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EXAMINATION OF URANIUM-2 w/o ZIRCONIUM EXPERIMENTAL  
FUEL SLUGS IRRADIATED IN EBR-I

by

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ABSTRACT

Six groups of uranium-2 w/o zirconium fuel slugs were irradiated in the first core of the EBR-I to burnups of 0.080 to 0.189 a/o at calculated temperatures of 307 to 383°C. Two groups of cast specimens were found to be more dimensionally stable than four groups of wrought slugs. Of the wrought slugs, the "as quenched" group showed less tendency to grow than the three groups which had some annealing after quenching. Specimens at burnups of about 0.189 a/o and at 383°C showed the onset of swelling as indicated by density measurements. The hardnesses of these specimens seemed but little affected by radiation, but there was an indication of softening with increasing irradiation temperature.

INTRODUCTION

Untermeyer<sup>(1)</sup> had shown in 1951 that small additions of zirconium to uranium would improve its dimensional stability under irradiation. Subsequently, Paine, Brown and Murphy,<sup>(2)</sup> Paine and Brown,<sup>(3)</sup> and Kittel and Paine<sup>(4)</sup> extended the experimental irradiation of uranium containing small additions of zirconium and were able to delineate more sharply useful compositions of the binary alloy. Concurrently, Zegler, Chiswik, Mueller, and Macherey<sup>(5)</sup> had studied the behavior of uranium-zirconium alloys under thermal cycling. The latter concluded that, although surface stability was generally improved, wrought uranium-zirconium alloys were not more dimensionally stable than beta-quenched unalloyed uranium. They found, however, that cast alloys did show an improvement in dimensional stability, being at least comparable to beta-quenched unalloyed uranium.

The results of the "pin cushion" irradiation tests by Paine and Brown and of the thermal cycling tests by Zegler *et al.*, indicated that an alloy of uranium-2 w/o zirconium was a promising fuel material for a fast reactor.

A number of experimental fuel rods containing enriched uranium-2 w/o zirconium in various heat-treated conditions were made up<sup>(6)</sup> and irradiated in the first core of the Experimental Breeder Reactor (EBR-I), a cross section of which is shown in Figure 1. The results of the tests are given in this report.

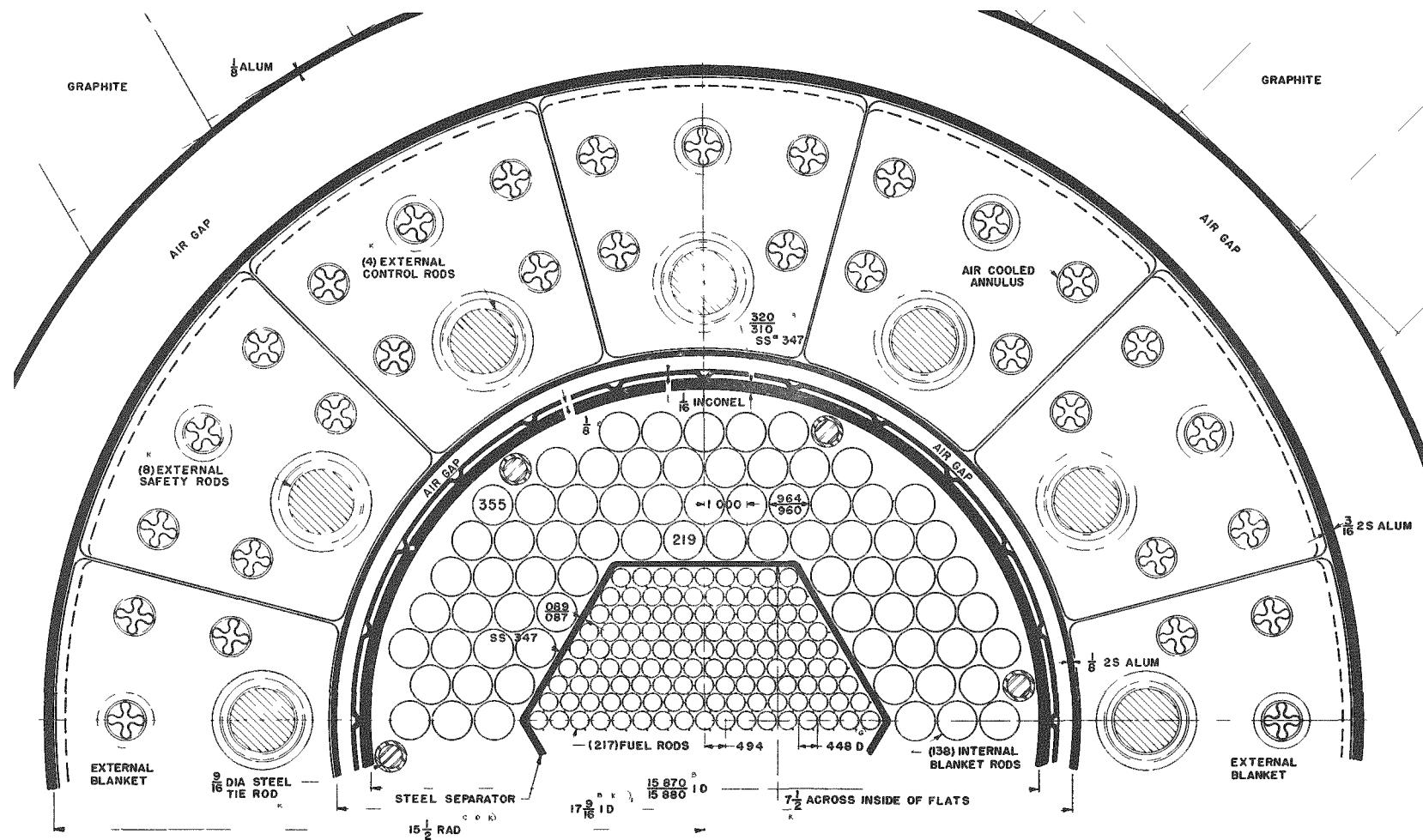


Figure 1. Horizontal Cross Section of EBR-I at Midplane

## HISTORY OF THE SPECIMENS

The experimental enriched uranium-2 w/o zirconium slugs were fabricated, heat treated, and assembled into fuel rods under the direction of A. B. Shuck.(6) A summary of the irradiated rods with 2 w/o zirconium in the fuel was as follows:

<u>Rod Numbers</u>	<u>Loading Scheme (Fig. 3)</u>	<u>Blanket Slugs</u>	<u>Fuel Slugs</u>
62-545-1 through 6	1	Natural U-2 w/o Zr	$U^{235}$ -2 w/o Zr
62-545-7 through 10	2	Natural U-2 w/o Zr	$U^{235}$ -2 w/o Zr
62-560-1	2	Natural U	$U^{235}$ -2 w/o Zr

The raw materials for the alloy fuel slugs were crystal bar zirconium wire and buttons of enriched uranium. They were melted together and cast into water-cooled copper molds. Casting No. 16F-52 yielded fourteen wrought slugs for the 545 series. The following procedures indicate the steps in fabrication:

1. The 1.300-in.-diameter casting was vacuum annealed for one hour at 800°C, furnace cooled to 550°C and held at that temperature for 2 hr, and then furnace cooled to room temperature.
2. The casting was then rolled to 0.836-in. diameter at 300°C in six passes.
3. The rod was reannealed and then rolled from a diameter of 0.836 in. to one of 0.520 in. in six passes.
4. It was again reannealed, rolled to 0.390-in. diameter, and separated into two parts.
5. One part was hot swaged from 0.390 in. to the finished diameter, 0.384 in.
6. The other portion was rolled to 0.375 in. and hot swaged to the finished diameter, 0.364 in.
7. The slugs were cut to length from the swaged rods.

All the wrought slugs in the 545 series, with but two exceptions, were made according to the above fabrication schedule. The two exceptions were the slugs in position 2 in rods 545-7 and 9. These two slugs came from casting 16F-62, which was of  $\frac{5}{8}$ -in. diameter. This casting was annealed and rolled to 0.480-in. diameter as described for casting 16F-52. It was then rolled to a 0.390-in. diameter and swaged to a 0.384-in. diameter. The two slugs were machine cut from the swaged rod.

