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PRELIMINARY STUDIES OF IRRADIATION
DAMAGE TO URANIUM-ZIRCONIUM ALLOYS.

By
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PRELIMINARY STUDIES OF IRRADIATION DAMAGE TO
URANIUM-ZIRCONIUM ALLOYS

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Metallurgy Division Program 6.1.1

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PRELIMINARY STUDIES OF IRRADIATION DAMAGE TO URANIUM ZIRCONIUM ALLOYS

by

S. H. Paine, F. L. Brown and W. F. Murphy

ABSTRACT

The addition of zirconium to uranium is shown to have a beneficial effect upon the irradiation stability of the fuel alloy. Preliminary methods used to discover this fact, and experimental problems encountered are described in this report.

I. INTRODUCTION

The general search for uranium-rich alloys with improved resistance to radiation damage was barely under headway in 1950 when the need for such a fuel material became very urgent at ANL in the engineering of the core of the liquid metal-cooled fast reactor, EBR-I. The use of zirconium as a stabilizer appeared to be a likely choice because of its high melting point, its high solid solubility in gamma uranium, and its low absorption cross section when purified from associated hafnium. Also, initial attempts to hot roll a 10 w/o Zr alloy at Battelle Memorial Institute (BMI) had appeared promising. It was desired to get an evaluation of the irradiation behavior of such an alloy as rapidly as possible.

S. Untermeyer suggested an accelerated qualitative test for the central thimble of the Argonne Heavy Water Research Reactor, CP-3', in which a 10% Zr alloy of U^{235} would be compared visually with an identical shape of unalloyed enriched uranium. Specimens would be kept thin to minimize self-shielding and would be sandwiched between aluminum blocks for fission heat removal.

Procurement of the enriched alloy from BMI was expedited, and irradiation was started late in 1950. The specimens were examined after three months, and the encouraging nature of the results was made the subject of an interim report.⁽¹⁾ The irradiation in CP-3' was continued with the intention of watching the test until other irradiations in higher flux facilities should overtake it.

Technical details and final results of the test are recorded in this report. In addition, a brief account is given of a single clad specimen irradiation in the Low Intensity Training Reactor at Oak Ridge. This test

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was also conceived and expedited by Untermeyer, and was intended to use a somewhat more massive specimen than could be accommodated in CP-3'. Both of these experiments were preliminary in nature. Their techniques, together with those of the "pin cushion" experiment reported elsewhere,^(2,3) have been superseded by Kittel's NaK capsule technique.⁽⁴⁾

II. DESCRIPTION OF TESTS

A. Argonne Heavy Water Reactor (CP-3') Test

The specimens irradiated in the ANL-CP-3' reactor were three discs or "wafers," approximately 1/4 in. in diameter and 1/32 in. thick. Three metallurgical histories were chosen for comparison:

- a. Normal uranium, alpha rolled (300° C), heated to 725° C and water quenched;
- b. Uranium-235, alpha rolled (300° C), heated to 725° C and water quenched;
- c. Uranium-235 having an alloy content of 10 w/o Zr (22 1/4 a/o), arc melted and rolled at 980° C (1800° F) in a Mo jacket into strip approximately 0.040 in. thick.

All specimens were alpha annealed at 575° C after rough machining. A specimen of the U-Zr alloy was retained for thermal cycling and for additional annealing tests.

Before encapsulation for irradiation the specimens were carefully measured with a micrometer, weights and densities were taken, micro-profiles of the flat surfaces were made, and the appearances of the wafers were recorded photographically.

Figure 1 is a sectional drawing of the aluminum capsule in which the specimens were placed for irradiation. The end knob is a grappling adapter. Depths of the recessed wells in the split, conical aluminum wedge were carefully gauged to accommodate the exact thickness of each wafer plus two zirconium foils which were included to prevent direct contact between uranium and aluminum. After careful fitting and assembling of a specimen with its foil and capsule components, the conical wedge was driven home and locked in place by the threaded end plug. The capsule was then held in a close-fitting copper cooling block while a circumferential weld was made between cap and cartridge.

Fig. 1. Specimen Container for Uranium Wafers (MY 1175-B)

Specimens were placed in a central recessed well (shown in section) on the axis of the inner split tapered pin.

