

Extruded Material

As mentioned earlier, one-half of the extruded specimens were given a heat treatment consisting of heating to 645°C, cooling to and holding at 500°C for one hour, and cooling to room temperature. The heat treatment greatly affected the behavior of the extruded alloys under irradiation, as shown in Table III and more clearly in Figures 8 through 11.

Considering, first, the as-extruded specimens, it was noted that whereas the unalloyed specimens shortened under irradiation, all of the alloyed specimens which could be completely removed from their irradiation capsules were found to have elongated. Thus, confirming the thermal expansion results, it appears that a different type of preferred orientation is present in alpha-extruded uranium-plutonium alloys from at least approximately 10 to 20 w/o plutonium than is present in unalloyed uranium.

In contrast to the performance of the castings described earlier, the as-extruded specimens maintained excellent surface smoothness during irradiation, indicating that extrusion effectively reduced the grain size present in the original cast billets.

The general lack of surface roughening on the irradiated as-extruded specimens enabled the irradiation growth rate, G_i, resulting from preferred orientation, to be calculated and is shown in Table III. The irradiation growth rate is calculated from the relation(8)

$$G_i = \frac{1}{L}\frac{dL}{dN} = \frac{\ln(L/L_o)}{\text{fraction of total atoms fissioned}} = \frac{\text{microin./in.}}{\text{fissions}/10^6 \text{ total atoms}}$$

where

L_o = initial length

L = final length

N = unit of atom burnup .

The above expression thus takes into account the fact that the amount of growth at any given level of burnup is dependent upon the instantaneous length of the specimen at that time.

Growth rates of the as-extruded uranium - 9.5 w/o plutonium alloy specimens could not be obtained because they could not be completely removed from their irradiation capsules. However, growth rates of the 14.1 and 18.7 w/o plutonium alloy specimens showed the latter alloy to be comparatively stable, with a G_i value of about 23. This is comparable to that observed in uranium stabilized by being quenched from the beta phase,(8) with the exception that beta-quenched uranium develops a greater degree of surface roughening.

The surface roughening observed in the extruded and heat-treated specimens was even more severe than that noted in the cast specimens, with the exception of the 18.7 w/o plutonium alloy. Based on the appearance of the irradiated specimens, it may be concluded that the heat treatment produced an unfavorably large grain size in the alloys. However, judging from the 18.7 w/o plutonium alloy specimens shown in Figure 11, the heat treatment appears to have accomplished its intended purpose in randomizing the texture resulting from fabrication.

Results from the extruded group of specimens indicated that resistance to irradiation damage was related to plutonium content. Thus, irradiated specimens in the lowest alloy group, which contained 9.5 w/o plutonium, were so badly damaged that most of them could not be completely removed from their irradiation capsules. On the other hand, the specimens in the highest alloy group, which contained 18.7 w/o plutonium, were noticeably better in comparison to the other alloys. The as-extruded specimens containing 18.7 w/o plutonium also showed the lowest anisotropic growth rates and the heat-treated specimens of the same alloy showed the least amount of surface roughening.

CONCLUSIONS

1. Uranium-plutonium alloy specimens containing up to at least 13 w/o plutonium and prepared by casting into copper molds are subject to severe irradiation damage which manifests itself as excessive surface roughening.

2. Uranium-plutonium alloy specimens containing up to at least 19 w/o plutonium and prepared by extrusion in the alpha phase elongate under irradiation but maintain good surface smoothness.

3. The preferred orientation present in uranium-plutonium alloys extruded in the alpha phase can be removed by suitable heat treatment, but with the particular treatment used in this investigation, severe surface roughening resulted in subsequent irradiations.

4. Although the effects of irradiation on cast uranium-plutonium alloys showed no particular dependence on alloy compositions, the extruded alloys showed better resistance to irradiation damage with increasing plutonium content.

5. If further studies of the effects of irradiation on uranium-plutonium alloys are undertaken, it is recommended that a range of compositions extending upward from about 18 w/o plutonium be investigated. In particular, further studies are indicated of the relatively good performance of the extruded U-18.7 w/o plutonium alloy.