The chill-cast slugs in the 545 series rods were made individually. Water-cooled, split copper molds with a funnel-shaped riser at the top were used. The molds were either 0.375 or 0.395 in. in diameter and 4 in. deep. Cold castings had diameters of 0.365 and 0.384 in., respectively. Eight castings, Nos. 16F-53, 54, 56 through 60, and 63, were made. They were very bright, with smooth surfaces. Six of the finished slugs had densities between 18.03 and 18.08 gm/cm<sup>3</sup>, which were comparable with that of wrought material. Two with densities of 17.75 and 17.95 gm/cm<sup>3</sup> evidently had some porosity.

The zirconium compositions of the 545 series fuel slugs ranged from 2.01 to 2.17 w/o. The degree of enrichment of the uranium was from 93.20 to 93.26 w/o.

The fuel slugs in the 560-1 rod were made from casting No. 16F-64. The fabricating procedures were essentially as described for casting 16F-52. The zirconium composition was 1.99 w/o and the uranium was enriched 91.17%.

The fuel slugs going into the various rods were heat treated as noted in Table I.

Table I

## IDENTIFICATION OF IRRADIATED ENRICHED URANIUM 2 w/o ZIRCONIUM ALLOY FUEL SLUGS

Fuel Rod Designation <sup>(1)</sup>	Radial Location of Rod in Reactor cm	Reactor Output, kwh	Burnup for Fuel Section of Rod, a/o	Slug Position in Rod (Fig. 3)	Heat Treatment <sup>(2)</sup>	Burnup per Slug, a/o
S-1-62-545-1	1 255	2,066 231	0 189	2	1	0 189
				3	2	0 189
S-1-62-545-2	1 255	2 066,231	0 189	2	3	0 189
				3	4	0 189
S-1-62-545-3	1 255	1,234 030	0 113	2	5	0 113
				3	6	0 113
S-1-62-545-4	2 174	2,066,231	0 187	2	1	0 187
				3	2	0 187
S-1-62-545-5	1 255	2 066 231	0 189	2	3	0 189
				3	4	0 189
S-1-62-545-6	1 255	2 066,231	0 189	2	5	0 189
				3	6	0 189
S-2-62-545-7	7 836	2 066 231	0 147	2	1	0 132
				3	2	0 171
				4	3	0 138
S-2-62-545-8	7 633	1,234,030	0 089	2	4	0 080
				3	5	0 103
				4	6	0 084
S-2-62-545-9	7 633	1,824,432	0 131	2	1	0 118
				3	2	0 152
				4	3	0 123
S-2-62-545-10	7 633	1,995 564	0 144	2	4	0 129
				3	5	0 167
				4	6	0 136
S-2-62-560-1	7 633	1,824 432	0 131	2	3	0 118
				3	3	0 152
				4	3	0 123

(1) First number after the 'S' designates loading scheme as shown in Figure 3

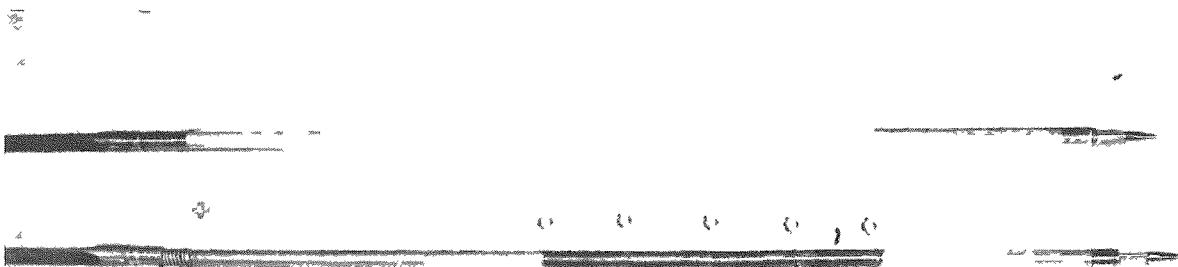
(2) Heat Treatments

- 1 Wrought heated 1 hr at 800°C water quenched
- 2 Wrought, heated 1 hr at 800°C, water quenched, 24 hr at 550°C
- 3 Wrought heated 1 hr at 800°C, air cooled in bomb
- 4 Wrought heated 1 hr at 800°C, quenched in 550°C lead bath for 1 hr, air cooled in bomb
- 5 Chill cast slugs
- 6 Chill cast slugs, annealed 1 hr at 550°C, vacuum cooled in cold chamber

The slugs were electropolished before placing them in the rods. None of the slugs was machined to diameter.

Weights and densities of the slugs were determined prior to putting them into a rod. Lengths and diameters were measured by means of a comparator and gauge blocks. A. B. Shuck supervised the preirradiation measurements.(6)

The assembled fuel rods were very similar to the rods with unalloyed uranium which composed the first core of EBR-I (Figure 2). A column of uranium, 20.25 in. long, was contained in a stainless steel tube. The column was spring loaded by a coil of stainless steel wire at the top. Tantalum spacers separated the slugs from one another. A NaK annulus of 0.010 or 0.020 in., depending on the diameters of the slugs, served as a heat transfer medium from the uranium to the stainless steel tube. The sizes of the slugs are indicated in Figure 3.



AS-28A

Approx.  $\frac{1}{4}$ X

Figure 2. A Typical EBR-I Fuel Rod Assembly. Upper rod is the assembled and welded fuel rod. Below it is the assembly of uranium slugs as inserted into the fuel tube. The spacers are shown separately.

These fuel rods were put into the reactor in the locations shown in Figure 4. Rods 545-1 through 8 were put into the reactor on January 13, 1953. Fuel rod 545-10 was inserted on January 20, Rods 545-9 and 560-1 were put in on February 5, 1953. With the exception of rods 545-3 and 8, all rods remained in the reactor until the end of the first core on January 24, 1954. Rod 545-3 was removed from the reactor and radio-graphed on April 8, 1953. Since the spring was not completely compressed by growth of the fuel, the rod was replaced in the reactor. On July 28, 1953, rods 545-3 and 8 were removed from the reactor and disassembled in the EBR hot cell. The kilowatt-hours accumulated by the reactor while these rods were in the core are indicated for each rod in Table I. During the period covered by the irradiation of the experimental fuel rods, the reactor was operated on a power-producing basis, except for a period of about one month around March 1953.



During power runs, a typical output was about 1158 kw thermal. The NaK inlet and outlet temperatures were 228 and 316°C, respectively, at a flow rate of 292 gpm.

After the irradiated rods were removed from the reactor, they were taken apart in the EBR hot cell. The fuel slugs from each rod were placed in an aluminum tube with plug closures at each end. The loaded aluminum tubes were shipped to the Lemont site in a shielded container in which they were identified by position. The slugs were there examined in the Metallurgy hot cells.

## RESULTS

When the aluminum tubes containing the irradiated enriched uranium-2 w/o zirconium slugs were removed from the shielded shipping cask at the Lemont site, the bottom plugs fell out of four, so that the slugs dropped out. This resulted in a mix-up of the slugs in each of these four tubes. The fuel rods involved were series 545, rod numbers 7, 8, 9, and 10. The slugs were identified by checking their weights against the preirradiation weights. Since each of these rods contained three fuel slugs, the radiation readings helped to identify the middle slug by its higher radioactivity as compared with the slugs above and below it in the rod. Identifying numbers on the ends of the fuel slugs were not sufficiently clear to be of much help. However, it is believed that the measurement data as given in this report are properly associated with the correct slugs and their positions in the reactor. Identification of the slugs is given in Table I.

When the slugs were removed from the aluminum tubes, they had a loose black powder on their surfaces. They were immediately weighed with whatever of the powder that remained with them. Then the slugs were scrubbed and rinsed with acetone, dried, and reweighed. The postirradiation weights are compared with the preirradiation weights in the Appendix. Also included in the Appendix are pre- and postirradiation lengths, diameters, and densities, as well as postirradiation hardnesses and radioactivity.

All the postirradiation, as-received, weights, with one exception, were greater than the preirradiation weights by amounts ranging from 1 to 41 mg. The single exception lost 2 mg. The acetone wash removed from 2 to 129 mg of material. The weights of the washed samples were generally less than the preirradiation weights by 1 to 118 mg. However, 5 of the 27 slugs examined gained from 2 to 26 mg over their original weight, and one slug showed no change. The changes of weight did not correlate with any recognized variables.

The procedures for measuring lengths and diameters are given in the Appendix. Since the ends of the slugs were machined, the preirradiation

length measurements were quite accurate. The slugs were relatively free from distortion after irradiation, and observed differences in repetitive measurements of length on the irradiated slugs ranged from 0.3 to 2.8 mils, with an average of 1.3 mils.

