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EFFECTS OF HIGH BURNUP AT ELEVATED TEMPERATURES
ON URANIUM-0.52 AND 1.62 w/o ZIRCONIUM ALLOYS

by

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EFFECTS OF HIGH BURNUP AT ELEVATED TEMPERATURES ON URANIUM-0.52 AND 1.62 w/o ZIRCONIUM ALLOYS

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ABSTRACT

An investigation of the effects of irradiation on uranium containing 0.52 and 1.62 w/o zirconium was made. Specimens of wrought material, variously heat treated, were studied as well as castings of the alloys. It was found that, although the wrought and heat-treated alloys were reportedly stable under thermal cycling, they elongated rapidly under irradiation. The best material studied was the 1.62% alloy in the as-cast condition. It was found to increase in length initially at a rate of about 5% per a/o burnup, after which the rate steadily diminished. Up to burnups of at least 5.3 a/o it retained a smooth surface. Two specimens of the cast 1.62% alloy, one irradiated to 2.1 a/o burnup at a calculated central temperature of 690°C and the other irradiated to 5.3 a/o burnup at a calculated central temperature of 620°C, swelled and increased in volume with the formation of a large central void surrounded by highly porous metal. The critical swelling temperature of uranium-1.62 w/o zirconium alloy was found to lie between 300 and 620°C, with some indications that it may be near 500°C.

INTRODUCTION

One of the chief problems associated with the operation of nuclear reactors in which uranium metal fuel elements are used is the dimensional instability which uranium exhibits when subjected to irradiation. Under irradiation, uranium has been observed to develop pronounced changes in length and considerable surface roughening. In general, the changes that are noted are found to proceed continuously with increasing irradiation and burnup of the fissionable atoms.

It has been found that, by quenching alpha-rolled uranium from the beta phase, the stability of the material under irradiation may be increased considerably.⁽¹⁾ Quantitative measurements made of the growth rate of beta-quenched uranium,⁽²⁾ however, show that it is still too unstable for general application in reactors in which it is desired to achieve burnups beyond 1 a/o, or approximately 10,000 MWD/T.

It has also been shown that uranium can be stabilized both under irradiation and under thermal cycling and the grain size refined by small additions of zirconium.⁽³⁻⁶⁾ The results of these early experiments, in which the irradiations were made on small pins and wafers, were of sufficient promise so that early in 1953 it was decided to make a more detailed investigation using irradiation specimens of sufficient size to permit accurate measurements of changes in dimensions, weight, density, and hardness. The purposes of the more detailed investigation were to determine the best composition and fabrication procedure and the ability of the optimum material to withstand burnups of several atom percent. Irradiations on specimens representing two different uranium-zirconium alloys were completed in July 1954, and the results obtained form the subject of this report.

SPECIMEN MATERIALS

As the presence of alloying atoms in a reactor core is closely connected with the problems of reactor economics, it was desired to irradiate alloys containing the lowest practicable concentration of zirconium. The choice of compositions was guided by the earlier irradiation and thermal cycling tests,^(4,6) which seemed to indicate that thermal cycling reactions were probably quite similar to those that might be expected from neutron irradiation.

The earlier irradiation and thermal cycling experiments had shown that 2 w/o zirconium in uranium was sufficient to increase both the dimensional and surface stability of uranium. This alloy was the most dilute that had been tested under irradiation, but compositions less than 1 w/o zirconium had been tested under thermal cycling. The thermal cycling tests had shown that, although additions of less than 1 w/o zirconium to uranium appeared to improve dimensional stability, they were not as effective as 2 w/o zirconium in eliminating surface roughening. Accordingly, two alloy concentrations, nominally 1 and 2 w/o zirconium, were selected for the new irradiations. In order to be able to obtain total atom burnups of several percent, the specimens were enriched 10% in U^{235} . The nominal specimen size was 1/8 in. in diameter and 3/4 in. long. Calculations indicated that specimens of this size and enrichment would not be subject to excessively large self-shielding and flux-depression effects. As shown in Appendix B, the combined self-shielding and flux-depression factor was found to be 0.810.

Heat treatments selected for the wrought specimens were based on the thermal cycling tests mentioned earlier, in which a number of variations were used.⁽⁶⁾ These experiments had indicated that the 1 w/o alloy would be most stable when given a long anneal in the beta phase and quenched. Specimens of the 1 w/o alloy were, therefore, prepared with this heat treatment, along with specimens quenched from the gamma phase and with as-cast material. For the 2 w/o alloy, the cycling tests indicated that either

slow cooling from the gamma phase, or an interrupted quench in which the alloy was allowed to transform at 500°C, would be a suitable heat treatment. Irradiation specimens for this study were therefore prepared with both heat treatments. As-cast 2 w/o alloy specimens were also prepared because the cycling tests and earlier pin irradiations had both shown that castings of this material were relatively stable.

It was also desired to compare the results of irradiations on the as-cast alloy specimens with those obtained on identical unalloyed specimens. Although, as mentioned earlier, information was available on the effects of irradiation on wrought and heat-treated unalloyed uranium, comparable data was not available on the irradiation behavior of cast material. Accordingly, 10%-enriched unalloyed specimens were also prepared in the as-cast condition.

A total of 36 specimens, including both alloyed and unalloyed material, were prepared for the investigation. Table I summarizes the fabrication and heat treatment of the specimens. A more detailed account is given in Appendix A, as well as chemical and isotopic analyses. The chemical analyses disclosed that the actual zirconium contents of the nominal 1 and 2 w/o alloys were 0.52 and 1.62 w/o, respectively.

EXPERIMENTAL PROCEDURE

The following properties of the alloy specimens (see Fig. 2) were measured before irradiation:

1. dimensions, measured to 0.0001 in.;
2. weight, measured to 0.1 mg;
3. density, measured to 0.05% by immersion in CCl_4 ; and
4. hardness, measured on the Rockwell A scale.

The same measurements, with exceptions for weight and density, were taken on the unalloyed specimens.

