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IRRADIATION BEHAVIOR OF Zr-U DRIVER TUBES FOR HWCTR

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DP-994, IRRADIATION BEHAVIOR OF
Zr-U DRIVER TUBES FOR HWCTR
by C. L. Angerman and G. R. Caskey, Jr.

This report summarizes the postirradiation measurement and metallographic examination of the HWCTR driver tubes, performed by the Reactor Engineering and Nuclear Materials Divisions. The driver tube performance was better than had been anticipated, under relatively severe irradiation conditions.

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Nuclear Materials Division

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ABSTRACT

The HWCTR was fueled with enriched uranium tubes of Zr - 9.3 wt % U alloy clad with Zircaloy-2 by coextrusion. Irradiation of these tubes up to 1.83 atom % burnup at temperatures up to 540°C and under 1200 psi coolant pressure resulted in volume increases up to 5%, only about half of the amount predicted by published data. The volume increases were caused by the formation of solid fission products and the precipitation of fission gases in small bubbles.

CONTENTS

LIST OF TABLES AND FIGURES

IRRADIATION BEHAVIOR OF Zr-U DRIVER TUBES FOR HWCTR

INTRODUCTION

The Heavy Water Components Test Reactor (HWCTR) was designed for the irradiation testing of full-size prototype fuel assemblies of uranium metal or uranium oxide for potential use in heavy-water-moderated and -cooled power reactors.[1] The reactor itself was fueled with 2.3-inch-diameter enriched uranium "driver" tubes of Zr - 9.3 wt % U clad with Zircaloy by coextrusion.[2] These tubes normally operated at central temperatures of 500-540°C and 1000-1200 psi system pressure to total atom burnups of 1.5-2.0%. Published data on the irradiation behavior of small samples of Zr alloys containing 6 to 22 wt % U indicated that volume increases as high as 10% might be anticipated under these conditions.[3,4]

Since only a limited amount of swelling could be tolerated and there were no data available on the performance of large fuel elements of this type, four of the driver tubes were measured at intervals during irradiation to monitor their swelling behavior. A detailed postirradiation examination was conducted to determine the cause of swelling, and postirradiation annealing experiments were performed to characterize the swelling that might occur at higher irradiation temperatures.

SUMMARY

Irradiation of the driver tubes up to 1.83 atom % burnup at time-averaged central metal temperatures up to 540°C and under 1200 psi coolant pressure resulted in volume increases up to 5.1%, which was less swelling than anticipated. This swelling occurred as an increase in wall thickness and was caused principally by the formation of solid fission products, with a lesser contribution due to precipitation of fission gases in small bubbles. The reactor pressurization, as well as impurities in the cores, may have limited the fission gas swelling. Much larger volume increases occurred during postirradiation annealing for 74 hours at 600°C under low external restraint.

The irradiation had no visible effect on the Zircaloy-2 cladding. Negligible corrosion of the cladding by the heavy water coolant occurred, as indicated by the absence of hydride precipitates in the cladding. The circumferential strain in the cladding due to the swelling of the core was less than 0.3%.

DISCUSSION

FABRICATION

The driver tubes, 2.30-inch OD by 1.96-inch ID clad with 0.015 inch of Zircaloy-2, were fabricated by coextrusion by Nuclear Metals, Inc.[2] Vacuum-induction-melted alloy of Zr - 9.3 wt % U was cast into graphite molds and machined to form cores for the coextrusion billet. The carbon content of the core was less than 2500 ppm. Billets were heated to 690°C and extruded. After extrusion the tubes were straightened, etched, and autoclaved 4 hours at 340°C in water and 4 hours at 400°C (1500 psi) steam.

At the extrusion temperature, the alloy is a two-phase mixture of alpha (α) Zr and gamma (γ) solid solution of Zr with 13 atom % U (Figure 1). Since the tubes were not heat treated after extrusion, except for autoclaving, the microstructure before irradiation was a finely divided lamellar array of epsilon (ϵ) phase particles within a matrix of α-Zr (Figure 2). The ϵ forms from γ by a eutectoid decomposition.

IRRADIATION

In the HWCTR, the driver tubes were arranged in a ring with positions for testing of experimental fuel elements within this ring. The operation of the reactor was divided into two different cycles; new driver tubes were charged at the start of the second cycle. The second cycle was operated at higher nominal temperatures than the first. In the first cycle, the nominal operating conditions for the driver tubes were 540°C maximum core temperature (510°C time-averaged), 1200 psi system pressure, and 1.83 atom % calculated maximum burnup; during the second cycle of operation, the conditions were 585°C maximum core temperature (540°C time-averaged), 1200 psi system pressure, and 1.54 atom % calculated maximum burnup. Surface temperatures of the Zircaloy-2 cladding were as high as 306°C in the second cycle. Temperature profiles along the lengths of the four tubes that were inspected are shown in Figure 3.

SWELLING DURING IRRADIATION

The two tubes that operated at the highest temperatures were chosen from each cycle for interim inspection during the course of irradiation. Measurements of outside and inside diameters, made periodically in a disassembly basin at several locations along the lengths of the tubes, indicated that up to 3.6% swelling had occurred. The final dimensional and volume changes (changes in cross-sectional area) and exposure profiles for the tubes are shown in Figures 4 through 7.