The sides of the slugs were surfaces which had been swaged or cast and then electropolished. These methods of fabrication resulted in variations in the diameters of each slug. The preirradiation diameters were recorded as maxima and minima. The differences between maximum and minimum values for the slug diameters ranged from 0.4 to 4.0 mils, the average being 1.7 mils. The largest variations in diameter were generally exhibited by the cast slugs. The water-quenched slugs were next, and the quenched and tempered slugs showed the smallest differences in diameters.

It was not practical to determine maximum and minimum diameters of the irradiated specimens. Consequently, averages of from 4 to 8 diameter measurements have been compared with the averages of the maximum and minimum diameters of the preirradiation data. The variation in diameter measurements after irradiation ranged from 0.4 to 10.0 mils, with an average of 2.8 mils. There is a tendency for the postirradiation measurements to reflect the preirradiation measurements with respect to slug history; that is, the irradiated cast slugs showed more variation in diameters than the others and the quenched and tempered slugs the least.

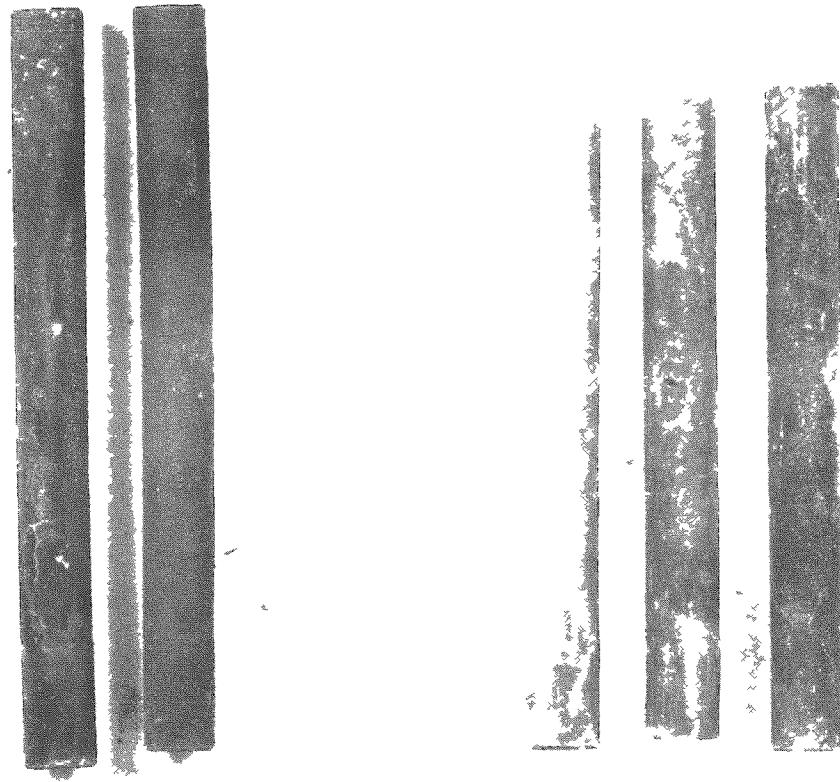
Both the length and diameter measurements after irradiation would be affected by the degree of surface roughening. Profile traces as shown in the Appendix indicate that this factor is relatively unimportant for these slugs. Profile traces were taken on representative slugs after irradiation. No preirradiation profiles had been taken, but traces of similarly prepared slugs were essentially smooth curves with only gradual slope changes along the lengths. The irradiated slugs showed some surface roughening, particularly in the slugs toward the bottom of the rods. The maximum variation observed on the short length of the traces was about 1.5 mils. A gradual change of about 3 mils was shown by one slug.

Pre- and postirradiation densities are given in the Appendix. The small weight changes and the fact that most slugs lost weight indicate that corrosion products on the surfaces of the slugs probably had only a slight effect on the determinations of postirradiation density.

Rockwell A hardness measurements of the irradiated slugs were made. No preirradiation hardness measurements had been made. The total spread of the hardnesses was from 62 to 76.5 Rockwell A for all conditions of heat treatment. Considering that six different preirradiation histories are involved, as well as differences in burnup and irradiation temperature, the relatively small spread in hardness values made it difficult to draw any conclusions.

The radiation readings described in the Appendix were taken for general interest. They were of some help in correcting a mix-up in identities as previously mentioned.

Photographs of the slugs from one rod of each loading scheme are shown in Figure 5(a) and (b). No gross distortion of the slugs was evident, although differences in length were obvious. The surfaces appeared generally clean and smooth.



16507

1X

(a) Rod S-1-62-545-4

16508

1X

(b) Rod S-2-62-545-10

Figure 5. Photographs of Typical Experimental Uranium-2 w/o Zirconium Fuel Slugs Irradiated in the First Core of EBR-I. In each photograph the slug at the left is from the top of the fuel column; slug at right is from the bottom.

The per cent of total atom burnup for the fuel sections of the rods examined has been calculated by extrapolation of the results obtained by radiochemical analyses<sup>(7)</sup> of unalloyed enriched uranium slugs removed from the reactor after a total of 412,588 kwh of thermal energy had been

produced by the core plus the inner blanket. Then, on the basis of fission rate curves at different radial locations,<sup>(8)</sup> the per cent burnup of total atoms per slug was calculated. The results are given in Table I.

Average slug temperatures were calculated on the basis of the burn-up rate and the temperatures of the NaK flowing past the fuel rods during reactor operation at 1158 kw. Approximate values were used for the temperature drop due to the stainless steel cladding, the NaK annulus, and the film coefficient.<sup>(9)</sup> Simplifying assumptions were made. The average temperatures and the temperature variation of slugs at the different locations in the reactor are given in Table II.

Table II

CALCULATED TEMPERATURES OF URANIUM-  
2 w/o ZIRCONIUM ALLOY SLUGS

Radial Location of Rod in Reactor, cm	Slug Position in Rod (Fig. 3)	Calculated Temperature Range, °C <sup>(1)</sup>	Volume Average Temperature, °C
1.255	2	279-409	339
	3	334-419	383
2.174	2	279-409	338
	3	334-419	382
7.633	2	261-366	309
	3	302-403	354
	4	337-404	374
7.836	2	261-365	307
	3	301-402	353
	4	337-404	374

(1) Temperature range is from the lowest surface temperature to highest central temperature, assuming there was no conduction along length.

For purposes of facilitating comparisons, the data for the slugs have been collected according to the heat treatment they received. This information is given in Table III.