In order for the irradiation to provide a maximum of useful data it was considered essential that the fuel specimens be exposed in the reactor in such a manner as to be initially, at least, free of all physical constraint. Any dimensional or surface instability which the specimen might develop under irradiation would thus be able to proceed at its natural, unrestrained rate. Accordingly, a zirconium irradiation capsule,⁽⁷⁾ with interior dimensions such that the specimen could freely move about, was designed. In order to make use of the high thermal flux available in the MTR it was also necessary to provide an efficient means of transferring heat developed in the specimen to the process water flowing past the capsule. This was

TABLE I

Summary of Fabrication and Heat Treatment of Specimens
(For details, refer to Appendix A)

Irradiation Specimen No.	Fabrication Specimen No.	Zr, w/o	Fabrication	Heat-treatment Designation	Heat Treatment
AA-225	Y16F-98B-1	0	Cast	A	None
AA-226	Y16F-98B-2	0	Cast	A	None
AA-227	Y16F-98B-3	0	Cast	A	None
AA-228	Y16F-98B-4	0	Cast	A	None
AA-1	16F-69-A	0.52	Cast	A	None
AA-2	16F-69-B	0.52	Cast	A	None
AA-3	16F-69-C	0.52	Cast	A	None
AA-4	16F-69-D	0.52	Cast	A	None
AA-5	16F-69-E	0.52	Cast	A	None
AA-6	16F-69-F	0.52	Cast	A	None
AA-7	16F-69-G	0.52	Swaged	B	725°C for 1 hr; water quenched
AA-8	16F-69-H	0.52	Swaged	B	Same
AA-9	16F-69-I	0.52	Swaged	B	Same
AA-10	16F-69-J	0.52	Swaged	B	Same
AA-12	16F-69-L	0.52	Swaged	B	Same
AA-36	16F-69-K	0.52	Swaged	B	Same
AA-13	16F-69-M	0.52	Swaged	C	800°C for 1 hr; water quenched
AA-14	16F-69-N	0.52	Swaged	C	Same
AA-15	16F-69-O	0.52	Swaged	C	Same
AA-16	16F-69-P	0.52	Swaged	C	Same
AA-17	16F-70-1	1.62	Cast	A	None
AA-18	16F-70-2	1.62	Cast	A	None
AA-19	16F-70-3	1.62	Cast	A	None
AA-20	16F-70-4	1.62	Cast	A	None
AA-21	16F-70-5	1.62	Cast	A	None
AA-22	16F-70-6	1.62	Cast	A	None
AA-23	16F-70-7	1.62	Swaged	D	800°C for 1 hr; air cooled
AA-24	16F-70-8	1.62	Swaged	D	Same
AA-25	16F-70-9	1.62	Swaged	D	Same
AA-26	16F-70-10	1.62	Swaged	D	Same
AA-27	16F-70-11	1.62	Swaged	D	Same
AA-28	16F-70-13	1.62	Swaged	E	800°C for 1 hr; 500°C for 1 hr; air cooled
AA-29	16F-70-14	1.62	Swaged	E	Same
AA-30	16F-70-15	1.62	Swaged	E	Same
AA-32	16F-70-17	1.62	Swaged	E	Same
AA-35	16F-70-16	1.62	Swaged	E	Same

accomplished before final closure by filling each capsule with sufficient eutectic NaK alloy to fully cover the specimen. A drawing of an assembled capsule and specimen is shown in Figure 1.

106-1760

Figure 1. Vertical section of an assembled irradiation capsule; the outer diameter of the capsule is 0.50 in.

14,625

2X

Figure 2. Appearance of a typical specimen before irradiation.

The 36 capsules were irradiated in groups stacked as vertical columns in MTR reflector A-pieces. Aluminum A-pieces were used, instead of the standard beryllium pieces normally present, as special small-diameter channels were required to accommodate the capsules. Details of location and exposure in the reactor are given in Table II.

In addition to an experimental specimen, each capsule also contained a thermal-flux monitor consisting of a few milligrams of aluminum-0.5 w/o cobalt-0.5 w/o manganese alloy, located as shown in Figure 1. The Co^{60} activity induced in the monitor was counted in solution using a scintillation spectrometer. Corrections for resonance activation of the cobalt were included. By interpolating between flux values obtained from adjacent monitors in the column of capsules it was possible to determine the relative thermal flux to which each specimen was exposed.

Temperatures existing in the specimens during irradiation were calculated from the flux determination, using conventional heat-transfer equations for cylindrical geometries.

Actual burnup determinations were made on four selected specimens, using two different methods. First, samples of each of the four specimens were subjected to chemical analyses for uranium and Cs^{137} to

TABLE II

Irradiation History of Specimens

Specimen No.	Date Inserted in MTR	Location	Date Removed from MTR	Actual Irradiation Time, days
AA-1	7-27-53	A-4	1-17-54	150
AA-2	7-27-53	A-4	5-19-54	251
AA-3	7-27-53	A-4	1-17-54	150
AA-4	7-27-53	A-6	9-24-53	52
AA-5	7-27-53	A-6	11-5-53	90
AA-6	7-27-53	A-6	8-26-53	26
AA-7	7-27-53	A-4	1-17-54	150
AA-8	7-27-53	A-4	1-17-54	150
AA-9	7-27-53	A-4	1-17-54	150
AA-10	7-27-53	A-6	9-24-53	52
AA-12	7-27-53	A-6	11-5-53	90
AA-13	7-27-53	A-4	1-17-54	150
AA-14	7-27-53	A-4	1-17-54	150
AA-15	7-27-53	A-6	9-24-53	52
AA-16	7-27-53	A-6	8-26-53	26
AA-17	7-27-53	A-4	1-17-54	150
AA-18	7-27-53	A-4	5-19-54	251
AA-19	7-27-53	A-4	1-17-54	150
AA-20	7-27-53	A-6	9-24-53	52
AA-21	7-27-53	A-6	11-5-53	90
AA-22	7-27-53	A-6	8-26-53	26
AA-23	7-27-53	A-4	1-17-54	150
AA-24	7-27-53	A-4	1-17-54	150
AA-25	7-27-53	A-6	9-24-53	52
AA-26	7-27-53	A-6	8-26-53	26
AA-27	7-27-53	A-4	5-19-54	251
AA-28	7-27-53	A-4	5-19-54	251
AA-29	7-27-53	A-4	1-17-54	150
AA-30	7-27-53	A-6	9-24-53	52
AA-32	7-27-53	A-6	8-26-53	26
AA-35	7-27-53	A-4	5-19-54	251
AA-36	7-27-53	A-6	8-26-53	26
AA-225	6-9-54	A-7	7-28-54	A-35
AA-226	6-9-54	A-7	7-28-54	A-35
AA-227	6-9-54	A-7	7-28-54	A-35
AA-228	6-9-54	A-7	7-28-54	A-35

determine the atom burnup.⁽⁸⁾ In the second method, uranium samples from the four specimens were analyzed mass-spectrographically, and from the changes in isotopic composition resulting from irradiation it was possible to compute the atom burnup (see Appendix B).