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TABLE I

Pre-irradiation Density and Hardness Measurements
On Uranium-Plutonium Alloy Specimens

Spec. No.	Pu, w/o[a]	Condition[b]	Density, gm/cc	Hardness, R_A[c]
AA-229	0	C	18.89	58.6
AA-230	0	C	18.89	57.5
AA-233	0	C	18.86	57.3
AA-231	3.7	C	18.81	61.1
AA-232	3.7	C	18.84	60.9
AA-234	3.7	C	18.88	61.6
AA-235	6.6	C	18.92	60.7
AA-236	6.6	C	17.94	61.2
AA-237	6.6	C	18.70	59.9
AA-238	13.0	C	18.75	62.3
AA-239	13.0	C	18.72	60.4
AA-240	13.0	C	18.78	59.6
BG-1	0	E	18.82	-
BG-2	0	E	18.90	-
BG-3	9.5	E	18.81	-
BG-4	9.5	E	18.71	-
BG-5	9.5	HT	18.88	-
BG-6	9.5	HT	18.86	-
BG-7	14.1	E	18.83	-
BG-8	14.1	E	18.87	-
BG-9	14.1	HT	18.80	-
BG-10	14.1	HT	18.78	-
BG-11	18.7	E	18.78	77.4
BG-12	18.7	E	18.80	-
BG-13	18.7	HT	18.80	-
BG-14	18.7	HT	18.80	-

[a] Balance is natural uranium

[b] C - Cast and annealed 1 hour at 500°C
E - Extruded at 450-525°C
HT - Extruded at 450-525°C, heated to 645°C, cooled to and held at 500°C for 1 hour, cooled to room temperature

[c] Hardness measurements were not made on most of the extruded specimens because of excessive brittleness of the alloys.

TABLE II

Effect of Irradiation on Cast Uranium-Plutonium Alloy Specimens

Spec. No.	Pu, w/o	Burnup, a/o	Irrad. Temp., °C[a]		Length Change, %	Diameter Change, %	Weight Change, mg
			Surface	Center			
AA-233	0	0.097	85	95	6.7	0 to 6.4	- 2.6
AA-229	0	.11	90	100	0	3.2 to 46	- 4.1
AA-230	0	.12	95	105	0	-6.4 to + 26	- 2.3
AA-234	3.7	.32	160	200	6.1	3.2	3.8
AA-232	3.7	.43	200	240	5.2	0 to 20	.8
AA-231	3.7	.66	280	340	5.6	0 to 33	3.1
AA-237	6.6	.42	170	200	7.4	0 to 14	-242.9
AA-235	6.6	.72	310	380	(b)	-	-
AA-236	6.6	.82	340	440	19.5	66 to 114	-532.3
AA-240	13.0	.32	150	190	17.5	20	3.1
AA-238	13.0	.68	280	350	12.0	6.4 to 54	- 15.1
AA-239	13.0	.84	330	430	14.6	0 to 94	-588.9

[a]Temperatures were calculated on the basis of conditions existing at the beginning of exposure.

[b]Disintegrated under irradiation.