Table III  
MEASUREMENTS DATA FOR IRRADIATED URANIUM-2 w/o ZIRCONIUM FUEL SLUGS

Preirradiation History <sup>(1)</sup>	Nominal Slug Diameter, in.	Burnup per Slug, a/o	Average Temperature, °C	% Length Change	Length Change $\Delta L$		% Diameter Change	Diam. Change Burnup microin./in. ppm Burnup	% Diam. Change % Length Change	% Density Decrease	Volume Increase, % Burnup, a/o	Hardness, RA
					Burnup	microin./in. ppm Burnup						
1	0.385	0.118	309	-0.39	-3.3	+1.04	+8.8	-2.67	0.25	2.1	69	
	0.385	0.132	307	0.00	0.0	+1.30	+9.8	-	0.31	2.4	72	
	0.365	0.187	338	+1.31	+7.0	+0.82	+4.4	+0.62	0.75	4.1	75	
	0.365	0.189	339	+0.99	+5.2	+0.54	+2.9	+0.55	0.71	3.8	70	
2	0.385	0.152	354	+1.05	+6.9	-0.26	-1.7	-0.25	1.00	6.6	67	
	0.385	0.171	353	+2.65	+15.5	-0.78	-4.6	-0.29	0.74	4.4	71	
	0.365	0.187	382	+2.96	+15.8	0.00	0.0	0.00	1.71	9.3	66	
	0.365	0.189	383	+2.55	+13.5	+0.54	+2.9	+0.21	2.80	15.3	64	
3	0.385	0.118	309	+3.01	+25.5	-1.04	-8.8	-0.34	0.46	3.9	71	
	0.385	0.123	374	+1.26	+10.3	-0.26	-2.1	-0.21	0.59	4.2	67	
	0.385	0.123	374	+1.29	+10.5	+1.04	+8.5	+0.81	0.76	6.2	65	
	0.385	0.138	374	+1.18	+8.5	0.00	0.0	0.00	0.76	5.6	67	
	0.385	0.152	354	+2.58	+16.5	-0.52	-3.4	-0.20	0.75	5.0	68	
	0.365	0.189	339	+2.59	+13.7	0.00	0.0	0.00	1.28	6.9	70	
	0.365	0.189	339	+2.53	+13.4	-0.27	-1.4	-0.11	1.10	5.9	68	
4	0.385	0.080	309	+1.35	+16.8	0.00	0.0	0.00	0.34	4.3	68	
	0.385	0.129	309	+2.09	+16.2	0.00	0.0	0.00	0.60	4.7	70	
	0.365	0.189	383	+2.12	+11.2	+0.55	+2.9	+0.26	2.16	11.7	65	
	0.365	0.189	383	+2.05	+10.8	+0.82	+4.3	+0.40	1.70	9.2	64	
5	0.385	0.103	354	-0.03	-0.3	+0.78	+7.6	-26.00	0.39	3.8	69	
	0.365	0.113	339	0.00	0.0	+0.27	+2.4	-	0.53	4.8	70	
	0.385	0.167	354	-0.12	-0.7	+0.78	+4.7	-6.50	1.10	6.7	69	
	0.365	0.189	339	-0.30	-1.6	+1.64	+8.7	+5.46	1.53	8.2	68	
6	0.385	0.084	374	+0.09	-1.1	+0.26	+3.1	+2.88	0.47	5.6	66	
	0.365	0.113	383	+0.16	+1.4	+1.09	+9.0	+6.81	0.82	7.4	67	
	0.385	0.136	374	0.00	0.0	+0.78	+5.7	-	0.78	5.7	68	
	0.365	0.189	383	+0.59	+3.1	+1.64	+8.7	+2.78	2.85	15.5	65	

## (1) Heat Treatments

1. Wrought; heated 1 hr at 800°C, water quenched
2. Wrought; heated 1 hr at 800°C, water quenched, 24 hr at 550°C
3. Wrought; heated 1 hr at 800°C, air cooled in bomb
4. Wrought; heated 1 hr at 800°C, quenched in 550°C lead bath for 1 hr, air cooled in bomb
5. Chill cast slugs
6. Chill cast slugs; annealed 1 hr at 550°C, vacuum cooled in cold chamber

## DISCUSSION

The program was specifically intended to determine if any of various treatments resulted in an improvement in the dimensional stability of a uranium-2 w/o zirconium alloy when irradiated. Basically, the treatments resulted in variations in the fineness of the microstructures and in degree of approach to phase equilibrium.

Four groups of specimens were heated to 800°C, at which a stable gamma phase exists. They were returned to room temperature by cooling in various ways as noted in Table I. Two groups were chill cast, and one of these groups was then annealed for one hour at 550°C. At all temperatures below about 610°C the equilibrium alloy would consist of a mixture of alpha and delta phases. None of the treatments allowed the system to achieve a condition of phase equilibrium.

This alloy, as quenched from 800°C, had a martensitic alpha structure supersaturated with zirconium.(5) Reheating to 550°C and holding for 24 hr resulted in coarsening of the martensitic needles and the formation of some spheroids of delta phase in the martensitic matrix. Slow cooling

from 800°C resulted in a coarse martensitic structure and the beginning of delta spheroid formation. Quenching from 800° into a lead bath at 550°C and holding for one hour produced a structure very similar to the previous two.

The chill-cast alloy had a fine martensitic structure. Some coarsening of the martensitic structure occurred when chill-cast specimens were annealed for one hour and vacuum cooled.

In addition to the treatments given to the alloy slugs, differences in temperature and burnup rate in the different locations in the reactor core also had to be considered as variables with respect to the measured changes in the fuel slugs. The temperatures involved in irradiation (Table II) probably promoted an approach to phase equilibrium.

Complete compression of the loading spring by the fuel did not take place in any of these rods. Of the ten rods of the 545 series, the fuel sections, excluding the blanket ends, had total measured elongations of from 0.006 in. to 0.177 in. The fuel section of the 560 series rod had a total elongation of 0.229 in. Since the allowable elongation was about 0.25 in., after making allowance for the blanket end sections, none of the springs in this set of fuel rods was completely compressed.

In Table III the data have been grouped according to the preirradiation histories and are arranged in the order of increasing burnup within each group.

Examination of the data from length measurements reveals that the cast slugs (Histories 5 and 6) had the smallest length changes (-0.12 to +0.59%; average +0.12%). The "as quenched" specimens (History 1) were next in order (-0.39 to +1.31%; average +0.48%). Slugs representing Histories 2, 3, and 4 were quite alike and showed greater length changes than those previously mentioned. Their length changes ranged from +1.05 to +3.01%, with an average of +2.08%.

The growth rate as indicated by the ratio of the length change to burnup was calculated for each slug. The cast slugs had values ranging from -0.7 to +3.1 microin./in.-ppm burnup. The average was +0.8. The chill-cast slugs had an average value of +0.2 microin./in.-ppm compared with +1.4 for the chill-cast and annealed slugs. The growth rate for the "as quenched" slugs ranged from -3.3 to +7.0 microin./in.-ppm burnup with an average of +2.2. The range of growth rate values for slugs with Histories 2, 3, and 4 was +6.9 to 25.5 microin./in.-ppm burnup. The averages were as follows: History 2, +12.9; History 3, +14.0; History 4, +13.8 microin./in.-ppm.

There is some indication of trends associated with temperature, but the picture is complicated by the fact that the slugs in the high-temperature regions were almost always those with the small diameters. However, there were seven specimens with History 3 (slowly cooled from 800°C), and five of them were 0.385 in. in diameter. There seems to be a clear indication for these five slugs that the ratio of length change per unit of burn-up decreases with increasing temperature. Considering length change alone, the colder the slug, the greater the length increase for this group of specimens.

The effects of thermal cycling on length changes of uranium-2 w/o zirconium specimens have been reported<sup>(5)</sup> for different heat treatments. The results are shown in Table IV. The heat treatments were similar to those given the experimental fuel slugs. The correspondence between the two sets of data is poor. There is a tendency for the specimens whose cooling from 800°C was interrupted to shorten on thermal cycling and to elongate on irradiation. The chill-cast samples tended to elongate and the water-quenched samples tended to shorten when thermally cycled; on irradiation, similarly treated slugs showed a mixed response, some shortening, some elongating, some not changing in length.

Table IV  
THERMAL CYCLING GROWTH AND HARDNESS RESULTING FROM VARIOUS TREATMENTS OF URANIUM-2 w/o ZIRCONIUM<sup>(5)</sup>

Treatment	% Growth $\frac{(L_n - L_0)}{L_0} \times 100\%$			Hardness <sup>(b)</sup> DPH - 100-gm Load			
	Cycles <sup>(a)</sup>			Edge	Mid-radius	Center	Average
	N = 250	N = 500	N = 1000				
Rolled at 300°C, heated 1 hr at 800°C, water quenched	-0.67	-1.04	-1.39	312 (66)	387 (70)	390 (70.5)	357 (68.5)
Rolled at 300°C, heated 1 hr at 800°C, air cooled	Nil	-0.15	-0.20	314 (66)	319 (66.5)	314 (66)	314 (66)
Rolled at 300°C, heated 1 hr at 800°C, 1 hr at 575°C, air cooled	-0.07	-0.22	-0.10	283 (64)	283 (64)	287 (64.5)	287 (64.5)
Rolled at 300°C, heated 1 hr at 800°C, 1 hr at 500°C, air cooled	Nil	Nil	Nil	266 (62.5)	287 (64.5)	302 (65.5)	281 (64)
Rolled at 300°C, heated 1 hr at 800°C, furnace cooled	+0.75	+1.86	+3.28	292 (64.5)	302 (65.5)	276 (63.5)	288 (64.5)
Chill cast	+0.25	+0.40	+0.50	333 (67)	302 (65.5)	302 (65.5)	316 (66)
Chill cast, heated 2 hr at 550°C, furnace cooled	+0.35	+0.60	+0.90	319 (66.5)	306 (65.5)	283 (64)	304 (65.5)

(a) Cycled between 50-550°C, 15 min heating, 5 sec cooling, 2 min holding time at upper and lower temperatures.

(b) Values in parentheses are Rockwell A conversion values.