Burnup values determined from the isotopic analyses were found to be more reliable and were normalized to the flux-monitor determinations to obtain burnups for all specimens. The burnups reported refer to the volume-averaged burnup of all atoms in the specimen. An earlier account,⁽⁹⁾ in which some of the preliminary results were published, reports surface burnups which were, of course, significantly higher than the average value throughout the interior of the specimens.

The chemical sampling was done by use of a solution of 12M HNO_3 and 0.6M HF, saturated with $\text{Al}(\text{NO}_3)_3$. The composition was designed to lessen the explosion hazard that results when finely divided UZr_2 is dispersed in nitric acid.⁽¹⁰⁾

IRRADIATION RESULTS

Although 36 specimens were irradiated, only 23 were examined subsequently. Examination of the remaining 13 was rejected on the basis of experience obtained as the irradiations proceeded.

Results on castings of the 1.62% alloy soon showed that they were much superior to the other materials being studied, and attention was therefore concentrated on this material. Moreover, the amount of troublesome air-borne activity released by the capsules containing specimens with higher burnups was such that it seemed advisable to limit the number of specimens of a given material to be examined, once the trend of its behavior under irradiation was established. This latter point was emphasized when one highly irradiated specimen, No. AA-15, spontaneously ignited and burned to oxide before measurements could be made and the specimen photographed. The decision to limit examinations of highly irradiated specimens where not definitely needed was based largely on this occurrence.

Table III summarizes the conditions under which the 23 examined specimens were exposed in the reactor and the changes in the specimens which were observed after irradiation. Changes in hardness are not included because reliable measurements of hardness could not be made; most of the irradiated specimens were too roughened or distorted. Furthermore, most of the relatively undamaged specimens which were subjected to hardness tests fractured when the major load was applied to the indenter. Dimensional changes in badly distorted or roughened specimens were determined by means of photographs rather than from micrometer measurements.

TABLE III

Summary of Irradiation Conditions and Changes Resulting from Irradiation

Specimen No.	Zr, w/o	Heat Treatment (a)	Burnup, a/o	Surface Temp, °C (b)	Center Temp, °C (b)	Length Change, %	Growth Rate, G_i (c)	Diameter Change, %	Weight Change, mg	Density Decrease, %	% Vol. Increase a/o Burnup	Relative Surface Appearance
AA-225	0	A	0.44	220	270	(d)	--	--	--	--	--	Rough
AA-226	0	A	0.69	330	400	(d)	--	--	--	--	--	Very rough
AA-227	0	A	0.88	420	510	(e)	--	--	--	--	--	
AA-228	0	A	0.94	440	540	(d)	--	--	--	--	--	Very rough
AA-6	0.52	A	0.29	210	260	6.3	21	12	2.4	1.8	6.2	Rough
AA-4	0.52	A	0.44	170	220	-2.8	-6.4	18	1.3	2.1	4.8	Rough
AA-5	0.52	A	2.7	540	690	(f)	--	--	--	--	--	
AA-36	0.52	B	0.12	110	130	12.5	98	-2.3	1.7	2.0	17	Smooth
AA-10	0.52	B	0.89	310	390	36	35	-10	4.3	4.6	5.4	Moderately smooth
AA-12	0.52	B	2.5	510	650	60(g)	19(g)	-3.1	-747.3	23.3	--	Moderately rough
AA-16	0.52	C	0.57	380	480	33	50	0 to -16	-2.1	6.7	13	Moderately rough
AA-15	0.52	C	1.4	460	580	(h)	--	--	--	--	--	Rough
AA-22	1.62	A	0.18	150	180	4.7	25	-0.64	1.7	1.7	9.4	Smooth
AA-20	1.62	A	0.68	240	300	3.2	4.6	0.16	-1.1	2.0	2.9	Smooth
AA-19	1.62	A	0.98	150	180	4.8	4.8	0.64	-1.8	3.4	3.7	Smooth
AA-17	1.62	A	1.6	210	260	6.3	3.8	2.23	2.2	8.3	5.6	Smooth
AA-21	1.62	A	2.1	540	690	23	9.9	10 to 70	49.8	--	71(i)	Smooth
AA-18	1.62	A	5.3	490	620	25	4.2	38 to 50	--	--	33(i)	Smooth
AA-26	1.62	D	0.68	470	590	55(g)	64(g)	-12 to +25	43.1	7.3	11	Smooth
AA-25	1.62	D	1.1	390	490	46(g)	34(g)	-12	54.2	19.5	22	Smooth
AA-32	1.62	E	0.41	280	370	45	91	-13	19.0	2.8	7.1	Moderately smooth
AA-30	1.62	E	1.4	500	630	(d)	--	4.7	--	--	--	Smooth
AA-29	1.62	E	2.1	280	350	57(g)	21(g)	-11	2.3	14.5	8.3	Smooth

(a) See Table I and Appendix A.

(b) Calculated for conditions at beginning of exposure

(c) $G_i = \ln(L/L_0) / \text{burnup} = \text{microin./in. per ppm burnup}$. Where rates are small, this expression is approximately equal to % length change per a/o burnup.

(d) Specimen was too distorted for measurement.

(e) Specimen could not be removed from irradiation capsule.

(f) Specimen disintegrated under irradiation.

(g) Value is too low because specimen had elongated into total available space.

(h) Specimen spontaneously ignited and burned before measurements could be taken.

(i) Volume calculated from dimensional analysis of photographs.

Unalloyed Uranium. One of the four specimens irradiated was so badly distorted that it could not be dislodged from its irradiation capsule. The remaining three specimens were also severely damaged, as shown in Figure 3. Although the conditions of the specimens were such that dimensional changes could not be determined with reasonable certainty, the samples appeared to have either retained their original lengths or shortened somewhat.

Neg. No.	16,166	16,167	16,168
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Specimen No.	AA-225	AA-226	AA-228
Burnup, a/o	0.44	0.69	0.94
Max. Irradiation			
Temp., °C	270	400	540

Figure 3. Effects of irradiation on as-cast unalloyed uranium.
Magnification 2X.

