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PRELIMINARY EXPERIMENTS ON IRRADIATION CYCLING AND PARTIAL BETA-PHASE IRRADIATION OF URANIUM

by

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PRELIMINARY EXPERIMENTS ON IRRADIATION CYCLING AND PARTIAL BETA PHASE IRRADIATION OF URANIUM

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ABSTRACT

A group of uranium specimens were irradiated under conditions in which some of the specimens were irradiated continuously while others were cycled in and out of a reactor. Although most of the specimens were irradiated under conditions in which they were entirely in the alpha phase, some were irradiated so that their centers were above alpha-phase temperatures. Both highly textured material (300°C rolled) and nominally randomly oriented material (300°C rolled and beta-quenched) were studied. It appeared that irradiation cycling of both types of material may result in greater elongation than would be anticipated on the assumption that the effects of irradiation and thermal cycling acting alone were additive. The material rolled at 300°C showed no external effects due to central irradiation temperatures being above those limiting the alpha phase. However, beta-quenched material, which was irradiated so that central temperatures were above those required for stability of the alpha phase, developed severe distortions which were greater under cycling conditions. It was also noted that 300°C rolled uranium begins to elongate under irradiation at burnups as low as 0.0002 a/o (2 MWD/T).

INTRODUCTION

The first core in the EBR-I fast reactor was made of unalloyed uranium which had been prepared by rolling at 300°C and subsequently quenching from the beta phase. Although at the time of preparation this procedure was the best known stabilizing heat treatment for uranium, it was realized that the fuel slugs would not be completely dimensionally stable during the life of the core. Earlier experiments at Argonne National Laboratory had showed that beta-quenched uranium would gradually elongate under thermal cycling[1] and under irradiation.[2] Since both of these effects were expected to occur during normal use of the reactor, the fuel element contained free space at the top into which the fuel slugs could elongate.

However, as significant amounts of fuel burnup were achieved in the core, reactivity losses gave increasing evidence that the uranium slugs were elongating at a greater rate than had been anticipated. Among the proposed explanations for this apparent behavior was the suggestion that concurrent irradiation and thermal cycling might, as far as dimensional stability was concerned, be somewhat more damaging than the effects of irradiation and thermal cycling alone would indicate, if it were assumed they were additive.

Irradiations of a limited number of uranium specimens were made early in 1955 in the MTR in order to test this hypothesis. The irradiation conditions were varied so that some specimens were irradiated continuously, whereas others were cycled into and out of the reactor. Also, by varying the specimen diameters, the irradiations were made under conditions where some of the specimens were in a temperature range such that they were entirely in the alpha phase, whereas other specimens were irradiated so that their centers were above alpha phase temperatures.

EXPERIMENTAL PROCEDURE

The primary purpose of the experiments was to compare the effects of continuous irradiation on uranium specimens with the effects produced by intermittent or cycling irradiation. Because of the ease with which capsules could be rapidly inserted and withdrawn from the MTR reflector, using hydraulic rabbits, it was decided to use these facilities for all irradiations, including those exposures to be made without cycling. Before irradiations were begun, surveys of the thermal neutron fluxes were made in the hydraulic rabbits, using monitor foils composed of 99 w/o Al-0.5 w/o Mn-0.5 w/o Co alloy. The induced activity in the cobalt was counted in order to determine the thermal flux. The flux profile obtained in rabbit VH-2 was found to be satisfactory and is shown in Figure 1. It was in this facility that all specimen irradiations were performed.

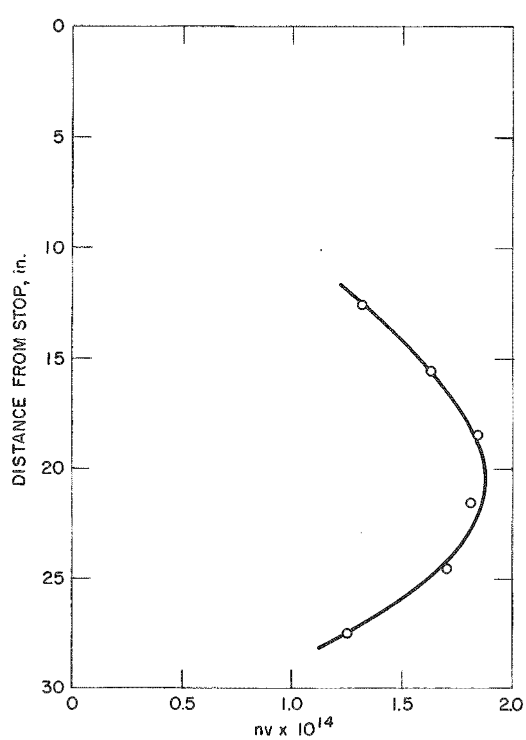


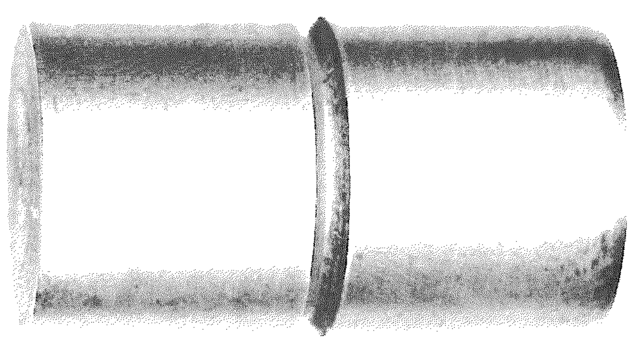
Figure 1. Thermal flux survey in MTR hydraulic rabbit VH-2.