The pertinent data from the interim inspections, corrected for systematic errors, are summarized in Table I. Some corrections were needed, because volume decreases of 1 to 3% were actually indicated at the ends of the tubes where the exposures and temperatures were low. Furthermore, the volume increases calculated from measurements of the outside diameter and wall thickness on individual ring sections cut from the tubes for subsequent metallographic examination were several percentage points greater than the increases calculated from the measurements made in the disassembly basin. Since these data indicate that a systematic error was present in the original measurements, all the volume data were adjusted to extrapolate to zero at zero burnup for subsequent discussions. These adjusted values are shown in parentheses in Table I.

TABLE I

Irradiation Conditions and Behavior of Driver Tubes

		Irradiation Conditions			Max. Dimensional Changes(c)				
Cycle No.	Tube No.	Calc. Burnup, atom %	TACMT(a), °C	Max CMT(b), °C	ΔOD, mils	Cladding Strain, %	ΔID, mils	ΔV/V, %	R(d), %/atom %
1	48	1.47	486	536	4	0.18	-10	3.6 (5.1)	3.4
		1.80	489	532	2	0.10	-11	3.2 (4.7)	2.6
1	22	1.18	501	531	3	0.13	- 6	2.5 (4.0)	3.4
		1.57	502	530	5	0.22	- 8	3.2 (4.7)	3.0
		1.83	508	528	5	0.17	- 8	3.4 (4.9)	2.7
2	1	0.32	533	539	-1	0	- 2	0.22	0.7
		0.76	534	569	2	0.09	- 7	2.75 (2.3)	3.0
		1.33	522	565	-1	0	- 6	1.16 (3.2)	2.4
		1.50	530	526	1 (5)*	0.02 (0.17)*	- 4 (-14)*	0.8 (4.3)*	2.9
2	18	0.32	523	539	0	0.15	- 4	1.09	3.4
		0.79	543	585	0	0.28	- 7	2.04 (1.5)	1.9
		1.38	534	572	1	0.08	- 5	1.96 (4.3)	3.1
		1.54	538	509	1 (7)*	0.03 (0.27)*	- 2 (- 9)*	0.56 (4.1)*	2.7

(a) Time-averaged central metal temperature.

(b) Maximum central metal temperature during irradiation.

(c) Where two sets of values are given, the values in parentheses have been adjusted so that the change in volume extrapolates to zero at zero exposure; the others are raw data obtained from measurements made on full-length tubes in the basin.

(d) Swelling rate - percent increase in volume per atom percent burnup.

* The data marked with an asterisk are based on direct measurements made on ring sections in a hot cell.

Inspection of the data on changes in outside and inside diameters indicated that the volume change was caused by an increase in wall thickness. The inside diameter decreased more than the outside diameter increased. Correspondingly, circumferential tensile strain in the outer Zircaloy-2 cladding was low, less than 0.3%. The average diameter decreased about 0.2% and the length increased about 0.5%, indicating some slight anisotropic growth. Hexagonal metals, such as zirconium, will grow when irradiated with fission fragments.[5]

The behavior of these Zr-U alloy tubes was distinctly better than anticipated. Available swelling data from the literature, derived from irradiation tests on small samples, predict volume increases of up to 10%, or about twice the observed swelling for the temperatures and exposures that prevailed during the irradiation.[3,4] The behavior of the HWCTR driver tubes is compared to the published data in Figure 8. The improved behavior was attributed to the smaller contribution of fission gas swelling in the drivers than in the small samples. Reactor pressurization, as well as impurities in the cores, may have restricted the agglomeration of the fission gases.

In some cases the HWCTR driver tubes stuck in their housing tubes after irradiation; however the tubes were easily removed with a small force. Such sticking would be expected, since a volume increase of about 4% would eliminate the clearance between the driver and housing; as shown in Table I, volume increases of as much as 5.1% were observed.

MICROSTRUCTURAL CHANGES DURING IRRADIATION

Specimens were cut for metallographic examination from the hottest position (maximum swelling) in each tube; specimens were also cut from near the ends of Tubes 22 and 48. The principal microstructural features were:

- Fission gas bubbles were observed in specimens irradiated at 490 to 540°C, but none were observed in the specimens irradiated at 400°C.
- Fission gas bubbles were often more prevalent along the α-ϵ interface than within the α phase.
- The ϵ phase particles were slightly distorted and spheroidized as compared to those in the as-extruded tube where the pattern was more regular and lamellar.
- No cracks were observed in either the core or cladding.

- No hydride or twins were seen in the Zircaloy cladding of the specimens from either high- or low-temperature regions of the tube.

Fission gas bubbles up to 0.4 micron in diameter were present in specimens irradiated at 490 to 540°C (Figure 9), but none were present in the specimens irradiated at 400°C. Volume increases due to the gas bubbles, calculated from measurements of bubble diameters and concentrations, ranged up to 2.1%, which when added to the increase due to solid fission products, 2.5% per atom % burnup, essentially accounted for the observed volume increases. Bubbles were sometimes located along the α-ϵ interface as well as throughout the α-zirconium. No bubbles could be positively identified within the ϵ phase in spite of its much higher uranium concentration.