Assuming that no volume changes have occurred and that the measurements have an equal degree of validity, the change in the diameter should be opposite to the change in length. Examination of the data in Table III shows that this rule does not generally hold, and it can be assumed that swelling and, to some degree, surface roughening of the specimens has occurred.

Profiles shown in the Appendix (Figures 9 and 10) do show that some slight roughening had occurred, particularly in the slugs in position 2 (bottom of fuel section). The specimen that showed the greatest surface roughness of those examined had a measured diameter increase of 1.30% (545-7, slug 2, History 1). Specimens with Histories 1, 5, and 6 increased in diameter to about the same extent; the averages were +0.92%, +0.87%, and +0.94%, respectively. Length changes for these specimens were both plus and minus. Diameter changes for slugs with History 4 ranged from 0.0% to +0.82% with an average of +0.34%; length changes were all positive. The slugs with Histories 2 and 3 had diameter decreases generally. The averages were -0.13% and -0.15%, respectively, but plus values are included. Again, there were indications of an effect due to either temperature or slug diameter.

The ratios of diameter change to burnup for the various slugs confirm the indications given by the per cent diameter changes.

The ratio of per cent diameter change to per cent length change should have a value of about -0.30 if there were no volume changes or errors in measurement. The values for this ratio in Table III are in most cases greatly different from the value of -0.30, again indicating swelling and roughening as contributing factors. The theoretical value is only approached for two slugs with History 2 and four slugs with History 3.

Density changes do not seem to have been affected by the preirradiation history of the slugs for the most part. In general, they tend to correspond to those changes that occurred in unalloyed uranium slugs with similar irradiation histories. It is evident, however, that several of the uranium-2 w/o zirconium slugs with the higher burnup (0.18 a/o) and irradiated at the higher calculated temperature ( $383^{\circ}\text{C}$ ) have begun to swell (see Figures 6 and 7). Similar data for unalloyed uranium(10) did not indicate swelling within the temperature range  $300\text{-}390^{\circ}\text{C}$  and with burnups up to 0.3 a/o. The suspicion exists that the calculated temperatures may be different from the actual temperatures by substantial amounts. However, temperatures of  $430\text{-}450^{\circ}\text{C}$  were subsequently measured in a fuel rod of uranium-2 w/o zirconium in the second core of EBR-I, in a location next to the center rod and slightly above the midplane. The calculated temperature in this area for these experimental fuel rods was  $415^{\circ}\text{C}$ . Although the actual temperatures may not be known, some swelling has occurred in those slugs in the regions of higher temperature in the reactor core. These data show general agreement with some recently published information.(11)

There is evidence that the ratio of volume increase to burnup is smallest for the "as quenched" specimens (History 1). These were also the hardest specimens and presumably the strongest. Hence, they were better able to resist the internal stresses arising from the accumulation of fission fragments.

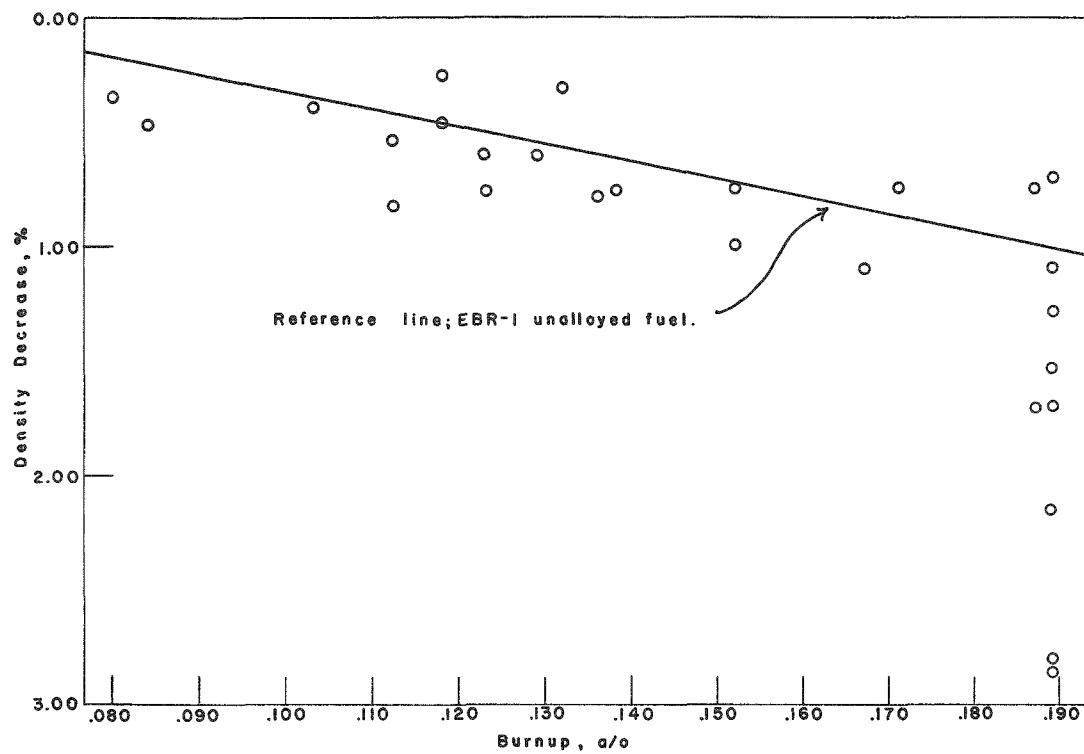


Figure 6. Density Decrease vs. Burnup for Experimental Uranium-2 w/o Zirconium Fuel Slugs Irradiated in the First Core of EBR-I

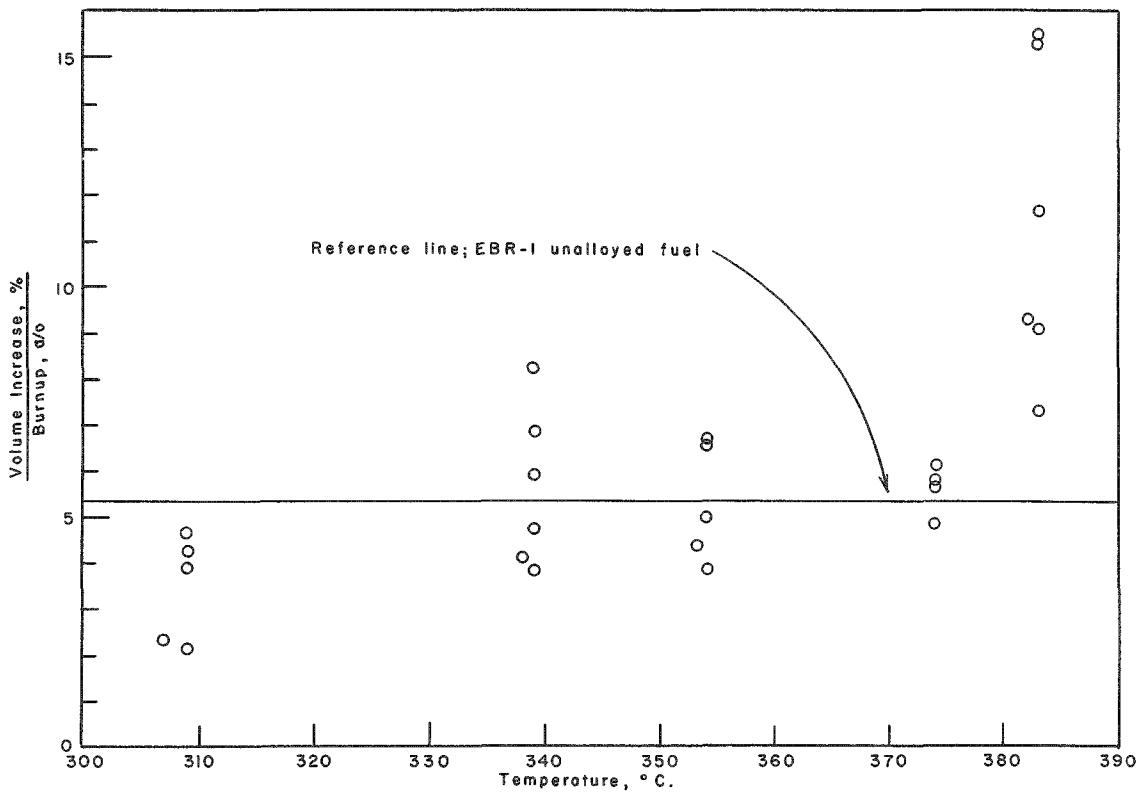


Figure 7. Volume Increase/Burnup vs. Temperature for Experimental Uranium-2 w/o Zirconium Fuel Slugs Irradiated in the First Core of EBR-I

The range of hardness for all the irradiated specimens was small (64 to 75 Rockwell A). Although the slugs with History 1 seemed slightly harder on the average, the other five groups seemed to have similar hardnesses. A plot of the hardnesses of the irradiated slugs against temperature (see Figure 8) shows that the hardness decreases with increasing irradiation temperature in the range 310-390°C.