Uranium-0.52 w/o Zirconium Alloy. The two as-cast specimens of this alloy recovered from their irradiation capsules are shown in Figure 4. An additional specimen taken to a higher burnup was found to have disintegrated into small particles during irradiation. As can be noted from the photographs, the amount of surface roughening and distortion which developed in these specimens is only slightly less than that which occurred in the unalloyed specimens described above.

The three irradiated beta-quenched specimens which were examined are shown in Figure 5. Some improvement is evident in surface smoothness as compared to the cast specimens of this alloy. However, one end of specimen AA-12 exhibited high-temperature swelling, and all three specimens developed appreciable elongations under irradiation. As shown in Table III, the values of G_i , the growth coefficient,⁽²⁾ ranged from 19 to 98. These growth coefficients were significantly higher than those

Neg. No.	14,626	14,873
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Specimen No.	AA-6	AA-4
Burnup, a/o	0.29	0.44
Max. Irradiation Temp., °C	260	220

Figure 4. Effects of irradiation on as-cast uranium-0.52 w/o zirconium alloy. Magnification 2X.

Neg. No.	14,631	14,874	14,875
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Specimen No.	AA-36	AA-10	AA-12
Burnup, a/o	0.12	0.89	2.5
Max. Irradiation Temp., °C	130	390	650

Figure 5. Effects of irradiation on swaged uranium-0.52 w/o zirconium alloy, quenched from 725°C. Magnification 2X.

obtained for beta-quenched unalloyed uranium.⁽²⁾ The beta-quenched alloy specimens, on the other hand, showed a somewhat improved surface smoothness as compared to similar unalloyed material.

Two capsules containing gamma-quenched specimens were opened. The specimen with the higher burnup, No. AA-15, ignited and burned to oxide soon after exposure to air. The remaining specimen is shown in Figure 6. The behavior of this specimen under irradiation was quite similar in all respects to the beta-quenched specimens described above.

14, 627

2X

Figure 6. Effect of irradiation on specimen AA-16, which received 0.57 a/o burnup at a maximum temperature of 480°C. The specimen material is uranium-0.52 w/o zirconium alloy quenched from 800°C.

Uranium-1.62 w/o Zirconium Alloy. All specimens of this composition showed considerably better surface smoothness after irradiation than did those of the 0.52 w/o zirconium alloy.

The behavior of the cast specimens was particularly good. All six of the irradiated cast specimens are shown in Figure 7. The original surface smoothness of the specimens was retained up to the highest burnup achieved, 5.3 a/o. With the exception of specimen AA-22, which received the lowest

burnup, the specimens appeared to be free of anisotropic irradiation growth. As can be noted in Figure 7 and Table III, the two specimens irradiated above 600°C developed large increases in volume. This is shown more clearly in Figure 8, in which has been plotted the rate of volume increase as a function of center irradiation temperature.

When specimen AA-21, which was irradiated at the highest temperature, was subjected to a hardness test the specimen fractured. Examination of the fragments revealed that a large void had developed within the specimen. The void extended along the axis of the enlarged portion of the specimen and the diameter of the void was about one-third that of the total specimen diameter. One of the fragments was examined metallographically, and the bubble-like structure found is shown in Figure 9.

Volume measurements on specimen AA-18, which was irradiated to 5.3 a/o burnup at a somewhat lower temperature, also indicated that it had swelled under irradiation. When this specimen was transversely sectioned, a similar large void was found in its interior, although not as large relative to the size of the specimen as was observed in specimen AA-21. A metallographic section of a part of specimen AA-18 is shown

Neg. No. 14,628 14,876 15,393 15,392 14,877 15,868

Specimen No.	AA-22	AA-20	AA-19	AA-17	AA-21	AA-18
Burnup, a/o	0.18	0.68	0.98	1.6	2.1	5.3
Max. Irradiation Temp., °C	180	300	180	260	690	620

Figure 7. Effect of irradiation on as-cast uranium-1.62 w/o zirconium alloy. Magnification 2X.

Figure 8. Effect of irradiation temperature on the rate of volume increase in cast uranium-1.62 w/o zirconium alloy.

23,444

100X

Figure 9. Metallographic section of specimen AA-21. The highly porous material shown above enclosed a large central void.

in Figure 10. It can be noted that, as in the case of specimen AA-21, a very porous region extended from the central void to the exterior of the specimen. Several radial cracks were also noted, one of which is shown in Figure 10.

25336

75X

Figure 10. Metallographic section of specimen AA-18, showing the porous structure which extended from the void on the left to the outer surface of the specimen on the right.

Although the two groups of swaged and heat-treated uranium-1.62 w/o zirconium alloy showed good surface smoothness after irradiation, all developed extensive elongations under irradiation. Specimens which were air-cooled from the gamma phase are shown in Figure 11, and it can be noted that one specimen, No. AA-26, had not only lengthened considerably but had departed considerably from a circular cross section. The other specimen, AA-25, shows incipient swelling at one end. Figure 12 shows specimens of the isothermally transformed alloy and illustrates the significant dimensional instability which was observed. One specimen, No. AA-30, shows severe high-temperature damage.

Neg. No.	14,629	14,878
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Specimen No.	AA-26	AA-25
Burnup, a/o	0.68	1.1
Max. Irradiation Temp., °C	590	490

Figure 11. Effect of irradiation on swaged uranium-1.62 w/o zirconium alloy air-cooled from 800°C. Magnification 2X.

Neg. No.	14,630	14,879	15,396
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Specimen No.	AA-32	AA-30	AA-29
Burnup, a/o	0.41	1.4	2.1
Max. Irradiation Temp., °C	370	630	350

Figure 12. Effect of irradiation on swaged uranium-1.62 w/o zirconium alloy, heated to 800°C and isothermally transformed at 500°C. Magnification 2X.