Some distortion and spheroidization of the ϵ phase were evident, although these were not as pronounced as in U - 2 wt % Zr alloys.[6] The difference between the two cases is probably due to the much greater irradiation growth in the case of the α-uranium matrix in the U - 2 wt % Zr alloy. The α-zirconium has a much lower growth rate and hence would distort less and not break up the ϵ phase pattern generated by extrusion. Distortion was more pronounced in the higher temperature portion of the tube in keeping with the greater exposure and growth in that region (Figure 10).

Cladding corrosion during reactor operation was negligible as judged by the absence of hydride in the cladding. No hydride was seen in any of the sections. Absence of twins in the cladding was in keeping with the very low cladding strains that were measured (Figure 11).

EFFECT OF POSTIRRADIATION ANNEALING

Operation of the drivers at maximum core temperatures of 600°C would generate a higher flux for irradiation of the test fuel in the HWCTR. In order to obtain an indication of the maximum swelling that might occur during irradiation at a temperature within the $\alpha + \gamma$ region, a specimen from Tube 22 (irradiated at 500°C) was annealed at 600 ±4°C for 2 hours, examined metallographically, annealed an additional 72 hours and re-examined. In contrast to irradiation, only low external restraint (due to the cladding) was present during the postirradiation anneals. The increases in volume of the specimen during annealing, based on pre- and post-annealing density measurements, was 10.4% after 2 hours of annealing and a total of 21.0% after 74 hours of annealing. Increases in the sizes and concentrations of gas bubbles accounted for this swelling, as shown in Figure 12. The volume increase after 74 hours is about twice as much as observed by Johnston[7], but the burnup was somewhat higher, 1.8 compared to 1.15 atom %.

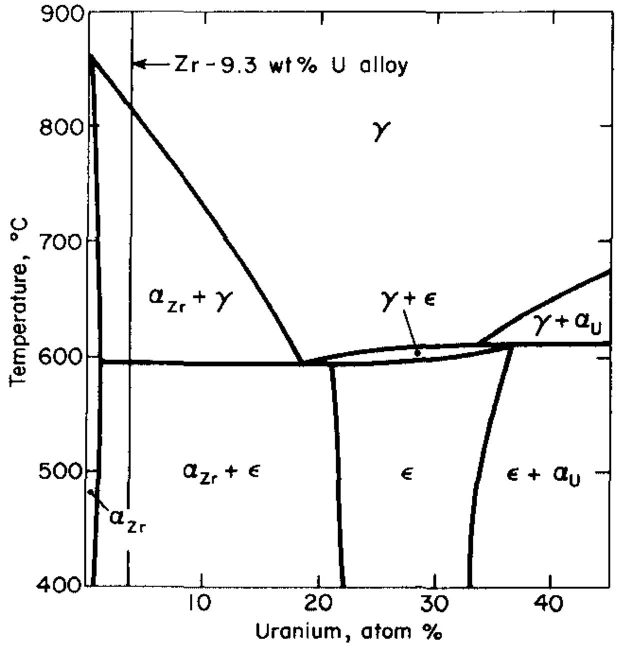


FIG. 1 ZIRCONIUM-URANIUM PHASE DIAGRAM

NEG. 58608 500X

a. Optical micrography showing distorted pattern of finely divided α and ϵ phases.

NEG. 1756H 6000X

b. Electron micrograph showing both lamellar and massive ϵ particles.