TABLE III

Effect of Irradiation on Extruded Uranium-Plutonium Alloy Specimens

Spec. No.	Pu, w/o	Condition [a]	Burnup, a/o	Irrad. Temp., °C[b]		Length Change, %	Growth Rate, G_i[c]	Diameter Change, %	Weight Change, mg
				Surface	Center				
BG-1	0	As Extruded	0.044	90	100	-3.82	- 87	3.12	0.1
BG-2	0	As Extruded	.048	95	105	-4.89	-102	3.76	.1
BG-4	9.5	As Extruded	.21	220	260	(d)	-	-	-
BG-3	9.5	As Extruded	.43	390	490	(d)	-	-	-
BG-6	9.5	Heat Treated	.16	170	210	1.20	-	0 to 65	-
BG-5	9.5	Heat Treated	.27	270	330	(e)	-	-	-
BG-7	14.1	As Extruded	.22	220	270	65	230	-54 to -11	-
BG-8	14.1	As Extruded	.43	390	490	96	160	-34 to 0	15.7
BG-10	14.1	Heat Treated	.12	140	170	58	-	65 to 140	-8.8
BG-9	14.1	Heat Treated	.42	380	480	42	-	6 to 97	-
BG-12	18.7	As Extruded	.28	270	350	6.7	23	0	-
BG-11	18.7	As Extruded	.37	360	460	8.4	22	0 to 22	-
BG-14	18.7	Heat Treated	.15	160	200	13	-	0 to 32	-7.6
BG-13	18.7	Heat Treated	.35	320	410	6.7	19	0 to 5.6	-

[a]Extrusion was at 450 to 525°C. Heat Treatment consisted of heating to 645°C, cooling to and holding at 500°C for 1 hour, and cooling to room temperature.

[b]Temperatures were calculated on the basis of conditions existing at the beginning of exposure.

[c]G_i = microin./in. per fission/10^6 total atoms.

[d]Specimen could not be entirely removed from irradiation capsule for measurements.

[e]Specimen ignited and burned before measurements could be made.

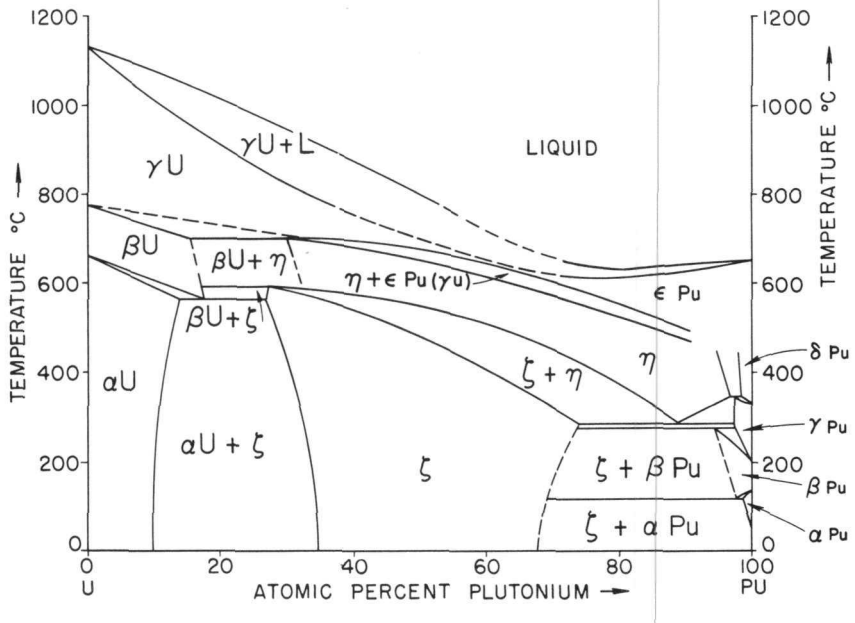
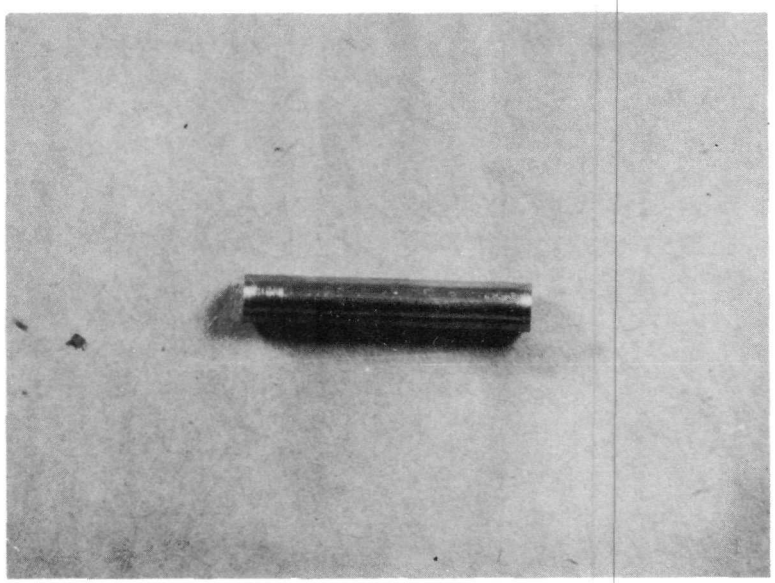