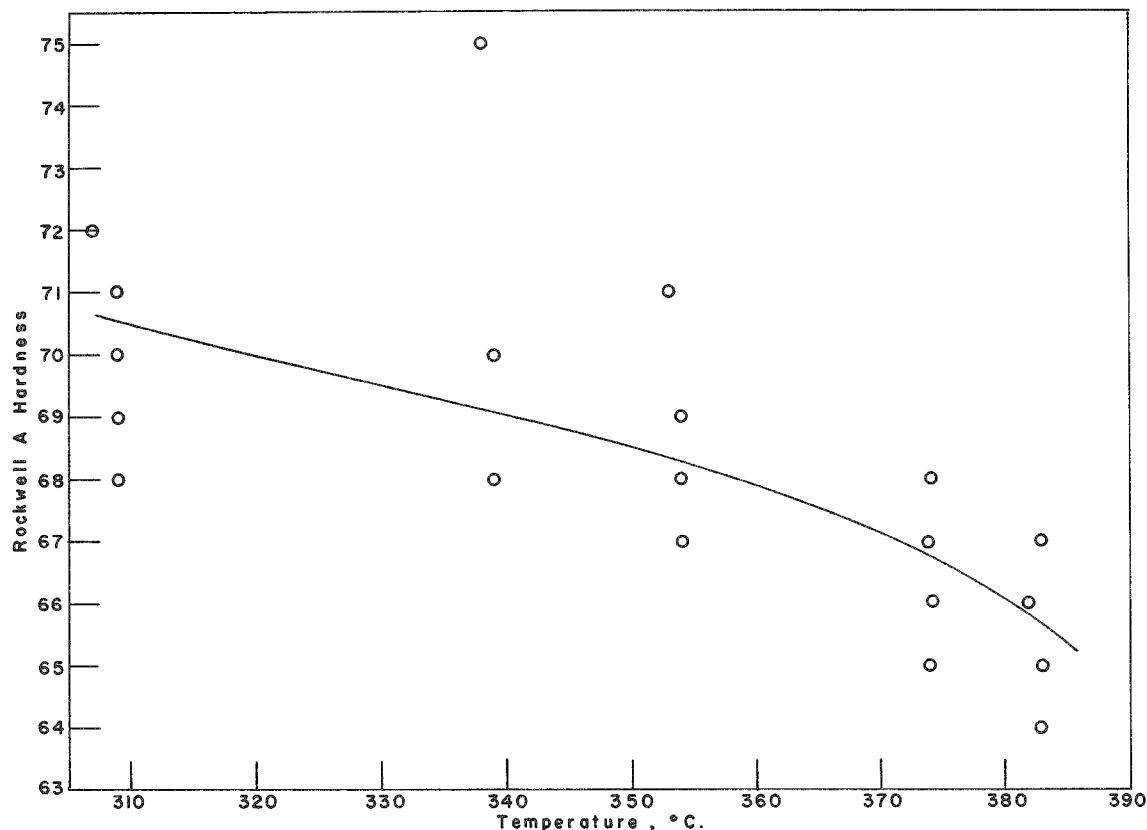


Figure 8. Hardness vs. Temperature for Experimental Uranium-2 w/o Zirconium Fuel Slugs Irradiated in the First Core of EBR-I

The hardnesses of uranium-2 w/o zirconium alloy after various treatments similar to those given the irradiated slugs have been reported(5) and are given in Table IV. The water-quenched samples had the highest hardness. Conversion of the DPH hardness values to Rockwell A, although not entirely valid, does seem to indicate that irradiation has not had an appreciable effect on the hardness of the alloy fuel slugs.

## CONCLUSIONS

1. Cast uranium-2 w/o zirconium fuel slugs showed the least tendency to change in length (average +0.12%) when irradiated. Of the four groups of wrought slugs, the "as quenched" changed length the least (average +0.48%). The three groups of wrought specimens in which some annealing occurred had about the same tendency to elongate (average +2.08%).

2. The ratio of length change to burnup was smallest for the chill-cast slugs (average +0.8 microin./in.-ppm burnup) and was +1.4 microin./in.-ppm burnup for the chill-cast and annealed slugs. Of the wrought slugs, the average ratio for the "as quenched" slugs was +2.2 microin./in.-ppm burnup. The other wrought slugs had an average ratio of about +13 microin./in.-ppm burnup.

3. Five slugs (of 0.385-in. diameter) which were slow cooled from 800°C provided data which indicated that the ratio of length change to burnup decreases with increasing temperatures in the range from 309 to 374°C. Excluding burnup as a factor, the colder the slug in this group of specimens the greater the length increase.

4. Slugs which were farther removed from the condition of phase equilibrium because of their preirradiation treatment had the greater dimensional stability when irradiated.

5. Density measurements indicated slight decreases in density with increasing burnup and were comparable to unalloyed uranium below 0.180 a/o burnup. Specimens with burnups of 0.187-0.189 a/o showed a tendency to swell. Swelling occurred in the specimens at the hottest calculated temperatures, 380-385°C. Unalloyed uranium with up to 0.3 a/o burnup did not swell at these temperatures in EBR-I.

6. Hardness does not seem to have been very much affected by irradiation. A tendency for the hardness to drop slightly with increasing irradiation temperature is noted.

## ACKNOWLEDGMENTS

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## REFERENCES

1. Untermeyer, S., Reduction of Irradiation Damage to Uranium by Small Zirconium Additions, ANL-4604 (March 1951).
2. Paine, S. H., F. L. Brown, and W. F. Murphy, Preliminary Studies of Irradiation Damage to Uranium-Zirconium Alloys, ANL-5537 (Feb 1956).
3. Paine, S. H., and F. L. Brown, "Pin Cushion" Irradiation Tests of Uranium and Its Zirconium Alloys, ANL-5538 (July 1958).
4. Kittel, J. H., and S. H. Paine, Effects of High Burnup at Elevated Temperatures on Uranium-0.52 and 1.62 w/o Zirconium Alloys, ANL-5406 (Aug 1959).
5. Zegler, S. T., H. H. Chiswik, M. H. Mueller, and R. E. Macherey, The Dimensional Stability of Uranium-Zirconium Alloys upon Thermal Cycling, ANL-5540 (to be published).
6. Shuck, A. B., The Manufacture of 16 Uranium-Zirconium Alloy Test Fuel Rods for EBR, ANL-5185 (to be published).
7. Levenson, M., Determination of the Conversion Ratio of the Experimental Breeder Reactor by Radiochemical Methods, ANL-5095 (December 1953).
8. Curtis, C. D., S. H. Klein, D. Okrent, W. C. Redman, and S. Untermeyer, A Physical Determination of the Conversion Ratio of the Experimental Breeder Reactor, ANL-5222 (August 1954).
9. Lichtenberger H. V., Experimental Breeder Project Report, December 1, 1948 through February 28, 1949, ANL-4274.
10. Murphy, W. F., A. C. Klank, and S. H. Paine, Examination of Uranium from the First Core of EBR-I, ANL-6113 (Sept 1961).
11. Bentle, G. G., Irradiation Swelling of Uranium and Uranium Alloys, NAA-SR-4893 (Sept 1961).

## APPENDIX

## MEASUREMENT DATA ON URANIUM-2 w/o ZIRCONIUM FUEL SLUGS

## WEIGHTS

Weights of the slugs were determined to 0.1 mg both before and after irradiation. The preirradiation weights were measured on the electropolished specimens just prior to loading them into the fuel rods. Each irradiated slug was weighed "as received," and after scrubbing and rinsing in acetone. For the purpose of reporting, the weights were rounded off to the nearest milligram. Pre- and postirradiation weights and weight differences are given in Table V.