DISCUSSION OF RESULTS

It was evident from the irradiated wrought specimen that a suitable stabilizing heat treatment had not been selected for either alloy. The selection of the heat treatments in this investigation was initially based on thermal cycling data. At that time this seemed to be a reasonable procedure, because earlier studies on unalloyed uranium had shown that its behavior under thermal cycling was similar in many respects to its behavior under irradiation. The present data indicate that for alloys used in this investigation a one-to-one similarity does not exist or, if it does, it is based on a presently unknown type of thermal cycle. Table IV summarizes irradiation and cycling growth rates for the present alloys as well as for unalloyed heat-treated uranium. A similarity in behavior is shown to exist for the unalloyed material, but for the alloys the only similarity noted was in the gamma-quenched alloy containing 0.52 w/o zirconium. This material was unstable under cycling as well as under irradiation. Although thermal cycling tests have been commonly used as an aid in developing stable uranium alloys, the present data, as well as data being obtained from irradiations of other alloys, show that it is unsafe to rely on thermal-cycling data as an indication of anisotropic dimensional stability under irradiation.

One can, however, use cycling tests as an indication of surface changes to be expected under irradiation. Since such changes are a function of the grain size in the material, metallographic examination of proposed fuel materials can also serve as a means of evaluating surface stability to be expected under irradiation.

The severe surface damage which developed under irradiation in the unalloyed material and to a somewhat lesser extent in the 0.52 w/o zirconium alloy is attributed to undesirably large grain sizes in the specimens. Such large grain sizes were not expected, because the specimens had been cast as thin sections in a cooled copper mold. The addition of 0.52 w/o zirconium to uranium evidently had little effect on the grain size of the material. The much greater surface smoothness of the irradiated 1.62 w/o zirconium alloy specimens indicated that a marked reduction in alpha grain size had occurred with this amount of added zirconium.

Extrapolation of the curves shown in Figure 8 indicates that the excellent performance of the uranium-1.62 w/o zirconium alloy in the cast condition may be limited to temperatures below approximately 500°C. Above the critical swelling temperature, swelling occurs at rates that would limit its usefulness as fuel material. Similar swelling of specimens irradiated at elevated temperatures has been observed in work on unalloyed uranium and on uranium-silicon alloys.⁽⁹⁾ In the study on unalloyed uranium, which was on powder compacts, the critical swelling temperature was found to be near 430°C.

TABLE IV.

Comparison of Growth Rates Obtained by Irradiation and by Thermal
Cycling on Wrought, Heat-treated Uranium and Uranium-Zirconium Alloys

Material	Heat Treatment	Irradiation Growth Rate, microin./in./ppm burnup of all atoms	Cycling Growth Rate, microin./in./cycle
Unalloyed U	alpha rolled, beta quenched	25 ^{(2)*}	15 ⁽¹⁾
Unalloyed U	600°C rolled	77 ⁽²⁾	180 ⁽¹⁾
Unalloyed U	300°C rolled	690 ⁽²⁾	500 ⁽¹⁾
U-0.67 w/o Zr	725°C for 1 hr, water quenched	35-98	3 ⁽⁶⁾
U-0.52 w/o Zr	725°C for 1 hr, water quenched		
U-0.67 w/o Zr	800°C for 1 hr, water quenched	50	40 ⁽⁶⁾
U-0.52 w/o Zr	800°C for 1 hr, water quenched		
U-2.0 w/o Zr	800°C for 1 hr, air cooled	64	-2 ⁽⁶⁾
U-1.62 w/o Zr	800°C for 1 hr, air cooled		
U-2.0 w/o Zr	800°C for 1 hr, 500°C for 1 hr, air cooled	91	0 ⁽⁶⁾
U-1.62 w/o Zr	800°C for 1 hr, 500°C for 1 hr, air cooled		

*Numbers in parentheses indicate sources of information listed in references.

The as-cast specimens which swelled in the present investigation, as well as certain other specimens described previously,⁽⁹⁾ are characterized by relatively large central voids surrounded by porous material. It is believed that the small pores were formed as a result of nucleation and growth of bubbles of volatile fission products, and that the large voids may have resulted from an effective inward movement and coalescence of bubbles. Because of the high radial temperature gradient (in the case of the present specimens, approximately 2000°C/in.) in the specimens, it would appear likely that an expanding gas bubble would preferentially strain the matrix metal on the inward side of the bubble, since this metal is at the highest temperature and would therefore have the lowest creep strength. A net inward motion of the bubble would thus accompany its expansion.

Using measurements of central void volume and specimen wall thicknesses made from enlarged specimen photographs, the tangential stress in specimen AA-21 in its final shape while at irradiation temperature was calculated to range as high as 6000 psi, depending upon what reasonable fraction of fission gas release into the void was assumed. The local effects of the small pores on stress was not taken into account. From available information on the mechanical properties of uranium-rich zirconium alloys,⁽¹¹⁾ it would appear that had the material surrounding the large central void been subjected to a tensile stress of the order of 6000 psi, considerable deformation might be expected.

Thermal-cycling stresses provide another possible mechanism which may have been responsible for the formation of the large central voids in specimen AA-18 and AA-21. Workers at Atomics International have shown that large central voids can be developed in uranium slugs by repeated thermal cycling into the beta-phase temperature range.⁽¹²⁾ Voids that appear to be approximately one-third the diameter of the slug have occurred after as few as 50 thermal cycles on 1-1/4-in. and 3/4-in.-diameter specimens. Similar voids have been developed in work at Argonne in which somewhat smaller uranium specimens were cycled in and out of a reactor such that alpha-beta cycling conditions existed.⁽¹³⁾ Although specimens AA-18 and AA-21 were subjected, respectively, to 54 and 14 shutdown cycles in the MTR, there is considerable doubt as to whether such small specimens would show the same reactions as the large uranium specimens described above. However, the possibility cannot be ruled out that thermal cycling resulting from reactor shutdowns may at least have contributed to the formation of the large central voids in the two specimens.

It has been suggested⁽¹⁴⁾ that if the voids resulted from the accumulation of fission gas at the center of the specimen, then presumably a small axial hole placed in the specimen before irradiation would act as a vent to permit the escape of gas and thus raise the permissible operating temperature of the fuel. Specimens designed to test this hypothesis have been irradiated and are awaiting examination.

CONCLUSIONS

1. The thermal-cycling results on the wrought uranium-zirconium alloys did not accurately predict their dimensional behaviors under irradiation.
2. Because of excessive surface roughening under irradiation, the 0.52 w/o zirconium alloy appears to be of little use as a reactor fuel material.
3. An alloy content of 1.62 w/o zirconium is sufficient to minimize surface roughening under irradiation to at least 5.3 a/o burnup.