Figure 1. Uranium-plutonium phase diagram, after Los Alamos.



17,225 2X

Figure 2. Typical specimen before irradiation.

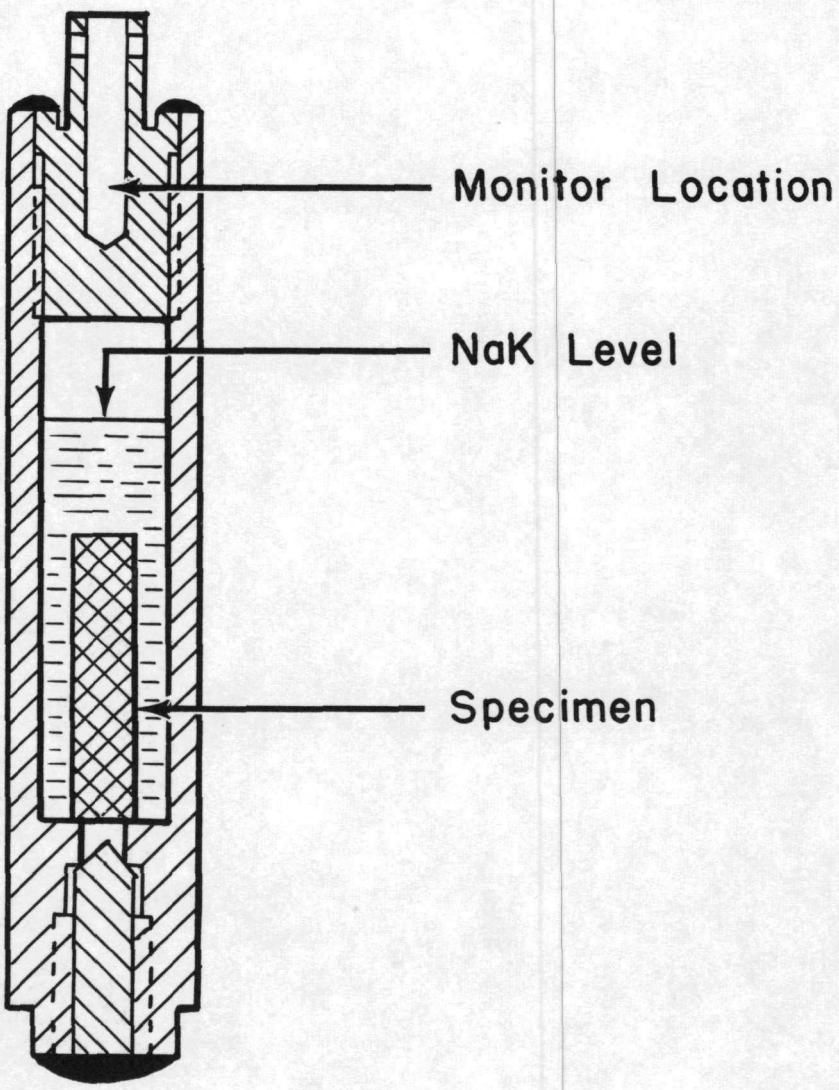


Figure 3. Assembled irradiation capsule. Length of the capsule is approximately 3 in. and the diameter is 1/2 in.