Table V  
WEIGHTS OF SLUGS

Fuel Rod Designation	Slug Position in Rod (Fig 3)	Weight gm			Weight Changes, mg		
		Original	Postirradiation				
			As Received	Scrubbed	B-A	C-B	C-A
S-1-62-545-1	2	117 648	117 661	117 644	+13	-17	-4
	3	118 234	118 237	118 230	+3	-7	-4
S-1-62-545-2	2	117 065	117 082	117 056	+17	-26	-9
	3	117 375	117 383	117 371	+10	-12	2
S-1-62-545-3	2	116 321	116 321	116 294	+6	-33	-27
	3	115 867	115 868	115 848	+1	-20	-19
S-1-62-545-4	2	117 482	117 502	117 437	+20	-65	-45
	3	118 160	118 172	118 153	+12	-19	-7
S-1-62-545-5	2	116 918	116 959	116 891	+41	68	-27
	3	117 199	117 210	117 081	+11	-129	-118
S-1-62-545-6	2	114 588	114 611	114 575	+23	-36	-13
	3	116 849	116 855	116 844	+6	-11	-5
S-2-62-545-7	2	114 892	114 895	114 891	+3	-4	-1
	3	115 319	115 317	115 314	-2	-3	-5
	4	115 181	115 184	115 180	+3	-4	-1
S-2-62-545-8	2	114 927	114 934	114 924	+7	-10	3
	3	113 785	113 794	113 777	+9	-17	-8
	4	112 992	112 997	112 989	+5	-8	-3
S-2-62-545-9	2	115 167	115 176	115 158	+9	-18	-9
	3	115 333	115 371	115 359	+38	-12	+26
	4	114 957	114 973	114 959	+16	-14	+2
S-2-62-545-10	2	115 030	115 040	115 032	+10	-8	+2
	3	113 654	113 666	113 651	+12	-15	-3
	4	113 455	113 460	113 450	+5	-10	-5
S-2-62-560-1	2	115 470	115 476	115 472	+6	-4	+2
	3	115 497	115 502	115 500	+5	-2	+3
	4	115 230	115 233	115 230	+3	-3	0

## LENGTHS AND DIAMETERS

The lengths and diameters of the slugs were measured prior to irradiation by means of a comparator and gauge blocks. The results were recorded to 0.0001 in. The diameters were given as maxima and minima.

Postirradiation measurements were made with micrometers mounted on blocks of aluminum. The micrometers were checked with standards. A ratchet on the adjusting knurl helped prevent overtightening. The readings were estimated to 0.0001 in. on the vernier.

At least three length measurements were taken on each slug, and the position of the slug with respect to the micrometer was reversed for each measurement. A minimum of four diameter measurements were taken along the length of each specimen after irradiation. The maximum and minimum diameters for each slug prior to irradiation were averaged and compared with the average of the postirradiation measurements. All measurements of both length and diameter were rounded off to the nearest 0.001 in. for use in this report. The average values of the postirradiation length and diameter measurements are compared with the preirradiation measurements in Table VI.

Table VI  
LENGTH AND DIAMETER MEASUREMENTS

Fuel Rod Designation	Slug Position in Rod (Fig. 3)	Length, in		Length Change		Diameter, in		Diameter Change	
		Preirradiation	Postirradiation	mils	%	Preirradiation	Postirradiation	mils	%
S-1-62-545-1	2	3 752	3 789	+37	+0.99	0.368	0.370	+2	+0.54
	3	3 751	3 847	+96	+2.55	0.368	0.370	+2	+0.54
S-1-62-545-2	2	3 751	3 848	+97	+2.59	0.366	0.366	0	0.00
	3	3 749	3 829	+80	+2.12	0.367	0.369	+2	+0.55
S-1-62-545-3	2	3 750	3 750	0	0.00	0.366	0.367	+1	+0.27
	3	3 732	3 738	+6	+0.16	0.365	0.369	+4	+1.09
S-1-62-545-4	2	3 751	3 800	+49	+1.31	0.367	0.370	+3	+0.82
	3	3 751	3 862	+111	+2.96	0.368	0.368	0	0.00
S-1-62-545-5	2	3 751	3 846	+95	+2.53	0.366	0.365	-1	-0.27
	3	3 751	3 828	+77	+2.05	0.366	0.369	+3	+0.82
S-1-62-545-6	2	3 751	3 762	+11	+0.30	0.366	0.372	+6	+1.64
	3	3 769	3 791	+22	+0.59	0.366	0.372	+6	+1.64
S-2-62-545-7	2	3 334	3 334	0	0.00	0.385	0.390	+5	+1.30
	3	3 333	3 421	+88	+2.65	0.386	0.383	-3	-0.78
	4	3 333	3 372	+39	+1.18	0.386	0.386	0	0.00
S-2-62-545-8	2	3 333	3 378	+45	+1.35	0.385	0.385	0	0.00
	3	3 334	3 333	-1	-0.03	0.384	0.387	+3	+0.78
	4	3 332	3 335	+3	+0.09	0.384	0.385	+1	+0.26
S-2-62-545-9	2	3 335	3 322	-13	-0.39	0.385	0.389	+4	+1.04
	3	3 334	3 369	+35	+1.05	0.386	0.385	-1	-0.26
	4	3 335	3 378	+43	+1.29	0.385	0.389	+4	+1.04
S-2-62-545-10	2	3 332	3 402	+70	+2.09	0.385	0.385	0	0.00
	3	3 333	3 329	-4	-0.12	0.384	0.387	+3	+0.78
	4	3 332	3 332	0	0.00	0.384	0.387	+3	+0.78
S-2-62-560-1	2	3 345	3 446	+101	+3.01	0.386	0.382	-4	-1.04
	3	3 325	3 411	+86	+2.58	0.387	0.385	-2	-0.52
	4	3 329	3 371	+42	+1.26	0.386	0.385	-1	-0.26

### DENSITIES

Densities before and after irradiation were determined by the method based on loss of weight on immersion in carbon tetrachloride. An immersion time of 5 min was allowed for temperature equalization of the

irradiated slugs. The results were adjusted to 20°C as a reference temperature. Comparison between the two sets of data is made in Table VII.

Table VII  
DENSITY, HARDNESS, AND RADIOACTIVITY

Fuel Rod Designation	Slug Position in Rod (Fig. 3)	Density gm/cm <sup>3</sup>		Density Decrease		Postirradiation Rockwell A Hardness	Radiation Reading, mr/hr
		Preirradiation	Postirradiation	gm/cm <sup>3</sup>	%		
S-1-62-545-1	2	18 050	17 921	0 129	0 71	70 0 <sup>+0 5</sup> 1 0	1400
	3	18 112	17 604	0 508	2 80	64 0 <sup>+2 0</sup> -2 0	1400
S-1-62-545-2	2	18 104	17 872	0 232	1 28	69 5 <sup>+0 5</sup> -1 0	1500
	3	18 109	17 717	0 392	2 16	64 5 <sup>+2 0</sup> -1 0	1550
S-1-62-545-3	2	18 025	17 929	0 096	0 53	69 5 <sup>+1 5</sup> -1 0	425
	3	18 049	17 901	0 148	0 82	67 0 <sup>+0 5</sup> -0 5	450
S-1-62-545-4	2	18 039	17 903	0 136	0 75	75 0 <sup>+1 5</sup> 2 0	1400
	3	18 111	17 801	0 310	1 71	66 0 <sup>+1 0</sup> -1 0	1400
S-1-62-545-5	2	18 108	17 908	0 200	1 10	68 0 <sup>+1 0</sup> -2 0	1300
	3	18 103	17 796	0 307	1 70	64 0 <sup>+1 0</sup> -1 0	1350
S-1-62-545-6	2	17 750	17 479	0 271	1 53	67 5 <sup>+1 5</sup> -2 5	1300
	3	18 028	17 514	0 514	2 85	65 0 <sup>+3 5</sup> -3 0	1400
S-2-62-545-7	2	18 054	17 998	0 056	0 31	72 0 <sup>+0 0</sup> -0 0	750
	3	18 077	17 943	0 134	0 74	71 0 <sup>+1 0</sup> -2 0	1000
	4	18 079	17 941	0 138	0 76	67 0 <sup>+0 5</sup> -1 0	800
S-2-62-545-8	2	18 078	18 016	0 062	0 34	68 0 <sup>+1 0</sup> -2 0	300
	3	18 062	17 991	0 071	0 39	69 0 <sup>+2 0</sup> -1 0	375
	4	17 948	17 864	0 084	0 47	65 5 <sup>+1 0</sup> -1 5	300
S-2-62-545-9	2	18 071	18 026	0 045	0 25	69 0 <sup>+2 0</sup> -1 0	1000
	3	18 036	17 855	0 181	1 00	66 5 <sup>+1 5</sup> -0 5	1200
	4	18 063	17 926	0 137	0 76	65 0 <sup>+0 5</sup> -1 0	950
S-2-62-545-10	2	18 072	17 964	0 108	0 60	69 5 <sup>+1 5</sup> -1 5	800
	3	18 076	17 876	0 200	1 10	69 0 <sup>+1 0</sup> -0 5	1200
	4	18 014	17 874	0 140	0 78	67 5 <sup>+1 5</sup> -1 0	950
S-2-62-560-1	2	18 103	18 020	0 083	0 46	70 5 <sup>+0 5</sup> -1 0	1000
	3	18 095	17 959	0 136	0 75	67 5 <sup>+1 0</sup> -1 5	1250
	4	18 097	17 990	0 107	0 59	66 5 <sup>+1 0</sup> -1 5	1000