Three of the sealed capsules were prepared for the initial irradiation period, one for each specimen. When the specimens were recanned for subsequent irradiation increments, however, a single capsule having three wells was used and the weld bead was omitted. There were four periods of exposure in the reactor, details of which are recorded in Appendix A.

The initial irradiation lasted three months. The capsules were then cut into three pieces each, and the wedging cones were driven out of the center sections. Figures 2-a) and -c) record the appearance of the specimens as decanned. Barrier foils came away from the normal uranium specimen readily, but were tightly adherent to both enriched wafers and had to be polished away. However, before this was done, preirradiation profile (Fig. 3) and linear dimension measurements were repeated. The process for removal of barrier foils from the U^{235} specimens is detailed in Appendix B. Because of the techniques used it was possible to do a metallographic study of the wafers after the foil was removed (Figs. 4 to 6); measurements of weights, densities and rechecks of linear dimensions were then made. These are recorded in Appendix C.

The specimens were returned to the reactor for a second period of four months. After this irradiation the zirconium barrier foils were again tightly adherent to the enriched specimens, and it was decided to record the appearance in a photograph and return the wafers to the reactor without disturbing the foils. Figures 2-b) and -d) show the specimens before and after the second irradiation period.

After the third irradiation the specimens received only visual examination. After the final exposure, careful stereophotos were made (Figs. 7-9), but no further measurements were taken. The normal uranium and U^{235} wafers were dissolved in 12N nitric acid and the average total atom burnup was determined by radiochemical analysis (see Appendix A for details).

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Fig. 2. Macrophotographs of Wafers after First and Second Irradiations (Magnified 6X)

(a)

Macro No.
11,053

(b)

11,835
11,834
11,836

(c)

11,049

(d)

11,262

Normal Uranium

U^{235}

U^{235} -10 w/o Zr

- (a) and (b). Edge views of wafers after first and second irradiations (0.7% and 1.8% surface depletion of U^{235} , respectively. See Table IV.)
- (c) and (d). Side views of wafers after first irradiation before and after polishing for metallographic examination. Note tightly adherent zirconium barrier foils on the two U^{235} wafers (c).

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FIGURE 3. SURFACE PROFILES OF WAFERS.

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Fig. 4. Beta-Treated and Irradiated Normal Uranium
(0.04% total atom burnup.)

10,880

Beta treated

100X

(Polarized light.)

11,119

Irradiated

100X

Fig. 5. Beta-Treated and Irradiated U^{235}
(0.17% total atom burnup.)

11,266

Beta treated

100X

(Polarized light.)

11,117

Irradiated

100X

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Fig. 6. Annealed and Irradiated U^{235} -10 w/o Zr Alloy
(0.16% total atom burnup.)

11,173

100X

11,264

100X

11,176

Annealed

500X

(Bright field.)

11,265

Irradiated

500X

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Fig. 7. Stereo Views of Normal Uranium Wafer after Final Irradiation
(0.4% total atom burnup.) (Magnified 9X)

Macro No.
14,481

14,480

14,479

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 Fig. 8. Stereo Views of U²³⁵ Wafer after Final Irradiation
 (1.74% total atom burnup.) (Magnified 9X).

Macro No.
 14,475

14,474

14,473

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Fig. 9. Stereo Views of U²³⁵-10 w/o Zr Wafer after Final Irradiation
(1.64% total atom burnup.) (Magnified 9X).

Macro No.
14,478

14,477

14,476

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B. Oak Ridge Low Intensity Test Reactor (LITR) Test

The pin for irradiation in the LITR was an alloy of 4.5 w/o Zr (11.5 a/o) with uranium enriched in U^{235} to 13%. After casting, the material was hot worked, oil quenched from 725° C, and annealed 48 hours at 575° C. The specimen was machined to 1/8 in. by 2 in. final size.

The linear dimensions, weight and density, and hardness of the specimen were carefully measured, and its appearance was recorded photographically. It was then assembled into a tightly fitting, finned aluminum capsule which was sealed by end welding an aluminum plug in place. The aluminum thickness in wall and fins was 1/32 in. The assembly was leak tested by autoclaving 18 $\frac{3}{4}$ hours at a water pressure of 90 psi. Figure 10 shows the preirradiation appearance of pin and capsule.