It was desired to irradiate some of the specimens with the temperatures of their centers above the region of stability of the alpha phase to determine the possible consequences to be expected if the fuel elements in

EBR-I were to be operated in such a manner. From the flux values shown in Figure 1 it was possible to calculate the critical diameter of a natural uranium specimen whose center would be at 668°C, the temperature at which alpha uranium transforms to the beta phase. However, for such a calculation to be meaningful, accurate knowledge of such factors as flux depression in and adjacent to the sample, end heat-conduction losses, thermal conductivities as functions of temperature, heat transfer coefficients, etc., would be required. Depending upon the values chosen for these factors, it was calculated that the critical diameter for a center temperature of 668°C might range from approximately 0.50 to over 0.55 inch. Similarly, the calculated surface temperature of the specimen ranged from 480° to 540°C.

Because of these uncertainties it was decided to determine experimentally the critical diameter by irradiating a preliminary group of specimens with diameters ranging both well above and below the computed diameter at which beta-phase operations was expected to occur. These initial specimens were made of material rolled at 300°C, so that they had a fine-grained microstructure. When uranium rolled at 300°C is heated even briefly into the beta or gamma phases, considerable grain coarsening results. By transversely fracturing fine-grained irradiated specimens with progressively larger diameters and examining the fracture surfaces, it was expected that the critical diameter could be determined, since a coarse-grained area would be evident if the center of any of the specimens had reached beta or gamma phase temperatures during irradiation. Examination of the fractured surfaces, however, was not expected to indicate conclusively whether the centers of the specimens had been heated above the beta phase into the gamma phase.

As shown in Figure 2, a circumferential notch was machined on these and all other specimens used in subsequent irradiations to facilitate a clean transverse break through the sample. To further increase the brittleness of the specimens, they were chilled in liquid nitrogen before fracturing.



24855 3X

Figure 2

Typical specimen before irradiation. The specimen has been circumferentially notched to facilitate transverse fracturing.

NaK capsules of the type shown in Figure 3 were used for containment of the specimens during irradiation. Each irradiation capsule contained an aluminum-cobalt alloy flux monitor, and atom burnups in the specimens were calculated from the flux values thus obtained. All the specimens were measured for dimensional changes resulting from irradiation. Each specimen was then fractured and the surfaces thus exposed were examined to determine if the center of the specimen had remained in the alpha phase during irradiation.

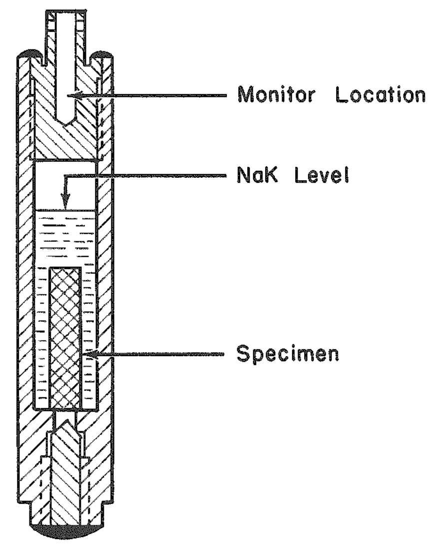


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Figure 3

Typical assembled irradiation capsule.

RESULTS AND DISCUSSION

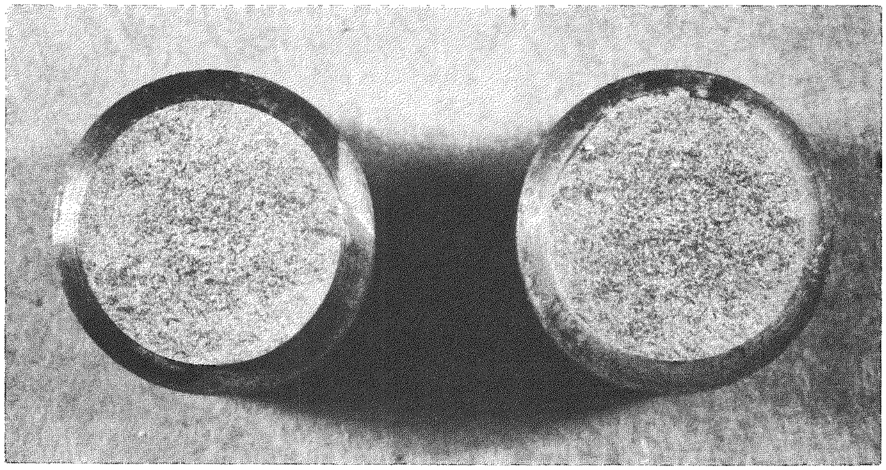
Table I shows the observations on the initial group of fine-grained specimens which were irradiated to determine the critical diameter for center-line beta-phase formation. Each specimen was irradiated for one hour. The appearance of the fracture surfaces from the 0.56-in. diameter