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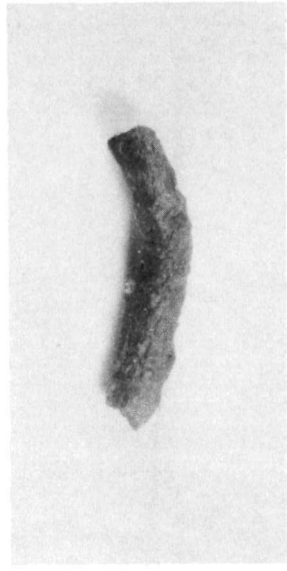
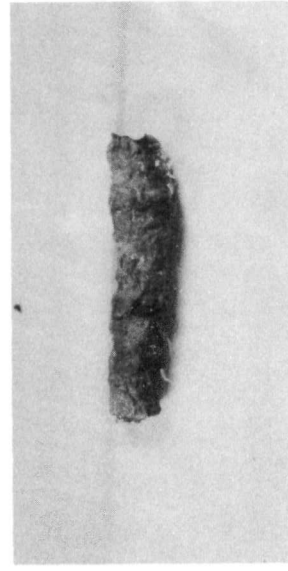
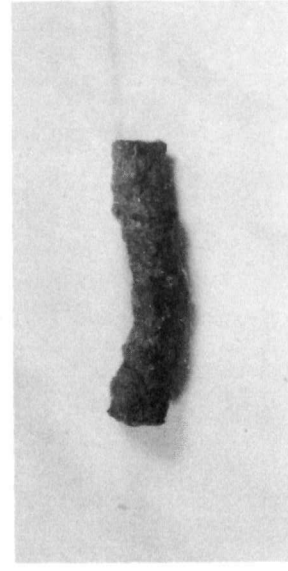
			
Specimen No.	AA-233	AA-229	AA-230
Max. Irrad. Temp., °C	95	100	105
Burnup, a/o	0.097	0.11	0.12

Figure 4. Effect of irradiation on cast unalloyed uranium. Magnification, 2X.

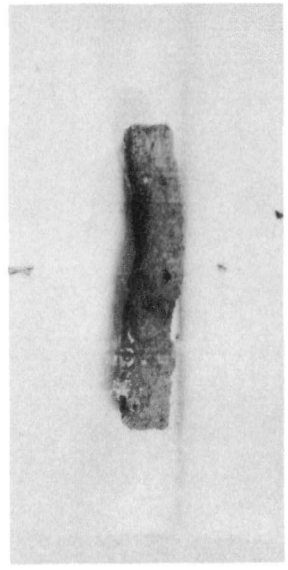
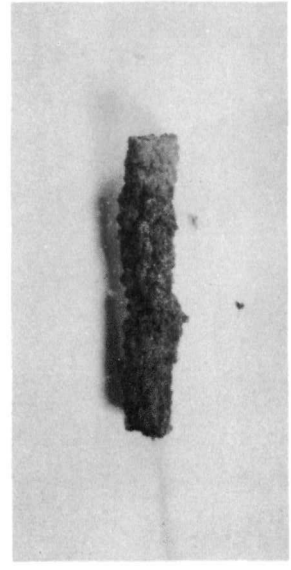
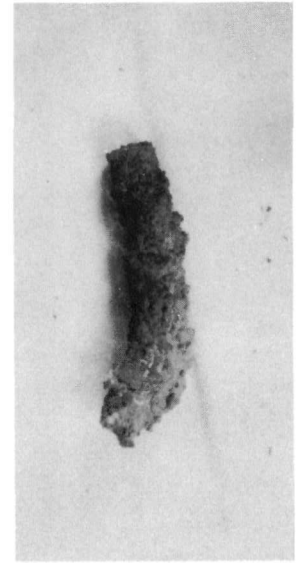
Photo No.	17,210	17,208	17,207
			
Specimen No.	AA-234	AA-232	AA-231
Max. Irrad. Temp., °C	200	240	340
Burnup, a/o	0.32	0.43	0.66

Figure 5. Effect of irradiation on cast uranium-3.7 w/o plutonium alloy. Magnification, 2X.