4. Cast uranium-1.62 w/o zirconium alloy is highly stable dimensionally under burnups of at least 1.6 a/o at temperatures below that at which swelling occurs.

5. The critical swelling temperature for cast uranium-1.62 w/o zirconium alloy lies between 300 and 620°C, with some indication that it may be near 500°C.

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APPENDIX A

CASTING AND FABRICATION OF 10% U^{235} -ENRICHED
URANIUM-ZIRCONIUM ALLOY PIN SPECIMENS FOR IRRADIATION TEST

by

A. B. Shuck

Two lots of uranium-zirconium alloy, designated 16F-69 and 16F-70 were made up for these specimens. S. M. Batch No. 2-60-1A1, 13.36% U^{235} -enriched uranium was diluted with Mallinckrodt reduction metal to bring the U^{235} concentration in the alloys down to approximately 10 w/o. Data on the production of the castings is given in Table V.

TABLE V

Furnace Charges and Computations

a) Casting No. 16F-69 (a&b)

S.M. Batch No. 2-653-1

Batch No.	Grams Net	U^{235} , w/o	U^{235} , gm	U^{238} , w/o	U^{238} , gm	U^{234} , w/o	U^{234} , gm	Zr, w/o	Zr, gm
2-60-1A1	68.212	13.36	9.113	86.33	58.887	0.31	0.211		
Normal U	22.257	0.721	0.160	99.273	22.095	0.006	0.001		
No. 3 XL Bar Zr	0.958							100.0	0.958
Total Grams	91.427		9.273		80.983		0.212		0.958
Calc. Anal., %		10.143		88.577		0.232		1.048	

b) Casting No. 16F-70 (a, b & c)

S.M. Batch No. 2-653-2A

Batch No.	Grams Net	U^{235} , w/o	U^{235} , gm	U^{238} , w/o	U^{238} , gm	U^{234} , w/o	U^{234} , gm	Zr, w/o	Zr, gm
2-60-1A1	68.417	13.36	9.140	86.33	59.064	0.31	0.212		
Normal U	23.071	0.721	0.166	99.273	22.903	0.006	0.001		
No. 3 XL Bar Zr	2.009							100.0	2.009
Total Grams	93.497		9.306		81.157		0.213		2.009
Calc. Anal., %		9.953		86.802		0.227		2.148	

Melting and Casting

Both charges were melted to 1500°C in thoria crucibles at a vacuum of approximately 2.5×10^{-5} mm of Hg and poured into a copper mold to form six 0.137-in.-dia. by 1-in. pin castings attached to a heavy sprue (see Figure 13). After casting, the pins were sawed from the sprue using an 0.012-in. milling cutter. The sprues were remelted and cast into a water-cooled copper mold, lined down to 1/4-in. dia.

AS-115

3/4X

Figure 13. Casting number 16F-69 and 16F-70
showing method of casting pin specimens

Mill Fabrication

The 1/4-in.-dia. rods were jacketed in 7/16-in.-dia. Armco ingot iron jackets and swaged at 600°C to approximately 0.180 in. The jackets were then stripped mechanically and the bare uranium swaged to 0.139 in. in four passes with a 300°C oil bath stress relief between each pass. The rod was then degreased, electrolytically cleaned and swaged to 0.126-0.128 in. (75% RA).

Heat Treatment

The rods were then parted in half, encapsulated in Vycor and were given the heat treatments shown in Table VI. The capsules of those specimens which were quenched were crushed while submerged in water.

Machining

The as-cast pins were turned and machined to length. The fabricated pins were notched, frozen in liquid nitrogen and broken to 25/32 in. and faced to length. Turnings from each casting were submitted for isotopic and chemical analysis (see Table VII). It will be seen that the zirconium content was found by analysis to be only one-half to three-quarters as great as had been anticipated.

TABLE VI

Heat Treatment of Specimen Materials

Designation	Heat Treatment
A	As cast.
B	Heated to 725°C in lead for 1 hr, water quenched, straightened and annealed in vacuum at 575°C (1/2 of 16F-69B).
C	Heated to 800°C in lead for 1 hr, water quenched, straightened and annealed in vacuum at 575°C (1/2 of 16F-69B).
D	Heated to 800°C in lead for 1 hr, air cooled (1/2 of 16F-70B).
E	Heated to 800°C in lead for 1 hr, transferred to 500°C lead for 1 hr, air cooled (1/2 of 16F-70B).

TABLE VII

Analyzed Compositions^(a) of Irradiation Specimens

Nominal Zr, w/o	Actual Zr, w/o	U ²³⁴ , w/o	U ²³⁵ , w/o	U ²³⁸ , w/o	C, ppm	N, ppm
0	0	0.11	9.9	90.0	117	51
1	0.52	0.23	10.4	89.4	605	53
2	1.62	0.23	10.3	89.5	440	20

(a) Uranium isotopic analysis is based on a total of 100 w/o uranium.

Electropolishing

Final electropolishing was done on these specimens just before loading into NaK-filled capsules.

Identification and Dimensioning

Tables VIII and IX show the identification, dimensions, weight and calculated composition of the U-Zr alloy pin specimens.

TABLE VIII

Identification, Dimensions, Weight Composition and Heat Treatment
of U-0.52 w/o Zr Specimens

Casting No.	Batch No.	Dimensions (in.)	Gross Weight (grams)	Uranium Weight (grams)	U ²³⁵ Weight ^a (grams)	Zirconium Weight (grams)	Heat Treatment ^b
16F 69-A	2.653-1A	0.125 x 0.751	2.792	2.777	0.289	0.015	A
B	B	0.1245-0.1250 x 0.7515	2.787	2.773	0.288	0.014	A
C	C	0.1243-0.1252 x 0.7502	2.808	2.793	0.290	0.015	A
D	D	0.1254-0.1258 x 0.7512	2.830	2.815	0.293	0.015	A
E	E	0.1255-0.1256 x 0.7514	2.832	2.817	0.293	0.015	A
F	F	0.1255-0.1252 x 0.7484	2.822	2.808	0.292	0.014	A
G	G	0.1266-0.1270 x 0.7514	2.909	2.894	0.301	0.015	B
H	H	0.1267-0.1272 x 0.7512	2.909	2.894	0.301	0.015	B
I	I	0.1266-0.1268 x 0.7520	2.912	2.897	0.301	0.015	B
J	J	0.1264-0.1267 x 0.7511	2.911	2.896	0.301	0.015	B
K	K	0.1268 x 0.7510	2.915	2.900	0.302	0.015	B
L	L	0.1262-0.1264 x 0.7511	2.901	2.886	0.300	0.015	B
M	M	0.1262-0.1266 x 0.7511	2.889	2.874	0.299	0.015	C
N	N	0.1260-0.1262 x 0.7516	2.861	2.846	0.290	0.015	C
O	O	0.1262-0.1266 x 0.7510	2.857	2.843	0.296	0.014	C
P	P	0.1261-0.1262 x 0.7505	2.868	2.853	0.297	0.015	C
Q	Q	Cracked (did not use)	2.850	2.835	0.295	0.015	C
R	R	Cracked (did not use)	2.846	2.831	0.294	0.015	C
Totals			51.499	51.232	5.328	0.177	

^aBased on 10.4% U²³⁵.