TABLE I

Effects of Irradiation on Initial Group of 300°C Rolled Specimens Used to Determine the Critical Diameter for Beta-Phase Operation

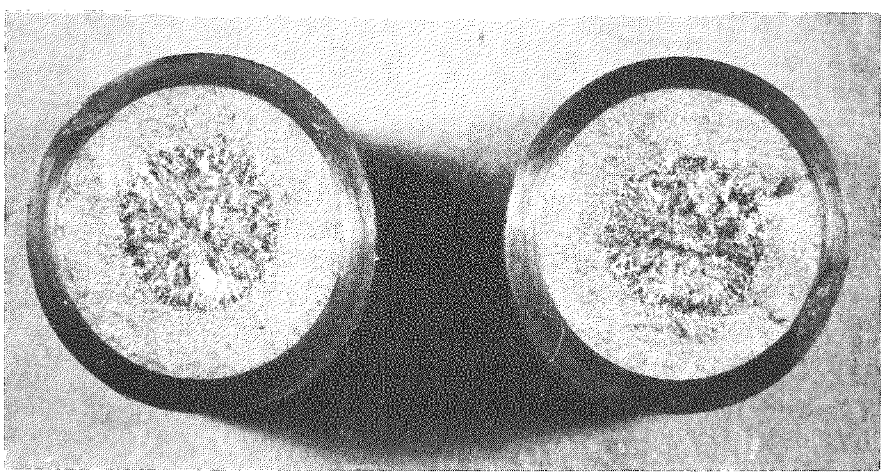
Spec. No.	Diameter, in.	Burnup, a/o	Length Change, %	Appearance of Fracture
AC-2	0.28	0.00024	0.10	Fine-Grained
AC-5	0.32	0.00024	0.27	Fine-Grained
AC-9	0.36	0.00023	0.30	Fine-Grained
AC-13	0.40	0.00017	0.05	Fine-Grained
AC-17	0.44	0.000068	0.05	Fine-Grained
AC-19	0.48	0.00025	0.65	Fine-Grained
AC-21	0.52	0.00026	0.43	Fine-Grained
AC-23	0.56	0.00027	0.55	Fine-Grained
AC-25	0.60	0.00027	0.29	Coarse-Grained Center

specimen, No. AC-23, which apparently remained wholly in the alpha phase, is shown in Figure 4. Figure 5 shows the fracture surfaces of the next larger specimen, No. AC-25, with a diameter of 0.60 in. A coarse-grained center is clearly evident, indicating that the center of this specimen was at least in the beta phase during irradiation.



17434 3X

Figure 4. Fracture surfaces of an irradiated 0.56-in. diameter 300°C rolled specimen, No. AC-23. The uniformly fine-grained structure indicates that the entire specimen remained in the alpha phase during irradiation.



17432 3X

Figure 5. Fracture surfaces of an irradiated 0.60-in. diameter 300°C rolled specimen, No. AC-25. The coarse-grained area indicates that the center of the specimen was in the beta phase during irradiation.

In addition to providing information on the diameter above which beta-phase formation would occur, this initial group of specimens was also of interest in determining the effect of very low burnups on highly textured uranium. An earlier investigation(2) had included determinations of irradiation growth rates in uranium rolled at 300°C with burnups ranging down to 0.0057 a/o. As shown in reference 2 the irradiation growth rate of fuel specimens may be expressed as a growth coefficient, G_i. The 300°C rolled specimens described in reference 2 showed an average value for G_i of 693 micro-in./in. per fission/10^6 total atoms. Although the present specimens received at most only one-twentieth of the minimum burnup of the specimens described in reference 2, it is apparent from the length changes reported in Table I that irradiation growth had already begun. After subtracting the growth in each specimen that would be expected from the thermal cycle it received, an average value for G_i of 820 micro-in./in. per fission/10^6 total atoms is obtained. It appears, therefore, that irradiation growth in highly textured uranium begins at burnups as low as 0.0002 a/o (2 MWD/T).

Following the irradiations on the initial group of specimens described above, two 0.52-in. diameter specimens were irradiated under cycling conditions. With this diameter the specimens remained entirely in the alpha phase during irradiation. One of the specimens, No. AC-22 had been rolled at 300°C. The other specimen, No. AC-61 had been rolled at 300°C and beta-quenched so that it was identical in preparation with the EBR-I fuel. The specimens were irradiated by inserting them into the reactor, using the VH-2 rabbit, for a 15-minute irradiation period, followed by removal from the reactor for a 5-minute cooling period in the canal, for a total of 50 cycles. Transfer times were estimated to be approximately five seconds. The changes in these specimens, which are designated as Group 1, are shown in Table II.