FIG. 2 MICROSTRUCTURE OF AS-EXTRUDED Zr - 9.3 wt % U ALLOY

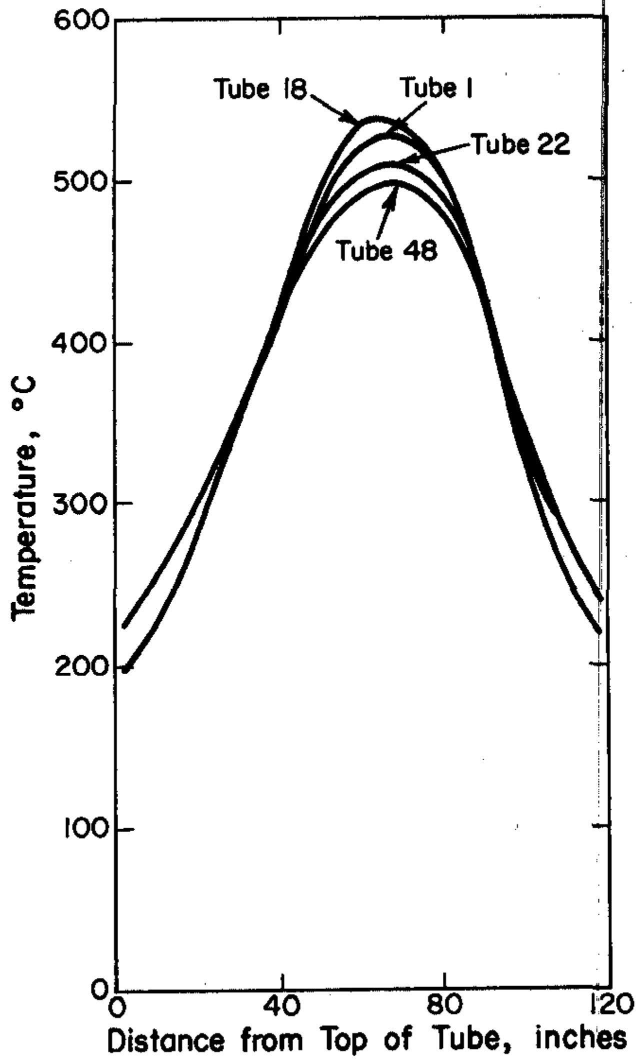


FIG. 3 PROFILES OF TIME-AVERAGED CENTRAL METAL TEMPERATURE

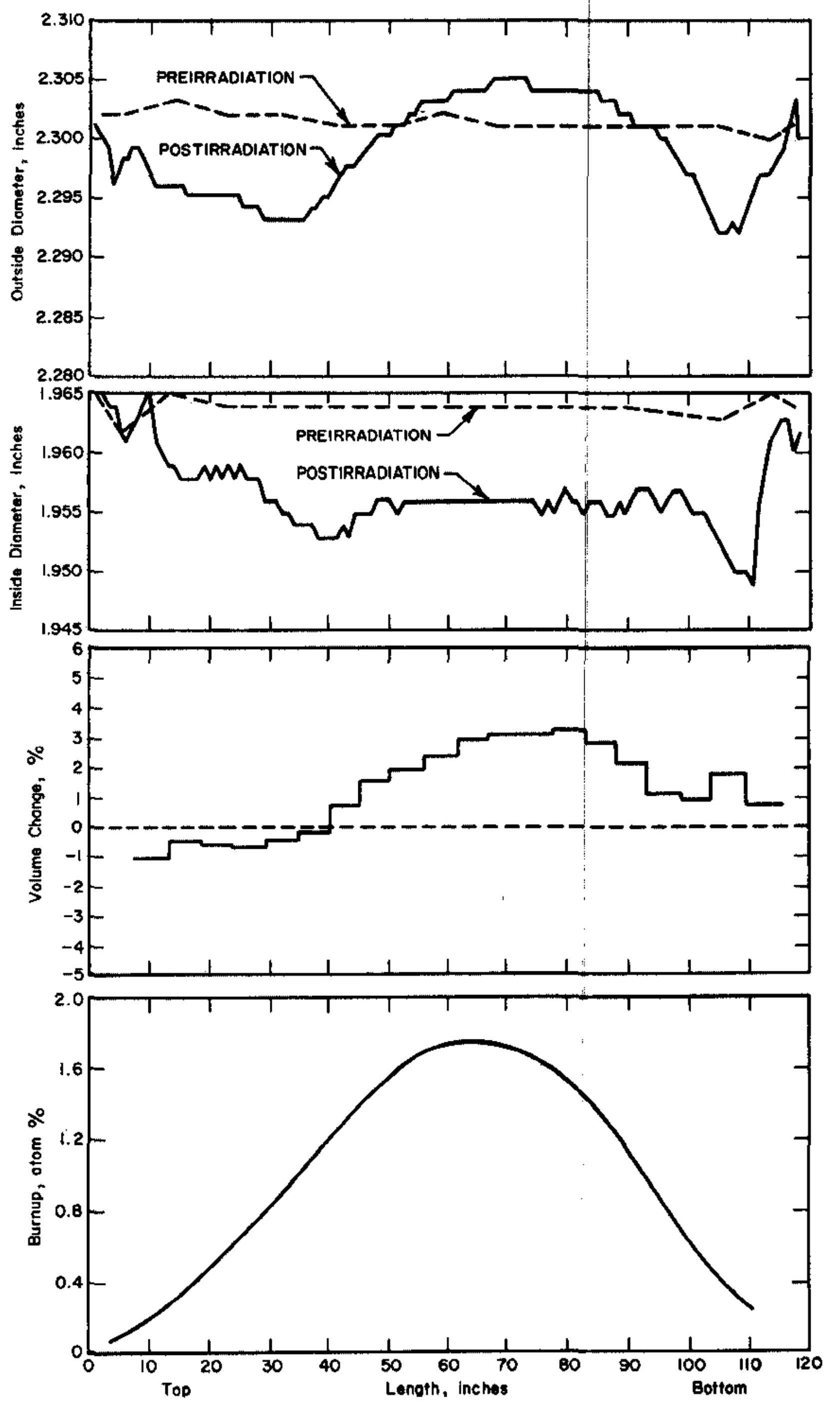