A special "A" type stringer was constructed to receive the sample in the LITR reflector. It was an aluminum can loaded with beryllium, having a 5/8 in. (I.D.) aluminum tube along its axis to act as a cooling water channel. A restriction at its midpoint was designed to hold the specimen capsule in the region of greatest flux. Irradiation commenced in October, 1951, and was terminated December 23, 1952. When visually inspected early in June the cartridge appeared to be as shiny as ever, but when removed in December considerable surface corrosion had occurred. It was estimated that it had received an integrated thermal flux of 2.5×10^{20} nvt and that perhaps 12% of the fissile surface atoms in the specimen would be depleted.

After shipment to ANL, the capsule was photographed and the specimen was decanned by dissolving away the aluminum jacket in aqueous 50% NaOH solution. Linear, density and hardness measurements were repeated (see Appendix C). After photography (Fig. 10) the specimen was accidentally dropped from a height of two feet and was broken into two pieces. The fracture and appearance of the surface were recorded in stereophotographs (Fig. 11). Approximately 3/8 in. of the specimen length was then dipped into 12N nitric acid to obtain an analytical specimen for direct determination of surface burnup.

III. DISCUSSION OF RESULTS

The measured changes induced in the wafer and pin specimens by irradiation are summarized in Table I, together with estimated total atom burnups. (For burnup computations and tabulation of actual measurements see Appendices A and C.) The small size of the wafers and sticking of the barrier foils combine to make the accuracy and meaning of measurements uncertain.

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Fig. 10. Specimen Pin and Its Capsule Before and After Irradiation
(0.19% total atom burnup.)

Macro 11,583

Macro 13,568

Macro 11,601

Macro 13,567

a) Appearance of pin before cladding and after stripping (approx. 1.5X)

b) Appearance of pin capsule before and after irradiation (approx. 1X)

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TABLE I

Summary of Irradiation Property Changes Induced in Wafer and Pin Specimens
(See Appendix C for actual measurements.)

Specimen	Loss in Weight (mg)	Decrease in Density (%)	Change in Diameter (%)	Change in Length (%)	Increase in Hardness (R "A" Points)	Average Total Atom Burnup ¹ (%)
U (Natural) Wafer	62	1.0	-0.3	-1.6	-	0.004
U ²³⁵ Wafer	156	1.0	2.5	-	-	0.17
U ²³⁵ - 10 w/o Zr Wafer	134	1.5	2.8	2.4	-	0.16
13% U ²³⁵ - 4-1/2 w/o Zr Pin	-	1.9	1.0	0.3	6.9	0.19

¹Burnup achieved by initial exposure of wafers, final exposure of pin.

All weight losses in the wafers were due to removal of material in the electropolishing operation after initial irradiation. The density decreases are understandable, except for the magnitude of the natural uranium change. The U²³⁵ and the U²³⁵ Zr alloy wafers show density change rates of about 6% and 9% per per cent total atom burnup, respectively, whereas the rate change in the natural uranium is of the order of 250%. This rate is obviously not realistic and the anomaly is heightened by the recorded negative changes in dimensions. Dimensional changes in the other wafers are in the right direction; surface roughening and the presence of barrier foil makes their magnitude greater than the density changes warrant.

It will be observed in Fig. 2 that the normal uranium and U²³⁵ wafers tended to remain circular in shape whereas the U²³⁵ Zr wafer became slightly oval. There is a reasonable doubt that the measurements of diameter show the true extent of this distortion.

It is of some interest to note in this connection that the U-Zr material under thermal cycling does not behave isotropically. Below 500 cycles there is a real difference between the average growth rates along and across the rolling direction. These rates decrease differentially. At 510 cycles they happen to be the same, but the trend at this point favors growth along the rolling direction rather than across it. The thermal cycling and irradiation growth effects are summarized for comparison in Table II; unfortunately, lack of definitive data for the irradiation specimen makes a real correlation impossible.

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TABLE II

Comparison of the Relative Behavior of U^{235} -10 w/o Zr Hot Rolled Alloy
Under Thermal Cycling and Under Irradiation
(See Appendix C for actual measurements.)