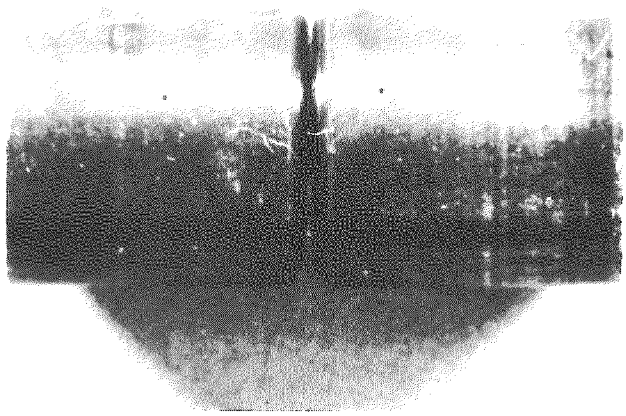
TABLE II

Effect of 0.040 a/o Burnup under Conditions of Both Irradiation Cycling and Continuous Irradiation

Spec. No.	Condition	Diameter, in.	Type of Irradiation	Irradiation Temp. Range	Length Change, %	Diameter Change, %	Remarks
				Group 1			
AC-22	300°C rolled	0.52	50 cycles(a)	alpha	5.0	-2.7	
AC-61	300°C rolled, beta-quenched	0.52	50 cycles	alpha	0.8	-0.1	
				Group 2			
AC-26	300°C rolled	0.60	50 cycles	alpha + beta	3.8	-1.7	
AC-65	300°C rolled, beta-quenched	0.60	50 cycles	alpha + beta	-	-	Developed central cavity and swelled so that specimen could not be removed from capsule.
				Group 3			
AC-62	300°C rolled, beta-quenched	0.52	12.5 hr continuously	alpha	0.2	-0.1	
AC-64	300°C rolled, beta-quenched	0.56	12.5 hr continuously	alpha	0.3	0.0	
AC-66	300°C rolled, beta-quenched	0.60	12.5 hr continuously	alpha + beta	0.9	-0.1	Developed a central projection at one end.

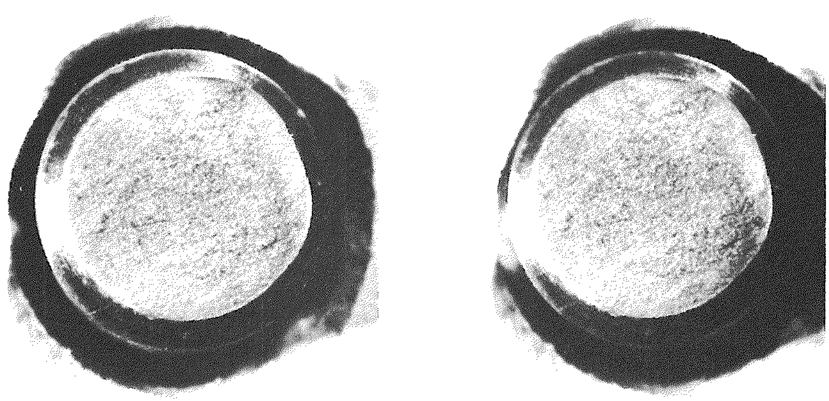
(a)A cycle consisted of a 15-minute irradiation followed by a 5-minute cooling period. Transfer times are estimated to be approximately 5 seconds.

The more highly textured specimen, No. AC-22, showed the greater elongation, as would be expected. Photographs of the irradiated specimens, before and after they were fractured, are shown in Figures 6 through 9.



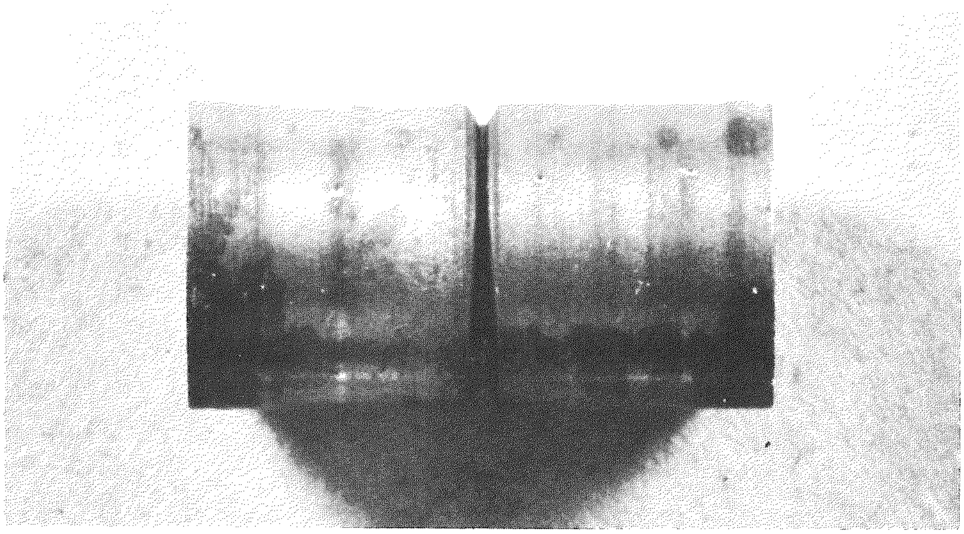
18620 3X

Figure 6. Appearance of an irradiation-cycled 300°C rolled specimen, No. AC-22, which remained in the alpha phase during irradiation.