FIG. 4 DIMENSIONAL CHANGE AND EXPOSURE PROFILES, TUBE 22

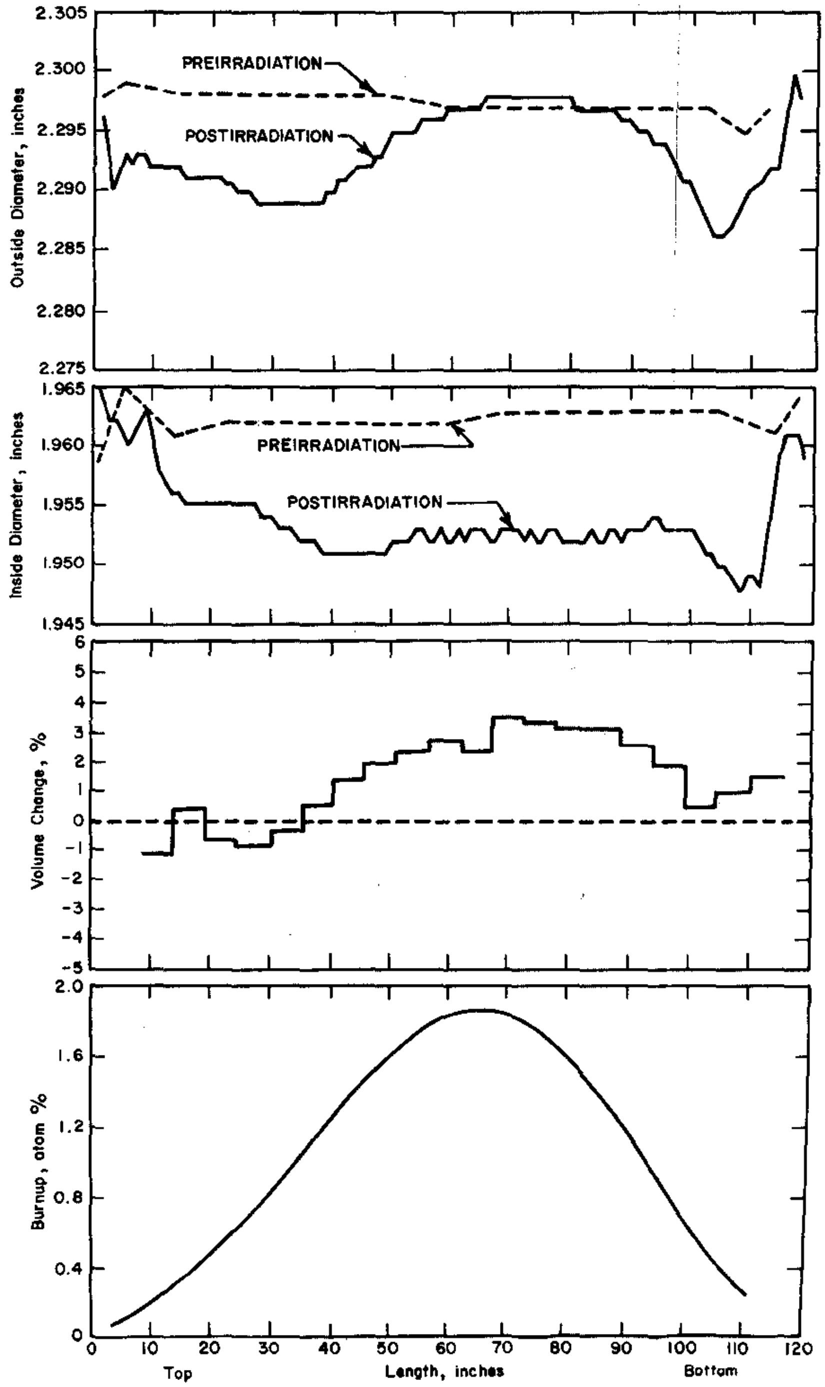


FIG. 5 DIMENSIONAL CHANGE AND EXPOSURE PROFILES, TUBE 48

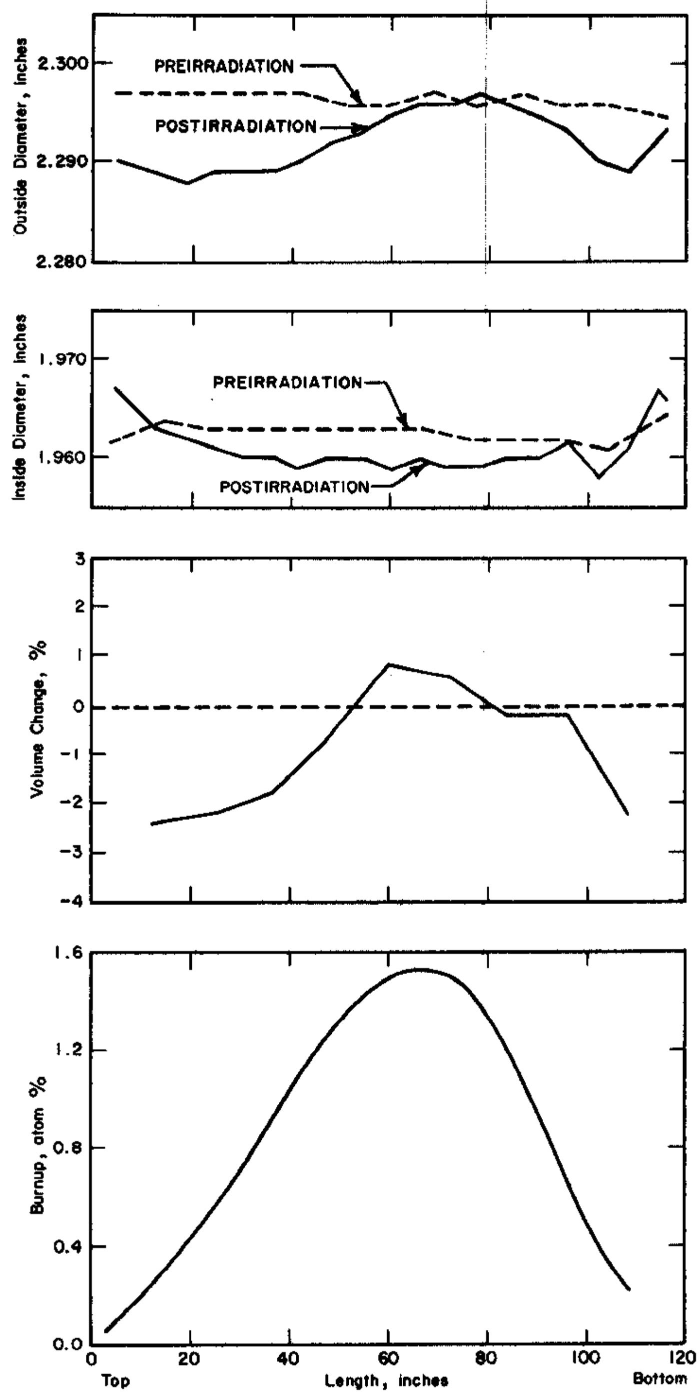