Growth Rates	Total Dose	Rolling Direction		
			⊥	Random
Average thermal cycling growth rate, G_t (Micro inches per inch per thermal cycle) ¹	10 cycles	190	280	-
	110 cycles	84	97	-
	510 cycles	42	42	-
Average irradiation growth rate, G_i (Micro inches per inch per ppm total atom burnup.)	0.16% burnup	(2)		17

¹Arbitrary heating and cooling cycle. See Table IX, Appendix C, for details.

²A circular disc became oval during irradiation, indicating that the irradiation growth rate is not the same in directions parallel (||) and perpendicular (⊥) to the rolling direction.

The use of tightly fitting capsules and barrier foils may have restrained small tendencies for the wafers and pin to change in dimensions during irradiation. For this reason the dimensional stability comparison between the various specimens is not really complete. However, the relative surface stabilities of the three wafer materials may be compared reasonably well by examining the appearance of the unrestrained edges. These are shown in Fig. 2 after the first and second exposures (0.7% and 1.8% surface depletion of U^{235} , respectively), and in Figs. 7-9 after maximum burnup (7.1% surface depletion of U^{235}) had been achieved. The zirconium alloy material proved to have a clear superiority in edge surface smoothness over the unalloyed U^{235} and also over the normal uranium even though the total atom burnup of the former was over forty times as great as that of the latter.

Some roughening of the specimens did occur on surfaces which were in close contact with the capsule, as shown by the profiles (Fig. 3). It should be mentioned, however, that the profiles of the enriched specimens were taken over the adhering barrier foils. There is the possibility that these were not completely joined to all points on the surfaces of the enriched wafers.

A woody type of roughening, which may be seen in Fig. 11, took place on the surface of the pin specimen. It was evidently suppressed at spots where the aluminum contact was tight. The two views in this figure

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Fig. 11. Stereo-Macrophotographs of Stripped Irradiated Pin Specimen
(0.19% total atom burnup.)

Macro 13,829

5X

a) View showing surface features and conchoidal fracture.

Macro 13,827

13X

b) Enlarged view of one end, showing that surface deformation was held down by restraint of the cladding.

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show some areas thinly coated with aluminum, conclusive evidence that surface adhesion between aluminum jacket and fuel metal did occur. The fracture surfaces of the pin were conchoidal, indicating that the material was brittle at room temperature.

The metallographic work, Figs. 4-6, was performed on a mockup basis under difficult experimental conditions, as may be judged from a reading of Appendix B. It was not entirely successful in defining the structures, but there is some indication of irradiation-induced changes both in the polarized light and bright field views. Increased chemical activity of the prepared surfaces is evident, together with pitting and staining problems produced by irradiation.

IV. CONCLUSIONS

The value of the two preliminary tests described above lies chiefly in the qualitative information obtained.

- 1) A 10 w/o Zr alloy of uranium showed much greater surface stability under irradiation than unalloyed uranium which had received the best known stabilizing treatment in its fabrication.
- 2) A $4\frac{1}{2}$ w/o Zr alloy of uranium showed excellent dimensional stability under irradiation in a totally confining finned capsule of 2S aluminum. It can be said that any tendency to change dimensions was successfully restrained by cladding only $1/32$ in. thick. This fuel alloy proved to be brittle at room temperature after the relatively low burnup of 0.19% of total atoms.
- 3) The tests demonstrated a tendency for the fuel materials to fission-weld to zirconium and aluminum when tight mechanical contact was made during irradiation.
- 4) Although the irradiation methods used had the advantage of rapid application, it is evident from the results of the study that unambiguous comparisons and quantitative data cannot be obtained from experimental arrangements which hold the fuel specimens in mechanical restraint.

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V. ACKNOWLEDGMENTS

We are indebted to Battelle Memorial Institute for the rolled U-10 w/o Zr material from which wafers were made. Actual fabrication of specimens was done by A. B. Shuck. Physical measurements were made by A. C. Klank and F. Pausche. Burnup analyses were made by E. H. Turk (Chemical Engineering Division). F. R. Taraba assisted in computations on flux and burnup. Thermal cycling of the U-Zr alloy was done by S. T. Zegler. S. Untermeyer (Laboratory Director's Staff) suggested the experiments and did much to expedite their performance.