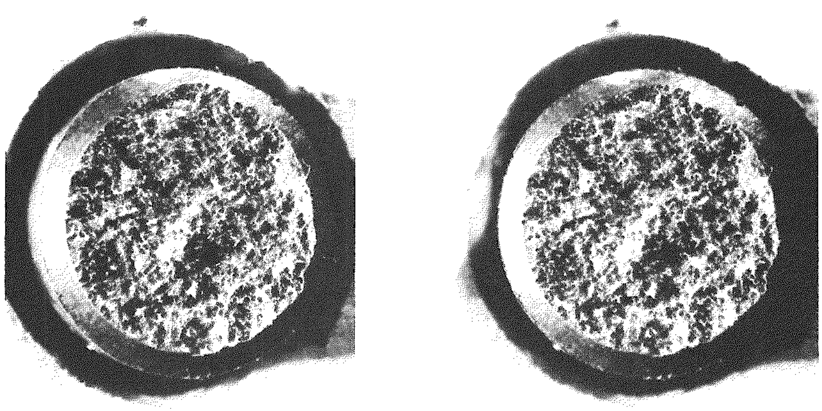
18832 3X

Figure 7. Fracture surfaces of the above specimen.



18622 3X

Figure 8. Appearance of an irradiation-cycled 300°C rolled and beta-quenched specimen, No. AC-61, which remained in the alpha phase during irradiation.



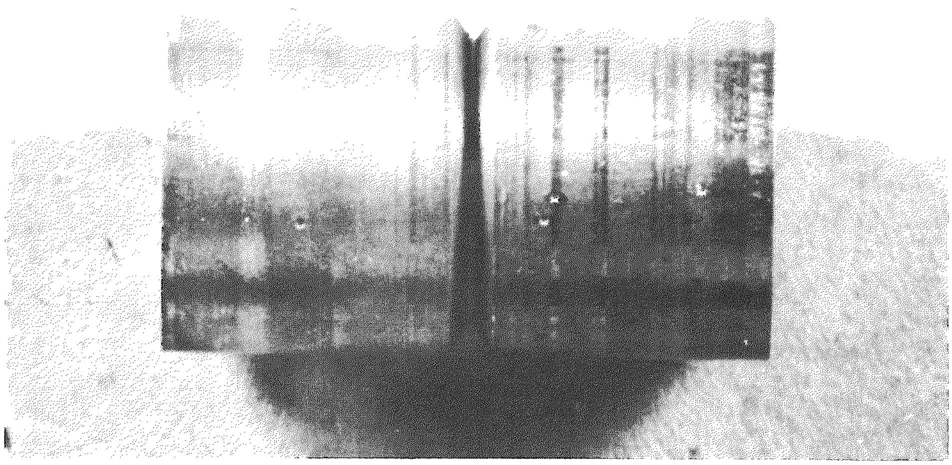
18839 3X

Figure 9. Fracture surfaces of the above specimen.

A second group of specimens, prepared in the same manner as the above Group 1 but with 0.60-in. diameters for partial beta-phase operation, were then irradiated. This second group was also given 50 irradiation cycles, and the results are shown in Table II.

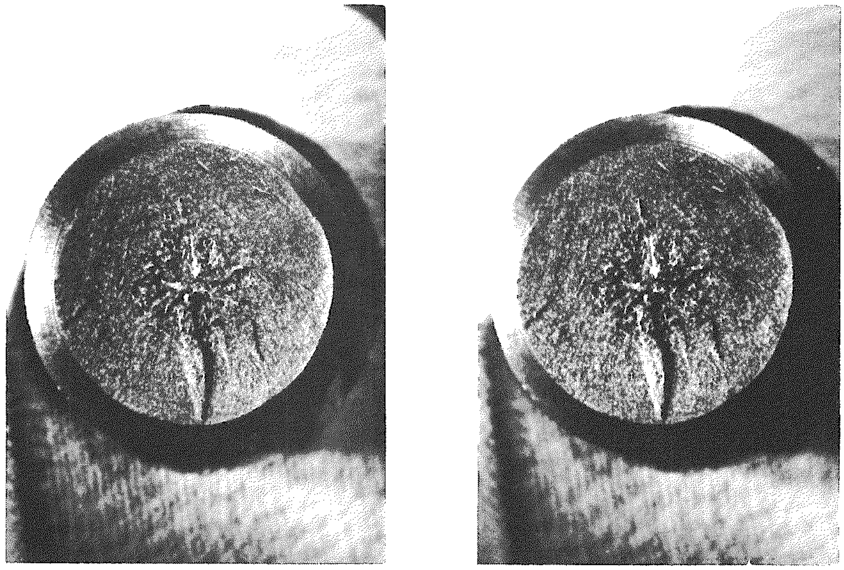
It can be noted that the more highly textured specimen, No. AC-26 developed only about 75% of the elongation under alpha-plus-beta irradiation cycling that was noted under alpha cycling. Figures 10 and 11 show this specimen before and after fracturing. From the fracture surfaces shown in Figure 11 it is apparent that the center of the specimen had been at a temperature above the limit of stability of the alpha phase.