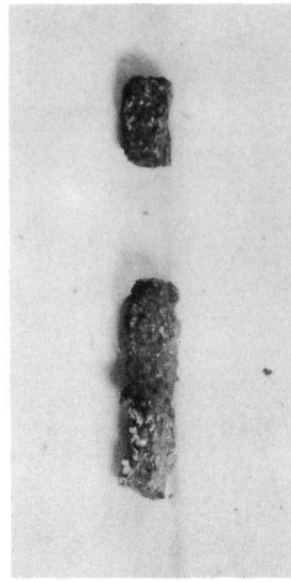
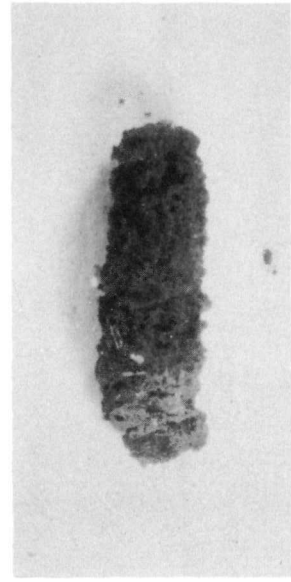
Photo No.	17,212	17,211
		
Specimen No.	AA-237	AA-236
Max. Irrad. Temp., °C	200	440
Burnup, a/o	0.42	0.82

Figure 6. Effect of irradiation on cast uranium-6.6 w/o plutonium alloy. A third specimen, No. AA-235, disintegrated during irradiation to 0.72 a/o burnup at 380°C. Magnification, 2X.

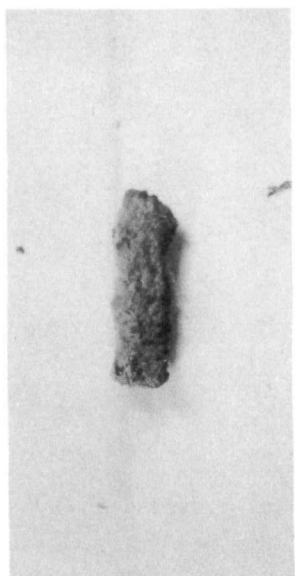
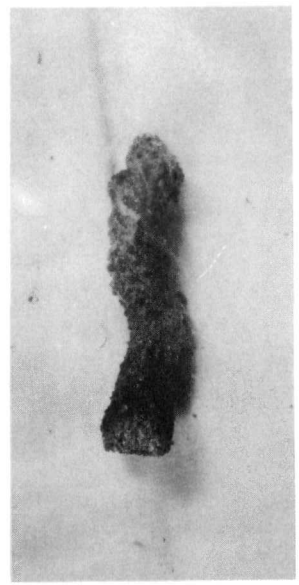
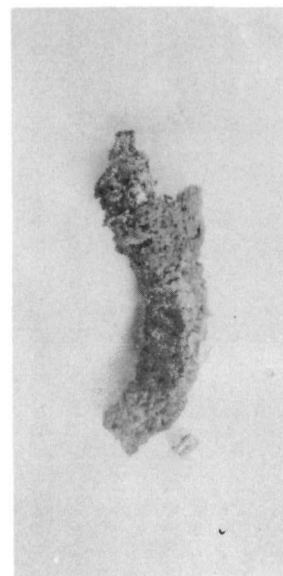
Photo No.	17,215	17,213	17,214
			
Specimen No.	AA-240	AA-238	AA-239
Max. Irrad. Temp., °C	190	350	430
Burnup, a/o	0.32	0.68	0.84

Figure 7.

Effect of irradiation on cast uranium-13.0 w/o plutonium alloy. The length of specimen AA-240 before irradiation was only 0.440 in. compared to the 0.750 in. length of the other two specimens. Magnification, 2X.