VI. BIBLIOGRAPHY

1. S. Untermeyer, et al., "Reduction of Irradiation Damage to Uranium by Small Zirconium Additions (Interim Report)," ANL-4604 (March 27, 1954).
2. S. Untermeyer, "Effects of Heat Treatment and Alloy Additions on Irradiation Stability of Uranium Metal," TID-73, Reactor Science and Technology, (December 1951), pp 131-134.
3. F. L. Brown and S. H. Paine, "Pin Cushion Irradiation Tests of Uranium and Its Alloys," ANL-5538, (to be published).
4. J. H. Kittel and P. Tedeschi, "A Capsule Design for Experimental High Flux Irradiation of Fuel Materials," ANL-4900 (October, 1952).

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IRRADIATION HISTORY AND BURNUP COMPUTATIONS

Exposure Data

Computation of Burnup

The original estimates of U^{235} surface depletion are higher than those obtained from the radiochemical results. In the case of the normal uranium, the average depletion of U^{235} is also the surface depletion and turns out to be 5.6%. For the U^{235} specimen, the surface depletion is only 2.4%, and even allowance for the fact that flux depression in the region of the specimen was ignored in the estimate justifies a number only slightly higher, 2.6%. This is considerably lower than the 7.1% estimate based on unperturbed flux. Some of this difference doubtless can be accounted for by the $1/v$ effect in the flux spectrum, as the thimble in which the irradiation was conducted was adjacent to the fuel rods. However, the greater part of the difference must be rationalized as uncomputed flux depression in the enriched specimen. The large difference between surface depletion numbers cited above for normal uranium and enriched uranium specimens seems to agree with this explanation.

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TABLE III

Irradiation Data and Rough Depletion Estimates

Dates of Irradiations		MWH of Reactor Operation		Estimated % Surface Depletion of U ²³⁵
Charged	Removed	Interval Exposure	Total Exposure	
12- 7-50	3- 5-51	260.4	260.4	0.7
4-23-51	8-27-51	409.2	669.6	1.8
11-28-51	10-27-52	1274.1	1943.7	5.2
11-10-52	5-11-53	697.9	2641.6	7.1

TABLE IV

Computed Burnup of Total Atoms (Wafer Specimens)

Estimated % Surface Depletion of U ²³⁵	Per Cent Average Burnup of Total Atoms		
	Normal Uranium	U ²³⁵ (94.5%)	U ²³⁵ -10 w/o Zr
0.7	0.004	0.17	0.16
1.8	0.01	0.44	0.41
5.2	0.03	1.28	1.20
7.1	0.04*	1.74*	1.64

*By Cs¹³⁷ analysis; all other values by proportion from the estimated surface depletion numbers.

TABLE V

Flux Perturbation Factors for Wafer Specimens

Specimen	Uranium Content (Atom %)	Flux Perturbation Factors (Ratios)		
		$\frac{\text{Surface Flux}}{\text{Unperturbed Flux}}$	$\frac{\text{Average Flux}}{\text{Surface Flux}}$	$\frac{\text{Average Flux}}{\text{Unperturbed Flux}}$
Normal U	100		1.00	
U ²³⁵ (94.5%)	100	0.90	0.78	0.70
U ²³⁵ -10 w/o Zr	78	0.96	0.88	0.84

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II. ORNL-LITR IRRADIATION

The finned capsule irradiation started in October, 1951, and was terminated on December 23, 1952, the total elapsed time being 14 months. It was estimated by ORNL personnel that the sample had received a total of 2.5×10^{20} nvt (thermal) and should have a surface depletion of approximately 12% of the U^{235} atoms.

Radiochemical analysis of Cs^{137} content, based on an accurately titrated determination of residual uranium, gave a burnup of 0.224% of the total uranium, or 1.72% of the U^{235} . The chemical sampling came from the surface of the specimen and, therefore, indicates the depletion of U^{235} at the surface. Average total atom burnup, allowing approximately 5% self-shielding in the specimen and taking account of zirconium content (10.9 a/o), was computed to be 0.19%. The burnup analysis results for the pin are, therefore, as in the case of the wafers, unaccountably low.

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APPENDIX B.