The beta-quenched specimen, No. AC-65, was found to have increased in diameter to such an extent that it could not be removed from its irradiation capsule. The appearance of this specimen in its capsule is shown in Figure 12. A transverse cut was then made through the entire assembly. This operation disclosed that a large central void had developed in the specimen, as shown in Figure 13. The void may conceivably have developed from swelling due to fission gas, for similar large central voids have been noted in other fuel specimens irradiated at high temperatures.[3] However, in view of the very low burnup which the specimen received, it is believed that a more probable explanation for the cavity is that it resulted from stresses associated with cycling into the beta phase. Rather similar



18621 3X

Figure 10. Appearance of an irradiation-cycled 300°C rolled specimen, No. AC-26, which was irradiated with its center in the beta phase.

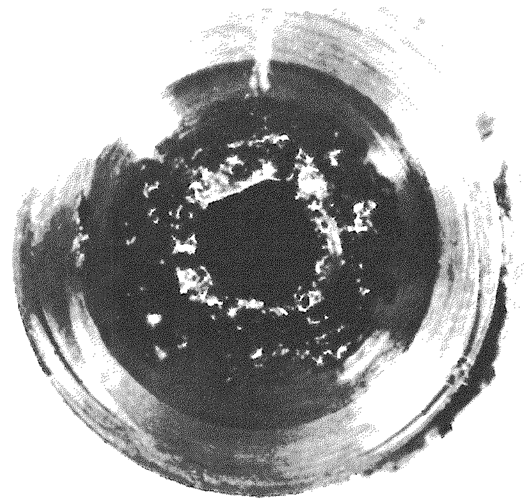


18834 3X

Figure 11. Fracture surfaces of the above specimen.

18626 3X

Figure 12. Appearance of an irradiation-cycled 300°C rolled and beta-quenched specimen, No. AC-65, which was irradiated with its center in the beta phase. The specimen had swelled to such an extent that it could not be removed from the irradiation capsule.



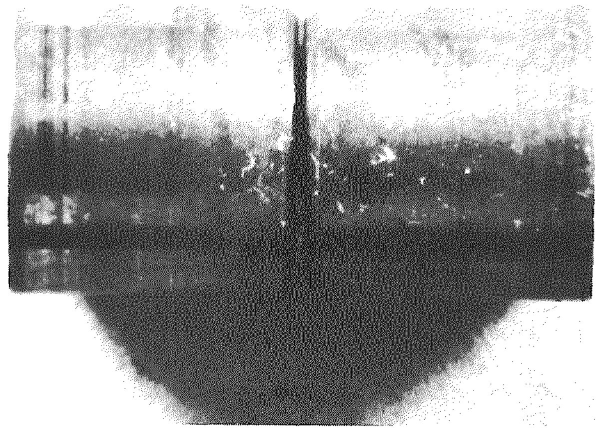
18625 3X

Figure 13. Transverse section of the above specimen. A large central void has developed during irradiation cycling.

cavities were obtained by Hayward[4] in uranium specimens thermally cycled 50 times between the alpha and beta phases. It is not known why the companion specimen rolled at 300°C, No. AC-26, did not show similar swelling. It is believed that the difference in behavior between the two specimens is probably related to the higher mechanical properties of the material rolled at 300°C, which would enable it to resist induced thermal stresses better, and to the pronounced difference in texture, which would alter the magnitude of the thermal stresses.

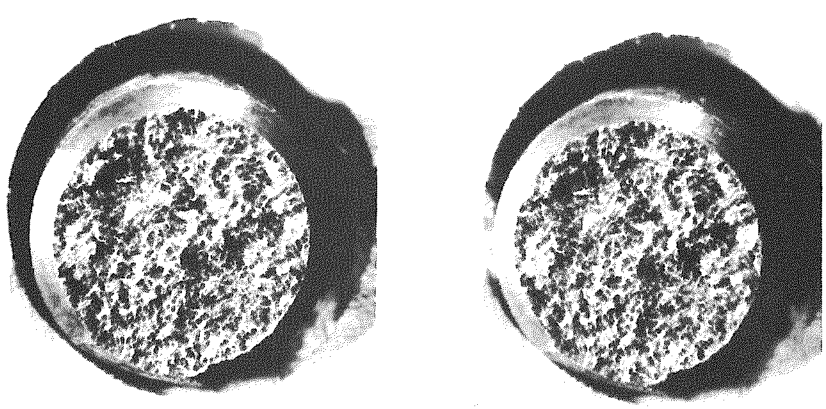
The third and final group of samples that were irradiated consisted of three specimens rolled at 300°C and beta-quenched, Nos. AC-62, AC-64, and AC-66, which were irradiated continuously for 12.5 hours. This was the same total exposure received by the cycled specimens in Groups 1 and 2. The specimens in Group 3 had diameters of 0.52, 0.56, and 0.60 inch respectively, so that both alpha-phase and alpha-plus-beta phase operation would be obtained. The changes which were observed are shown in Table II. Photographs of these specimens before and after fracturing are shown in Figures 14 through 20.