FIG. 6 DIMENSIONAL CHANGE AND EXPOSURE PROFILES, TUBE 1

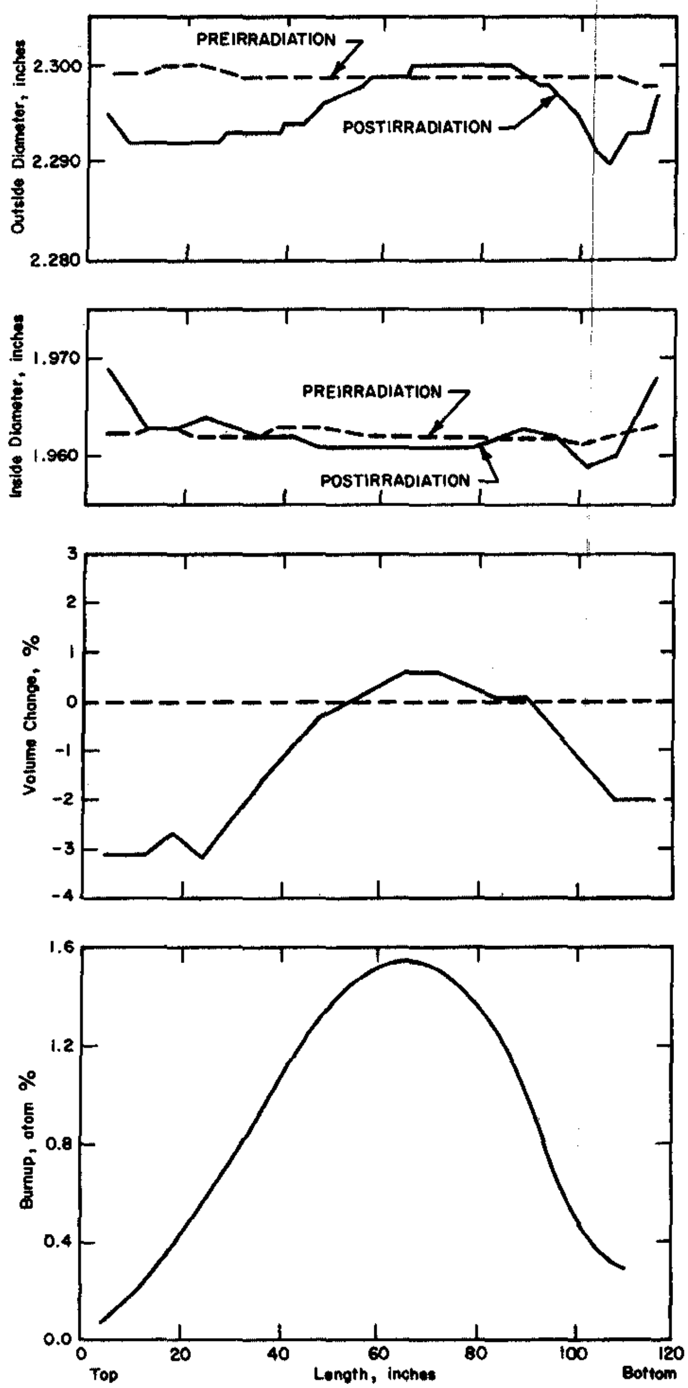
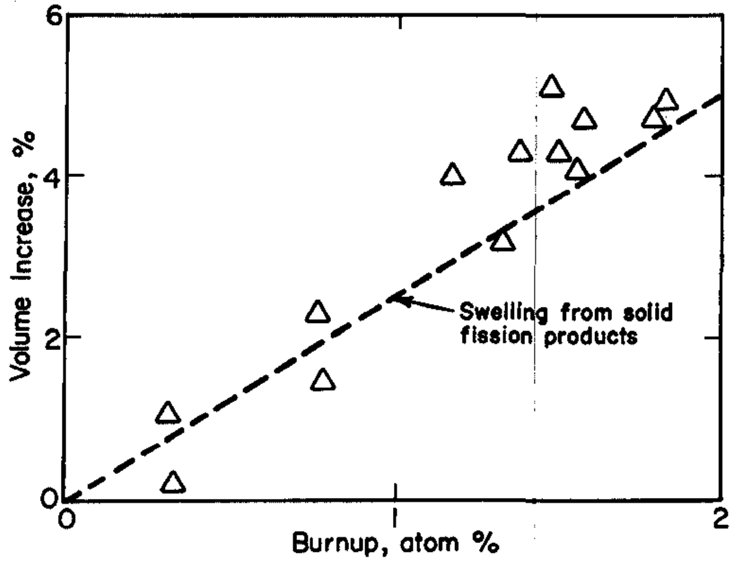
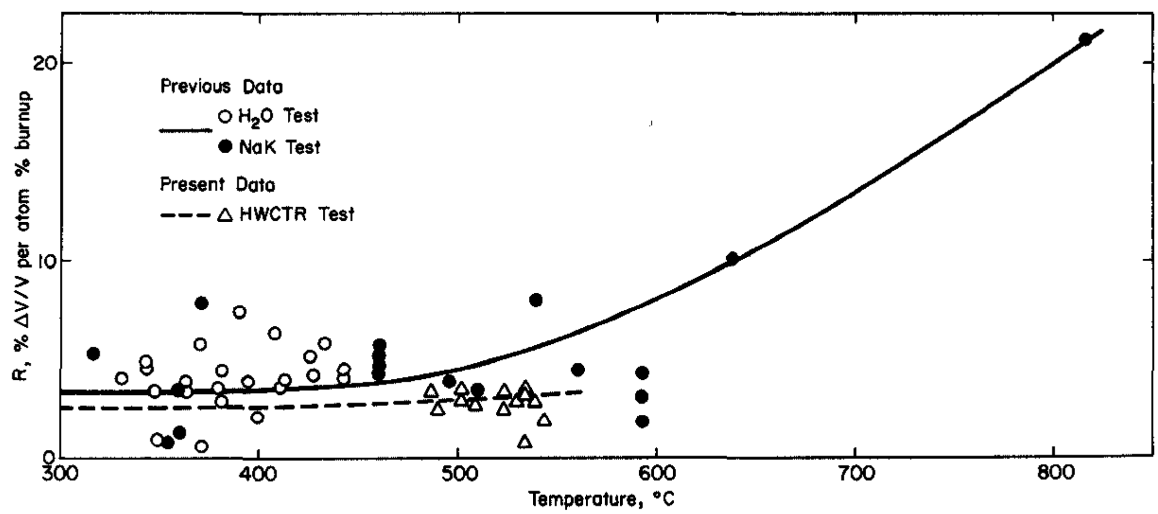