DETAILS OF FOIL REMOVAL AND METALLOGRAPHY

Removal of Zirconium Foil

The zirconium foil adhered slightly to the normal uranium specimen, but was easily peeled off with a knife. The foils on the enriched specimens were firmly adherent and could not be removed in the same manner. An attempt was then made to abrade the foil from the enriched uranium. This method was found to be tedious and time consuming. Other methods were considered, and it was decided to try electrolytic attack followed by abrasion, a method which proved successful and is described in detail.

Most of the foil was removed electrolytically in a perchloric-acetic acid bath (see Table VI). However, cold experiments had indicated that the uranium would be attacked equally as well as zirconium, and, therefore, electrolysis of the irradiated specimens was discontinued short of the time required for complete removal. Final removal of foil and initial rough polishing was done by abrading on Behr Manning grade 400 Speed Wet Durite abrasive paper wetted with kerosene-paraffin solution.

TABLE VI

Electrolytic Polishing Procedure

Electrolyte	Material	Radio-Activity	EMF, volts	Current ma/cm ²	Time, min
89% Acetic acid 9-1/2% Perchloric acid (70%) 1-1/2% Distilled water	Zirconium	None	11-1/2	550	0.3 mil removed per minute (on thickness)
	2-mil zirconium foil on U ²³⁵	25 R at 8"	11-1/2	400	5
	2-mil zirconium foil on U ²³⁵ -10% Zr	25 R at 8"	11-1/2	400	9
	U ²³⁵ -10% Zr	None	16	210	1-1/2
	U ²³⁵ -10% Zr	25 R at 8"	24	800	1/2
28% Orthophosphoric acid 28% Ethylene glycol 44% Ethyl alcohol	U ²³⁸	None		30	10 minutes on polished surface
	U ²³⁸	7-1/2 R at 8"	2-3/4	24	120
	U ²³⁵	None	2-3/4	24	120
	U ²³⁵	25 R at 8"	2-3/4	24	120

A specimen holder was devised to facilitate abrading with master-slave manipulators. This holder consisted, principally, of a 1/2" thick slab of brass with a depression in the center of a flat face. The specimen was

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cemented into the depression so that the surface to be polished projected slightly above the flat surface of the brass slab. In operation the holder (with specimen cemented into place) was inverted and stroked remotely on the abrasive paper. A screw system was provided whereby the specimen could be lowered or raised in the depression to control the amount of metal removal.

Rough Polishing

Following abrasion on the grade 400 paper, the enriched specimens were abraded on grade 600 Speed Wet Durite paper wetted with kerosene-paraffin solution. The specimens were rinsed with alcohol after each abrasion operation and were removed from the holder at the completion of rough polishing by dissolving the cement with acetone.

Final Polishing

Final polishing was done electrolytically. Conventional phosphoric acid electrolyte was used on the normal and enriched specimens; perchloric-acetic electrolyte was used on the uranium-zirconium alloy specimen (see Tables VI and VII). It was noted that the irradiated specimens did not respond to electrolytic polishing in quite the same manner as the corresponding unirradiated material; however, this discrepancy may have been caused by slight differences in the electrical contacts, as considerable difficulties were experienced in remote handling of the specimen clamping device.

TABLE VII

Electrolytic Etching Procedures

Electrolyte	Material	Radioactivity	EMF, volts	Current, ma/cm ²	Time, min
89% Acetic acid 9-1/2% Perchloric acid (70%) 1-1/2% Distilled water	U ²³⁵ -10% Zr	None	6	100	1
28% Orthophosphoric acid 28% Ethylene glycol 44% Ethyl alcohol	U ²³⁸ U ²³⁸	7-1/2 R at 8" 7-1/2 R at 8"	1-1/2 1-1/2	30 30	60

Preliminary examination after final polishing was made in the cave, using a bench microscope outfitted with a periscope. Photomicrographs were made outside of the cave with a Bausch & Lomb metallograph. A special uranium casket was made to shield the specimens during transit between cave and camera and during photomicrography. The specimens were cemented into place in this casket.

Photomicrography

Photomicrographs of the irradiated specimens and of corresponding unirradiated material are shown in Figs. 4-6.