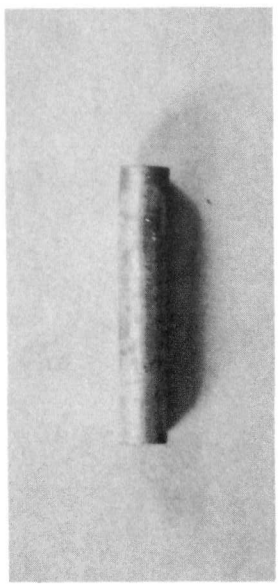
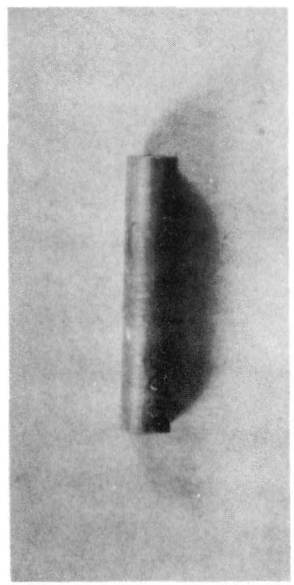
Photo No.	17,663	17,664
		
Specimen No.	BG-1	BG-2
Max. Irrad. Temp., °C	100	105
Burnup, a/o	0.044	0.048

Figure 8. Effect of irradiation on as-extruded unalloyed uranium. Magnification, 2X.

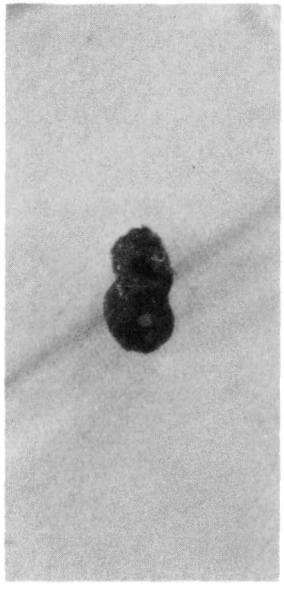
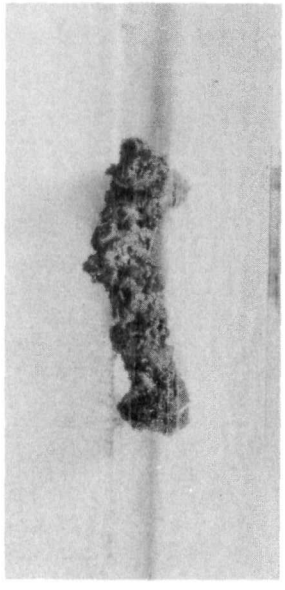
Photo No.	17,665	18,740
		
Specimen No.	BG-3	BG-6
Condition	As-Extruded	Heat Treated
Max. Irrad. Temp., °C	490	210
Burnup, a/o	0.43	0.16

Figure 9. Effect of irradiation on extruded uranium-9.5 w/o plutonium alloy. Part of specimen BG-3 could not be removed from its irradiation capsule. Magnification, 2X.

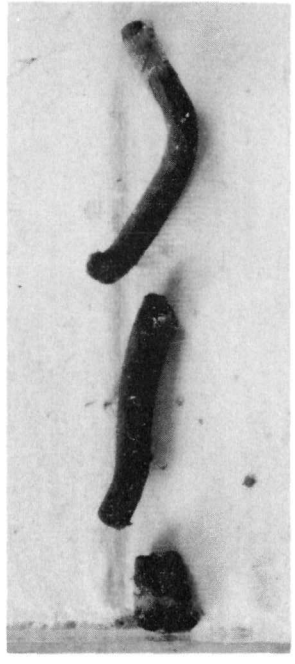
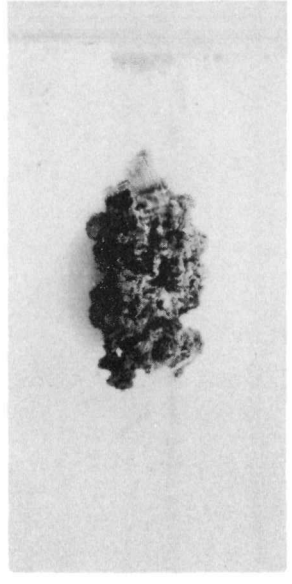
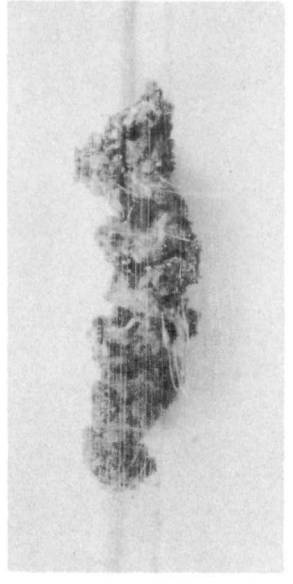
Photo No.	18,741	17,666	17,667	18,742
				