FIG. 7 DIMENSIONAL CHANGE AND EXPOSURE PROFILES, TUBE 18

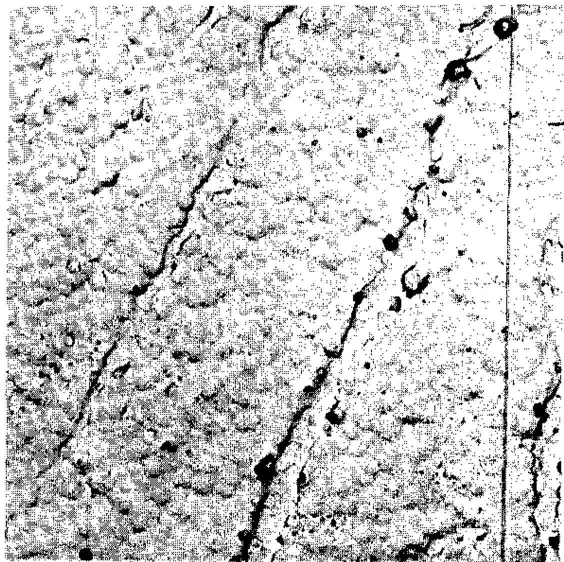


a. Variation in swelling with burnup for HWCTR tubes. Data may be represented by a linear relationship, although some departure may be indicated at the higher burnups and temperatures.



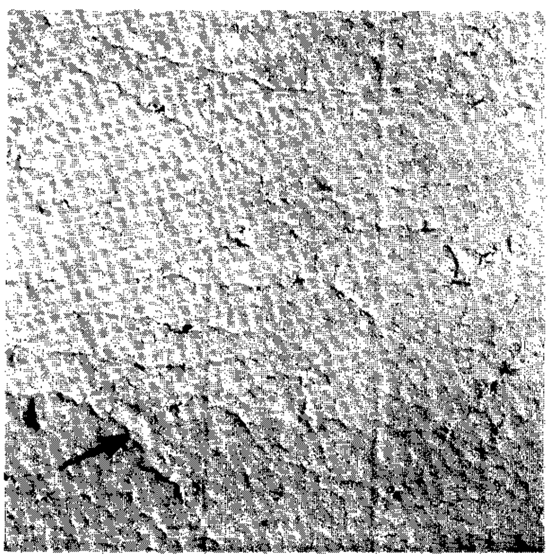
b. Variation of normalized swelling, R, with temperature.