## HARDNESSES

Rockwell A hardness measurements were made on the irradiated slugs. No preirradiation hardnesses had been determined. At least four hardness measurements were made along the length of each slug. The average hardness and the variations in hardness are given in Table VII.

## RADIATION

The radiation from a 1-in. section of each slug near its center of length was measured through a port hole in the hot cell wall. The parts of the slug not exposed were shielded with 4 in. of lead. The distance between the slug and the radiation meter was 105 cm. An alpha, beta, gamma meter was used with all shields open. The readings obtained are given in Table VII.

## PHOTOGRAPHS

Photographs at 1X magnification were taken on the fuel slugs from one rod of each loading scheme. In general the slugs selected were representative of the other slugs in this group of specimens. The photographs are presented as Figure 5 (a) and (b).

## PROFILES

Profile traces were taken on several slugs after irradiation. A 1-in. section near the center of length was selected. The length of the trace was magnified four times on the recorder and the vertical displacement of the stylus was magnified approximately 300X. Typical profiles are shown in Figures 9 and 10.

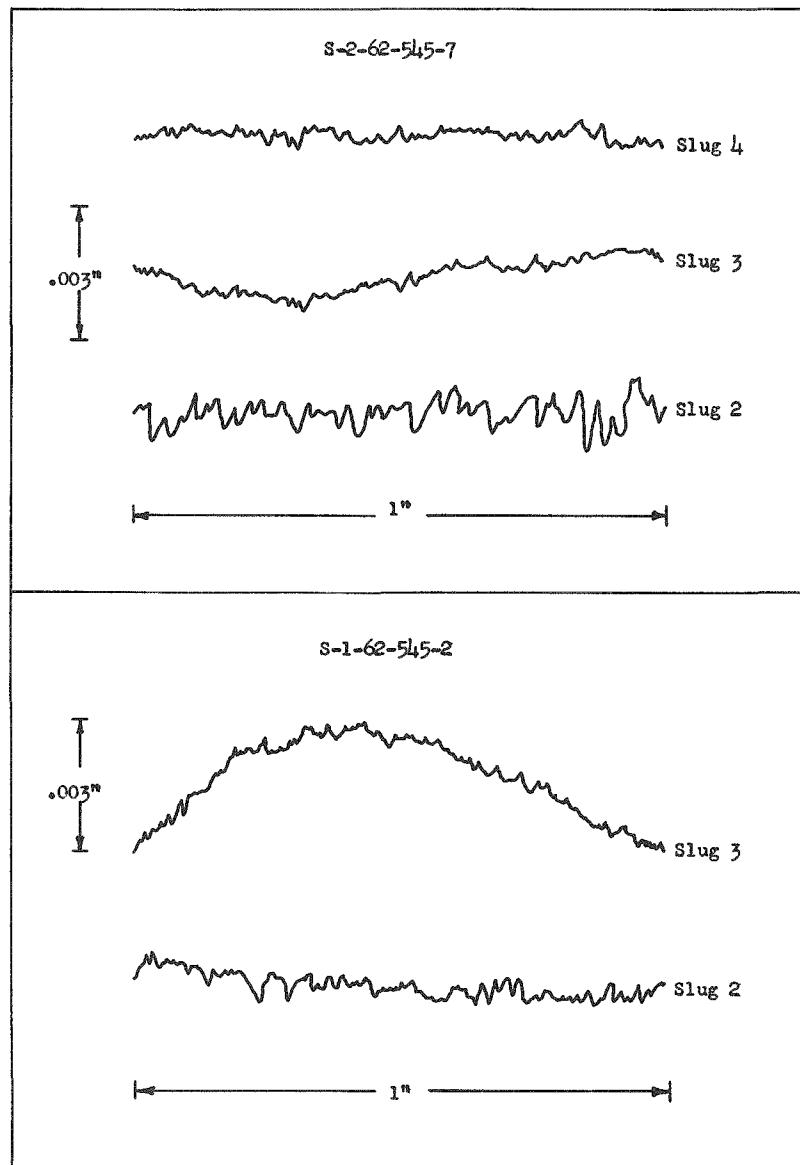


Figure 9. Profile Traces on Irradiated Slugs

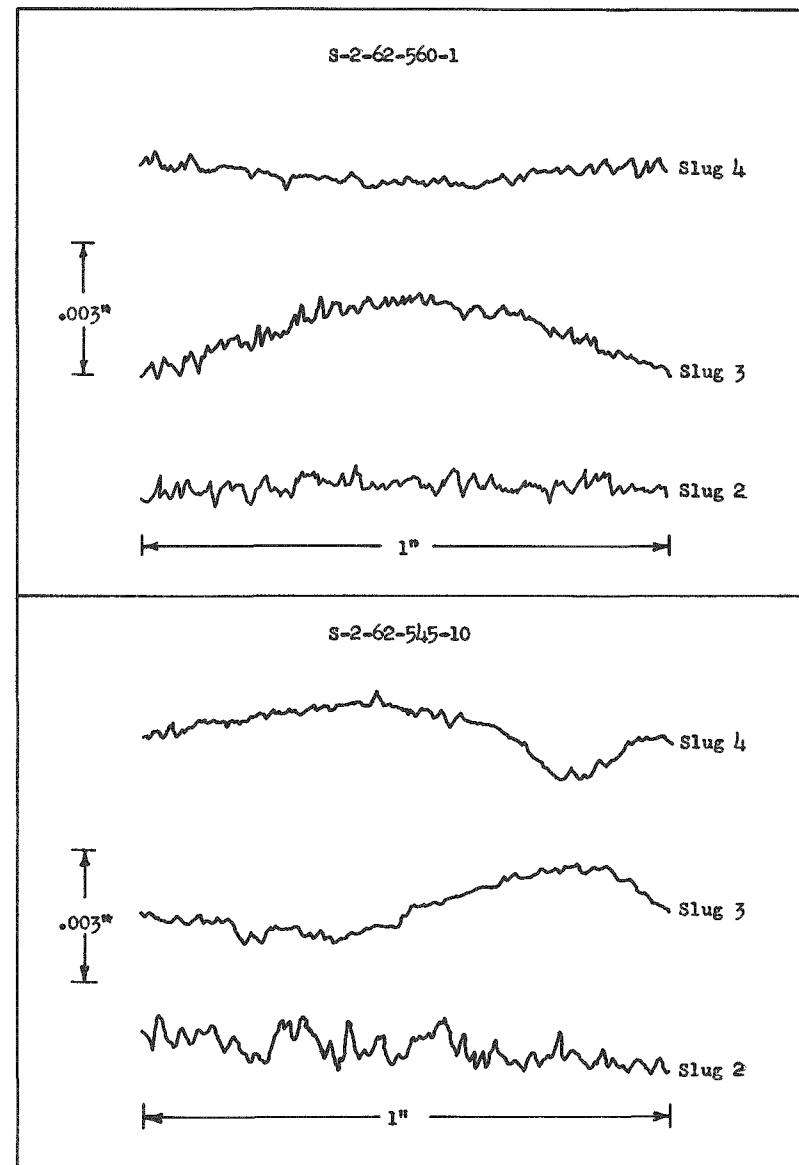


Figure 10. Profile Traces on Irradiated Slugs