The specimen of greatest interest in this group was No. AC-66, which, as indicated by its fracture, was irradiated with its center at a temperature above the limit of stability of the alpha phase. A small dome-shaped projection developed at the center of one end, as is shown in Figure 18. An end-on view of this projection is shown in Figure 19. As can be noted in Figure 19, the small projection from the center of the specimen



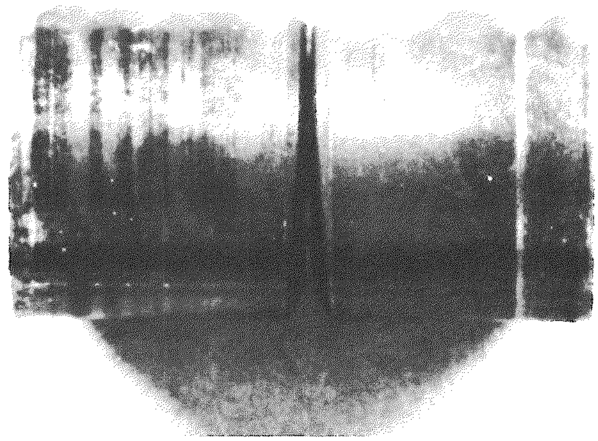
18623 3X

Figure 14. Appearance of a continuously irradiated 300°C rolled and beta-quenched specimen, No. AC-62, which remained in the alpha phase during irradiation.



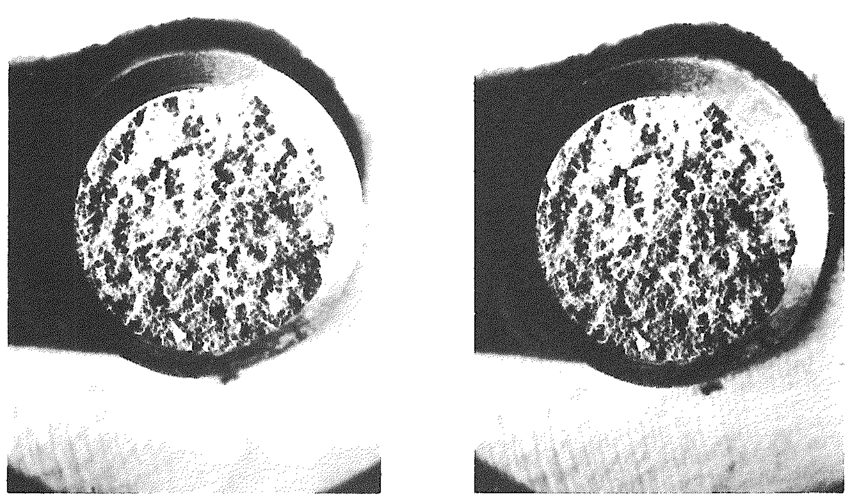
18843 3X

Figure 15. Fracture surfaces of the above specimen.



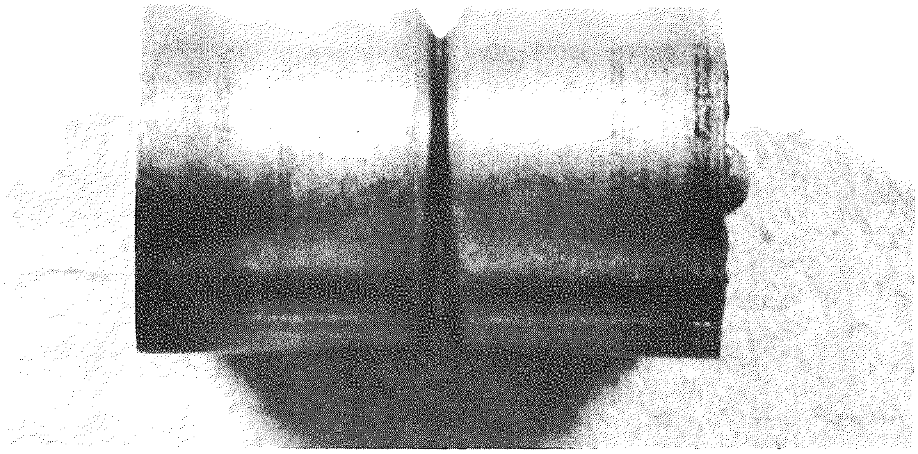
18624 3X

Figure 16. Appearance of a continuously irradiated 300°C rolled and beta-quenched specimen, No. AC-64, which remained in the alpha phase during irradiation.



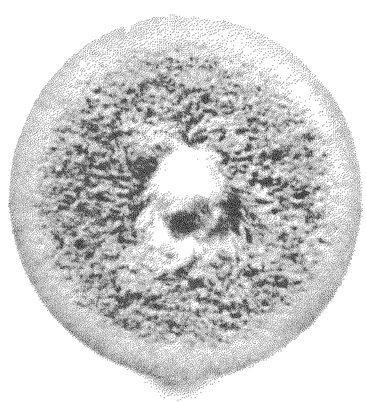
18844 3X

Figure 17. Fracture surfaces of the above specimens.