Specimen No.	BG-7	BG-8	BG-10	BG-9
Condition	As Extruded	As Extruded	Heat Treated	Heat Treated
Max. Irrad. Temp., °C	270	490	170	480
Burnup, a/o	0.22	0.43	0.12	0.42

Figure 10. Effect of irradiation on extruded uranium-14.1 w/o plutonium alloy. Magnification, 2X.

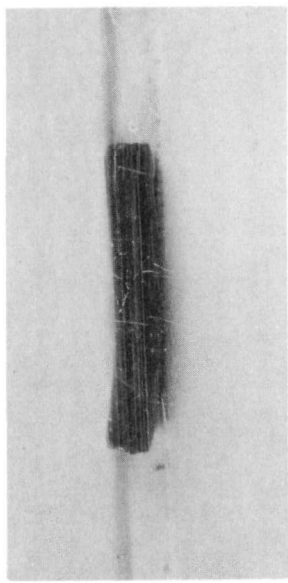
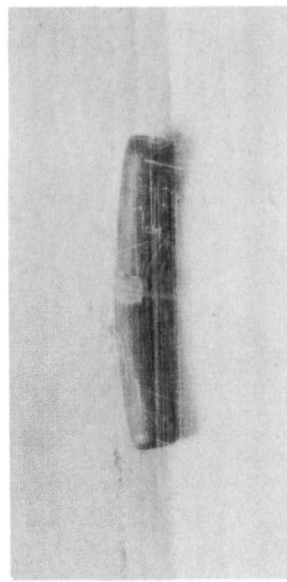
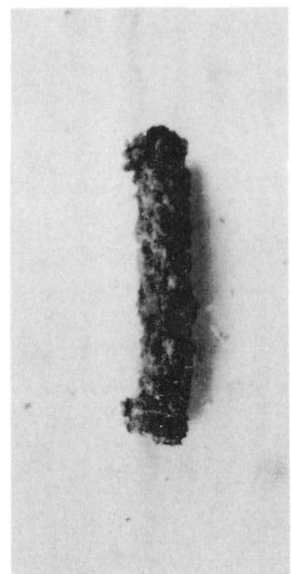
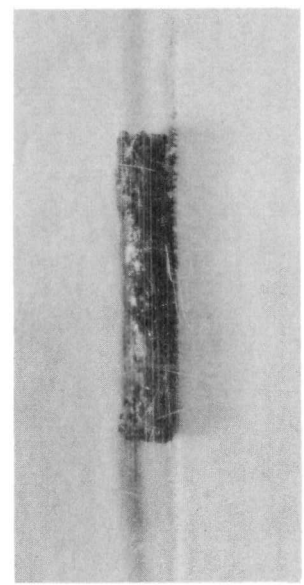
Photo No.	18,744	18,743	17,668	18,745
				
Specimen No.	BG-12	BG-11	BG-14	BG-13
Condition	As Extruded	As Extruded	Heat Treated	Heat Treated
Max. Irrad. Temp., °C	350	460	200	410
Burnup, a/o	0.28	0.37	0.15	0.35

Figure 11. Effect of irradiation on extruded uranium-18.7 w/o plutonium alloy. Magnification, 2X.