^bSee Table VI.

TABLE IX

Identification, Dimensions, Weight Composition and Heat Treatment
of U-1.62 w/o Zr Specimens

Casting No.	Batch No.	Dimensions (in.)	Gross Weight (grams)	Uranium Weight (grams)	U ²³⁵ Weight ^a (grams)	Zirconium Weight (grams)	Heat Treatment ^b
16F 70- 1	2-653-2A 1	0.1250-0.1252 x 0.7510	2.762	2.718	0.280	0.045	A
2	2	0.1251-0.1258 x 0.7518	2.767	2.722	0.280	0.045	A
3	3	0.1248-0.1252 x 0.7477	2.736	2.692	0.277	0.044	A
4	4	0.1247-0.1254 x 0.7510	2.754	2.710	0.279	0.045	A
5	5	0.1248-0.1252 x 0.7514	2.728	2.684	0.276	0.044	A
6	6	0.1248-0.1250 x 0.7502	2.649	2.606	0.268	0.043	A
7	7	0.1272-0.1264 x 0.7502	2.829	2.783	0.287	0.046	D
8	8	0.1270 x 0.7511	2.845	2.799	0.288	0.046	D
9	9	0.1268-0.1271 x 0.7514	2.843	2.797	0.288	0.046	D
10	10	0.1268-0.1270 x 0.7512	2.835	2.789	0.287	0.046	D
11	11	0.1282-0.1274 x 0.7504	2.887	2.840	0.293	0.047	D
12	12	Cracked (did not use)	2.853	2.807	0.289	0.046	D
13	13	0.1268-0.1272 x 0.7512	2.876	2.830	0.291	0.047	E
14	14	0.1268-0.1266 x 0.7512	2.864	2.817	0.290	0.046	E
15	15	0.1262-0.1268 x 0.7503	2.848	2.802	0.289	0.046	E
16	16	0.1278-0.1272 x 0.7500	2.843	2.797	0.288	0.046	E
17	17	0.1265-0.1270 x 0.7515	2.861	2.815	0.290	0.046	E
18	18	Cracked (did not use)	2.884	2.837	0.292	0.047	E
Totals			50.664	49.845	5.132	0.821	

^aBased on 10.3% U²³⁵.

^bSee Table VI.

APPENDIX B

BURNUP COMPUTATIONS

Because the specimens were partially enriched it was expected that they would depress the flux in their vicinity so that the flux monitors, which were approximately an inch away from the specimen, would indicate fluxes higher than actually existed at the surfaces of the specimens. For this reason, the outer surfaces of four specimens were chemically sampled, as described earlier. The solutions thus obtained were analyzed and two different methods were used to correlate their cobalt-indicated fluxes, given in Table X, column A, with the actual degree of burnup which had occurred in them.

TABLE X
Percent Uranium Burnup Numbers Obtained by Various Methods of Analysis

Specimen No.	Distance Above or Below Reactor Midplane (in.)	Cobalt Foil Flux Indication (10^{20} nvt)	Burnup from Cesium-137 Fission Product Indication	Surface Burnup Computed from Isotopic Analysis Data					
				$\alpha = 0.182$		$\alpha = 0.150$		$\alpha = \text{variable parameter}$	
				Equation (5)	Equation (7)	Equation (5)	Equation (7)	Equation (8) ^a	Equations (5) + (7)
		A	B	C	D	E	F	G	H
AA-20	-13.5	2.23	0.66	0.84	0.64	0.87	0.77	0.136	0.88
AA-19	+18.0	3.56	0.21	1.23	1.03	1.27	1.25	0.152	1.26
AA-17	-13.0	5.65	1.54	2.00	1.73	2.06	2.10	0.155	2.05
AA-21	+ 1.5	9.56	0.96	2.69	2.37	2.77	2.86	0.157	2.76

^aNumbers in column G represent the various values of α computed from equation (8). All other numbers in columns B to H are percentages burnup numbers.

One of the methods was a radiochemical analysis of fission-product Cs^{137} , for which a calibrated procedure had been developed.⁽⁸⁾ The burnup numbers obtained by this method are given in Table X (column B). They are based on 113 counts of Cs^{137} per minute for each 10^{12} fissions.

The second calibration method was worked out from an isotopic analysis of the uranium contained in the sample solutions (see Table XI). The burnup numbers listed in Table X, columns C to H, were obtained from these data by computations which will now be discussed.

Computation of Burnup from Mass Spectrograph Data

Let the pre- and postirradiation percentages of U^{234} , U^{235} , and U^{236} be represented as ratios by the letters a_0 , a ; b_0 , b ; c_0 , c , respectively, and ignore the small differences between weight and atom percent. Let

TABLE XI

Weight Percent Abundance of Uranium Isotopes before and after Irradiation

Specimen No.	U ²³⁴		U ²³⁵		U ²³⁶		U ²³⁸	
	Pre a ₁	Post a ₂	Pre b ₁	Post b ₂	Pre	Post C	Pre d ₁	Post d ₂
AA-20	0.23	0.215 ± 0.003	10.3 ± 1	9.40 ± 0.08	-	0.117 ± 0.005	89.5	90.27 ± 0.08
AA-19	0.23	0.219 ± 0.003	10.3 ± 1	8.97 ± 0.09	-	0.190 ± 0.003	89.5	90.62 ± 0.09
AA-17	0.23	0.210 ± 0.002	10.3 ± 1	8.12 ± 0.09	-	0.321 ± 0.002	89.5	91.35 ± 0.09
AA-21	0.23	0.210 ± 0.003	10.3 ± 1	7.34 ± 0.07	-	0.441 ± 0.005	89.5	92.01 ± 0.07

N_0 be the original whole amount of uranium in a specimen, and N the corresponding final amount. Let B stand for the total uranium burnup ratio; assume that the burnup is due to U^{235} fissions only, and let F be the amount of U^{235} fissioned.