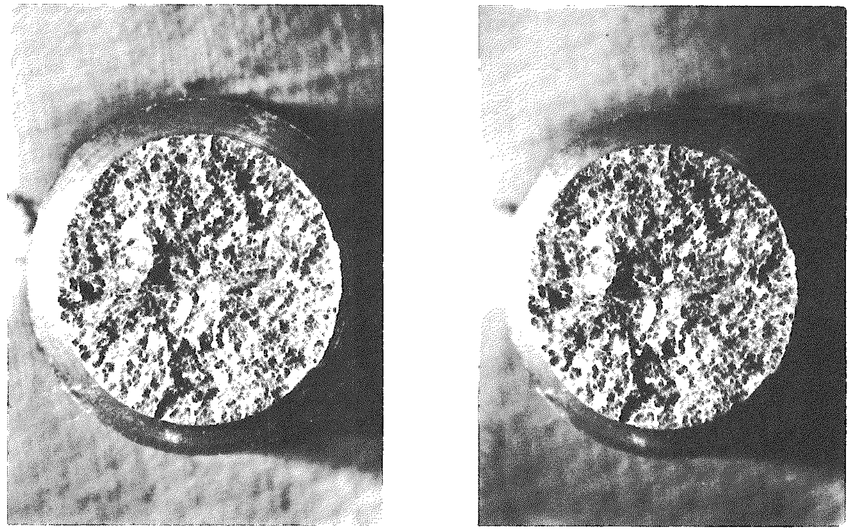
18628 3X

Figure 18. Appearance of a continuously irradiated 300°C rolled and beta-quenched specimen, No. AC-66, which was irradiated with its center above the alpha phase. A projection has formed on the right end of the specimen.



18629 3X

Figure 19. View of right end of above specimen.



18846 3X

Figure 20. Fracture surfaces of specimen No. AC-66. A radial array of fracture planes may be seen in the center of the specimen.

is surrounded by a relatively wide zone of roughened surface. This is, in turn, surrounded by an annulus with the smooth appearance of the original surface before irradiation. The existence of the three zones suggests that the specimen may have been operated with the center in the gamma phase, an intermediate annulus in the beta phase, and the outer part of the specimen in the alpha phase. The flux monitor attached to the capsule, however, did not indicate that this specimen received a higher integrated flux than the other specimens of similar diameter. Thus, in order for the center of specimen AC-66 to have reached the gamma phase, heat transfer must have been impaired in some manner or a period of high-flux irradiation occurred, which was effectively compensated for in the flux monitor by a period of lower flux level. It is believed that the projection resulted from the volume increase that would occur when the center transformed to the beta (or gamma) phase, since these phases are substantially lower in density than the alpha phase.(5) If the center of specimen AC-66 did reach the gamma phase, this may explain why similar projections did not develop on the specimens which may have been irradiated only in the beta phase.

Table III shows a comparison of the actual elongations of the specimens which were cycled in the alpha phase with the total elongations that might be predicted from known thermal cycling and irradiation growth rates of the materials. It can be noted that the actual elongation in each case is higher than predicted, indicating that the effects of concurrent irradiation and thermal cycling may be greater than simply the additive effect from either phenomenon acting alone. However, statistics would require that more specimens be irradiated to establish this preliminary indication as an experimental fact.

TABLE III

Comparison of Actual Elongations of Irradiation-Cycled Specimens with Predicted Elongations from Irradiation and Thermal Cycling Acting Alone

Specimen Number	AC-22	AC-61
Condition	300°C rolled	300°C rolled, beta-quenched
Predicted Elongation from Irradiation, %	2.8	0.2
Predicted Elongation from Thermal Cycling, %	1.8	0.2
Total Predicted Elongation, %	4.6	0.4
Actual Elongation, %	5.0	0.8

CONCLUSIONS

Based on the limited number of specimens used in this investigation, the following conclusions may be drawn:

1. Uranium which has been rolled at 300°C so that it is highly oriented begins to elongate under irradiation at burnups as low as 0.0002 a/o (2 MWD/T).

2. Irradiation cycling of uranium rolled at 300°C and of uranium rolled at 300°C and beta-quenched may result in greater elongations than would be predicted by assuming additivity of the effects of irradiation and thermal cycling acting alone.

3. Although highly textured uranium prepared by rolling at 300°C is more subject to anisotropic irradiation growth than is nominally randomly oriented material, rolled at 300°C and beta quenched, it is much less subject to gross distortion resulting from partial beta-phase irradiation.

4. The distortion which occurs in uranium, rolled at 300°C and beta quenched, irradiated partially in the beta phase is more severe if cycling conditions exist during irradiation.

ACKNOWLEDGMENTS

The author is indebted to S. L. Friederichs, project engineer at the MTR, who performed the irradiation cycling. Measurements on the specimens before and after irradiation were done by R. J. Fousek, F. Pausche and C. H. Gebo.

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