FIG. 8 COMPARISON OF SWELLING BEHAVIOR OF HWCTR DRIVER TUBES WITH PREVIOUSLY PUBLISHED IRRADIATION DATA[3,4]

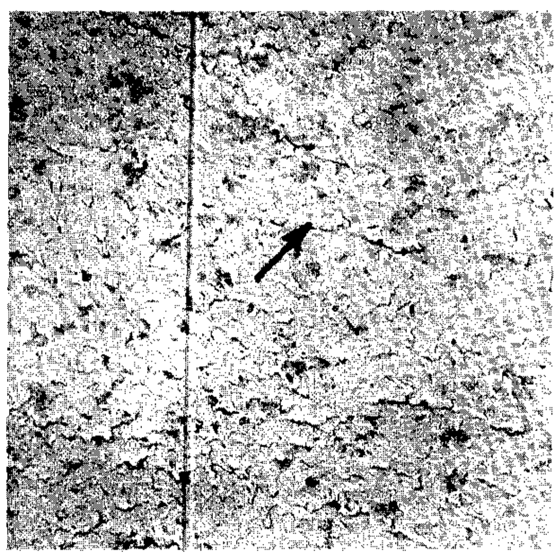


Fissión gas bubbles (A) along interface between α and ϵ phases and (B) in the α-zirconium matrix. Exposure = 1.89 atom % burnup, temperature = 500°C.

FIG. 9 FISSION GAS BUBBLES FORMED DURING IRRADIATION

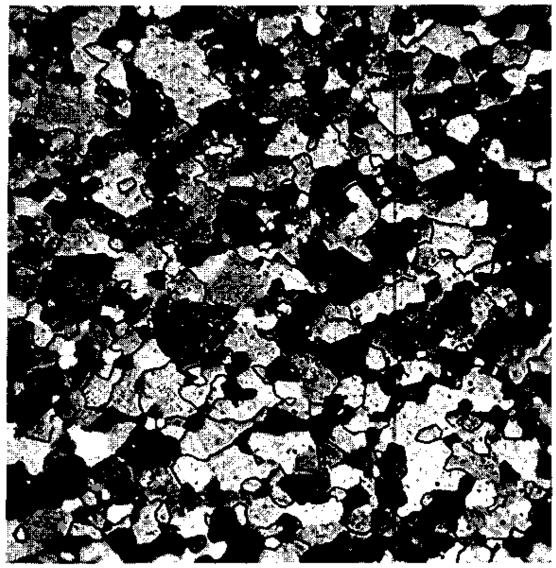


a. Region of alloy irradiated at 400°C to 1.10 atom % burnup. Pattern of ϵ phase altered very little from that of as-extruded alloy (compare Figure 2b).



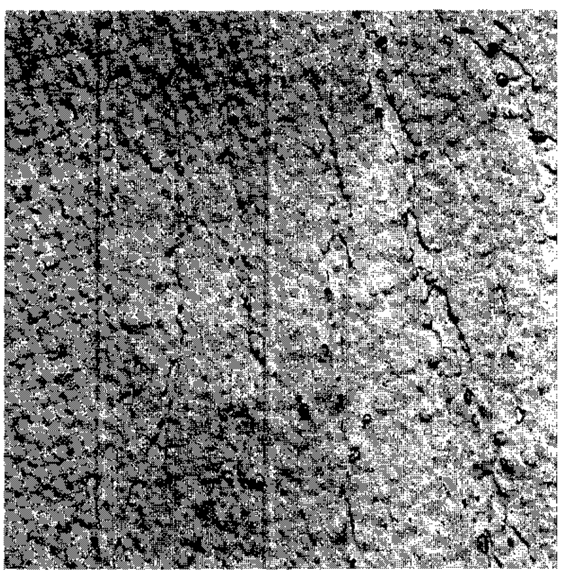
b. Region of alloy irradiated at 485°C to 1.80 atom % burnup showing distorted pattern of ϵ phase.

FIG. 10 DISTORTION OF ϵ PHASE DURING IRRADIATION



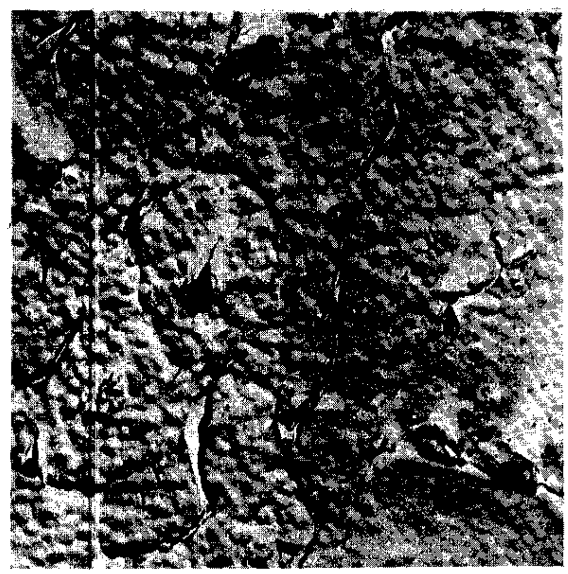
NEG. 62925 500X

FIG. 11 TYPICAL MICROSTRUCTURE OF IRRADIATED ZIRCALOY-2 CLADDING
No twins or zirconium hydride particles



6000X

As irradiated at 500°C



6000X

Annealed 74 hr at 600°C

FIG. 12 GAS BUBBLES PRODUCED BY POSTIRRADIATION ANNEALING (74 hr at 600°C)

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