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APPENDIX C

EXPERIMENTAL DATA

TABLE VIII

Properties of Specimens Before and After Irradiation¹

Specimen	Weight (g)		Density (g/cc)		Diameter (in.)		Length (in.)		Rockwell A Hardness	
	Before	After	Before	After	Before	After	Before	After	Before	After
U (Natural) Wafer	0.4968	0.4348	19.219	19.008	0.2515	0.2501	0.0326	0.0330 ²		
	.4967	.4352	19.214	18.999	.2518	.2518	.0325	.0320		
	.4969	.4345	19.187	18.999	.2523	.2519	.0324	.0317		
	.4968		19.191		.2531	.2520	.0324	.0319		
								.0314		
	0.4968	0.4348 ³	19.197 ⁴	19.002	0.2522	0.2515	0.0325	0.0320	(Averages)	
U ²³⁵ Wafer	0.4913	0.3360	18.903	18.670	0.2512	0.2600	0.0331	-		
	.4913	.3359	18.955	18.791	.2506	.2560	.0328	-		
	.4914	.3355	18.842	18.693	.2518	.2570	.0328	-		
	.4912		19.160		.2520	.2578	.0328	-		
	0.4913	0.3358 ³	18.900 ⁴	18.718	0.2514	0.2577	0.0329		(Averages)	
U ²³⁵ -10 w/o Zr Wafer	0.3558	0.2217	15.919	15.657	0.2469	0.2535	0.0297	0.0318 ²		
	.3559	.2217	15.667	15.648	.2467	.2535	.0297	.0301		
	.3560	.2216	15.881	15.656	.2469	.2562	.0296	.0304		
	.3560		15.869		.2473	.2529	.0295	.0295		
					.2469			.0305		
					.2471			.0296		
								.0301		
	0.3559	0.2217 ³	15.890 ⁴	15.654	0.2470	0.2540	0.0296	0.0303 ⁵	(Averages)	
U ²³⁵ -4-1/2 w/o Zr Pin	7.1351 ⁶	7.1362 ⁶	17.897	17.570	0.1246 ⁷	0.1258	2.0010 ⁴	2.007	63.4 to 64.2	69.0 to 73.8
			17.897	17.548						

¹After initial irradiation of wafers (0.7% surface depletion of U²³⁵) and final irradiation of pin (0.19%) total atom burnup.

²Thickness at center of wafer; other measurements made closer to edges.

³Weights taken after removal of metal by electropolishing.

⁴Average of three only.

⁵Thickness measurements include foil barriers.

⁶Average of two.

⁷Average of six.

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TABLE IX

Dimensional Changes Induced in a Rolled U-10 w/o Zr
Alloy Specimen by Thermal Cycling^{1,2,3}

	Length (inches)	Change in Length (in.)	$\frac{L-L_0}{L_0} \times 100$	Width (inches)	Change in Width (in.)	$\frac{W-W_0}{W_0} \times 100$
Before Cycling	1.070 (L_0)			0.5044 (W_0)		
After 10 Cycles	1.072	0.002	+0.19	0.5058	0.0014	+0.28
After 110 Cycles	1.080	0.010	+0.93	0.5098	0.0054	+1.07
After 510 Cycles	1.093	0.023	+2.15	0.5152	0.0108	+2.14

¹Holding alternately in 50°C and 550°C NaK for periods of 5 min, with transfer times of 5 sec between hot and cold zones.

²No measurable changes occurred in thickness of specimen, initially 0.039" thick.

³Before cycling, specimen was annealed 18.5 hr at 575°C in vacuum. The following Rockwell C hardness observations were made: As received, R_C 37-43; after 2 hr at 575°C, R_C 30; after 18.5 hr at 575°C, R_C 25-30; after 510 thermal cycles, R_C 16-30. It is believed that the scatter in annealed hardness readings is due to surface roughness of the specimen.

Discussion

The work was complicated by the nonexistence of established electrolytic polishing procedures for uranium-zirconium alloys, and by the lack of time and unavailability of irradiated material to determine fully satisfactory procedures for irradiated uranium as well as irradiated uranium-zirconium alloy. Furthermore, handling of the small specimens was extremely difficult with available cave equipment; available time did not permit designing and building more adequate tools.

A further difficulty was the limitation on the field of view imposed by the special holder used as a shield on the metallograph. The specimen was quite small, yet not more than 40% of the polished area could be examined. In the examination of important specimens of this type it is desirable to be able to examine the entire polished face.

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