The rate of fissioning depends only on N_5 , the amount of U^{235} present at any given time, and $\sigma_f U^{235}$, the cross section for fission. Therefore,

$$\frac{dF}{dt} = \sigma_f U^{235} \phi N_5 \quad (1)$$

The solution to this equation is

$$F = (\sigma_f / \sigma_t) [N_{O,4} + N_{O,5} - N_4 - N_5] \quad (2)$$

Here $N_{O,4}$ and $N_{O,5}$ are the original amounts of U^{234} and U^{235} present; N_4 and N_5 are the final amounts. It will be observed that $\sigma_f / \sigma_t = 1 / (1 + \alpha)$. Substitution of the isotopic analyses into the equation gives

$$F = [(a_0 + b_0) N_0 - (a + b) N] / (1 + \alpha) \quad (3)$$

Uranium may be assumed to be lost during irradiation only through fission in U^{235} and parasitic capture in U^{236} and U^{238} . The ratio of total fissions to total uranium lost depends on the relative amounts of isotopes present, as well as on the cross sections for the reactions. In the specimens under consideration, fission accounts for at least 95% of the uranium lost, and we may substitute in (3) the approximation

$$N = N_0 - F \quad (4)$$

without introducing more than a 1% error in the final results. Equation (3) then becomes

$$B = F / N_0 = (a_0 + b_0)(a + b) / [(1 + \alpha) - (a + b)] \quad (5)$$

It will be seen from the equation that evaluation of the uranium burnup levels depends only on the isotopic analysis for U^{234} and U^{235} . An independent check might be made, based on α and the isotopic analysis of U^{236} . Then, if $N_{O,6}$ and N_6 represent the original and final amounts of U^{236} present, the following relationship exists:

$$F = [N_6 - N_{O,6}] / \alpha \quad . \quad (6)$$

When the approximation (4) is used and $N_{O,6}$ is set equal to zero, this burnup equation becomes

$$B = F / N_O = c / (\alpha + c) \quad . \quad (7)$$

Evaluation of both (5) and (7) depends upon the value of α used. For example, if the neutrons are considered to be in equilibrium at the MTR water temperature, the value $\alpha = 0.182$ may be used. Results from this choice are shown in Table X, columns C and D, and it will be seen that agreement between the two computations is not very good.

The results from both equations may be brought into agreement by using values of α obtained from their common solution:

$$\alpha = c(1 - a_O - b_O) / [(a_O + b_O) - (a + b + c)] \quad . \quad (8)$$

Solutions for equation (8) appear in Table X (column G). These values range from $\alpha = 0.136$ to $\alpha = 0.157$, and have an average value of 0.150. When applied to equations (5) and (7), the average value produces burnup numbers which are in good agreement (see columns E and F).

It is conceivable that the ratio of capture to fission cross sections will vary somewhat along the length of a vertical hole in the MTR reflector, if the hole is close enough to the fuel to intercept an epithermal component of the neutron spectrum. In such a case one would expect the value of α to be symmetrical about the midplane of the reactor. It is apparent from Table X that the computed values of α do not correlate with position of the specimens in the reactor, but they do correlate with the degree of surface uranium burnup. It is also probable that they correlate directly with the degree of approximation introduced by ignoring the capture of U^{236} and U^{238} , an error which increases in absolute size in direct proportion to the burnup. Therefore α is considered to be a variable parameter, probably numerically different from the effective capture-to-fission cross-section ratio, but useful in obtaining reasonably accurate burnup numbers in the present analysis. When used in this way the burnup numbers shown in the final column of Table X were obtained.

Correlation of Monitor Flux Data with Burnup Analysis Data

The surface uranium burnup values obtained from cobalt monitors, radiocesium analysis, and mass spectroanalysis are summarized in Table XII. The best correlation appears between the monitors and mass spectro results. Analysis reveals that an average drop of $14 \pm 8\%$ in effective flux may be expected between the monitors and the surfaces of the uranium specimens. Because the data are few the standard deviation is relatively large.

TABLE XII

Comparison of Surface Uranium Percent Burnups by Various Methods of Analysis

Specimen No.	Cobalt-60 Foil Indication ^a	Cesium-137 Fission Product Indication	Computed from Mass Spectrograph Data ^a	
			$\alpha = 0.182$	α Variable ^b
AA-20	0.91	0.70	0.84	0.88
AA-19	1.40	0.20	1.23	1.26
AA-17	2.30	1.90	2.00	2.05
AA-21	3.30	1.11	2.69	2.76

^aThe least squares relationship between $\text{Co}^{60}(\text{C})$ and Mass Spectro(M) results is $M = 0.86C + 0.08$.

^bThe values shown in this column were used as a basis to obtain the average burnups shown in Table III, using the Co^{60} -indicated flux available for each specimen.

Correlation of the radiocesium data is poor, particularly for specimens AA-19 and AA-21. The possibility that surface cesium content of the test specimens was somewhat depleted by contact with, and solution in, NaK during irradiation was not explored. Although it was observed that the burnups obtained from the fission product analyses are all definitely lower than those obtained from the isotopic analysis data, as would be expected if surface solution of cesium had occurred, it should be noted that the largest discrepancy occurred with specimen AA-19 which was irradiated at the lowest temperature.

Computation of Average Total Atom Burnup

It is customary to express data of radiation damage as a function of the percent fission of all the atoms in the material. Surface uranium burnup data, therefore, are subject to two additional corrections. One of these is needed because of attenuation of neutron flux in the body of the specimen. The other is required to take account of alloy atoms present. The corrections

are listed below and were applied to the surface burnups shown in the final column of Table X to obtain the average burnups shown in Table III.

Correction Factor	Alloy Composition	
	0.52% Zr	1.62% Zr
1) Self-shielding Factor (average to surface flux ratio)	0.810	0.810
2) Alloy Content Factor	0.987	0.959
Combined Correction Factor	0.799	0.777