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IRRADIATION OF TZM - URANIUM DIOXIDE FUEL PIN AT 1700 K

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16. Abstract

A fuel pin clad with TZM and containing solid pellets of uranium dioxide (UO_2) was fission heated in a static helium-cooled capsule at a maximum surface temperature of 1700 K for approximately 1000 hr and to a total burnup of 2.0 percent of the uranium-235. The 0.76-cm-outside-diameter fuel pin had a fueled length of approximately 5.0 cm and a tube wall thickness of 0.125 cm. The density of the fueled core of stacked solid pellets was 95 percent of theoretical density, and the core had 6.4 g of UO_2 of 93.15 percent enrichment. The fuel-clad gap was 0.01 cm. The fuel pin contained a void of approximately 36 percent. About 3 percent of the void was in the porosity in the fuel pellets, and approximately 32 percent was in a plenum at the end of the fuel region. The remainder was in the clearance between the fuel and the clad.

After irradiation the fuel pin was measured to check dimensional stability and examined metallographically to determine the effect of the irradiation. Burnup was determined in several sections of the fuel pin.

The results of the postirradiation examination indicated

1. A transverse, intergranular failure of the fuel pin occurred when the fuel pin reached 2.0-percent burnup. This corresponds to 1.33×10^3 kW-hr/cm^3, where the volume is the sum of the fuel, clad, and void volumes in the fuel region.

2. The maximum swelling of the fuel pin was less than 1.5 percent on the fuel-pin diameter.

3. There was no visible interaction between the TZM clad and the UO_2.

4. Irradiation at 1700 K produced a coarse-grained structure, with an average grain diameter of 0.02 centimeter and with some of the grains extending one-half of the thickness of the clad.

5. Below approximately 1500 K, the irradiation of the clad produced a moderately fine-grained structure, with an average grain diameter of 0.004 centimeter. The maximum grain size was less than 0.1 of the clad thickness. Irradiation at this temperature probably would have resulted in increased fuel-pin life.

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SUMMARY

A fuel pin clad with TZM and containing solid pellets of uranium dioxide (UO_2) was fission heated in a static helium-cooled capsule at a maximum surface temperature of 1700 K for approximately 1000 hours and to a total burnup of 2.0 percent of the uranium-235. The 0.76-centimeter-outside-diameter fuel pin had a fueled length of approximately 5.0 centimeters and a tube wall thickness of 0.125 centimeter. The density of the fueled core of stacked solid pellets was 95 percent of theoretical density, and the core had 6.4 grams of UO_2 of 93.15 percent enrichment. The fuel-clad gap was 0.01 centimeter. The fuel pin was approximately 36 percent void. About 3 percent of the void was in the porosity in the fuel pellets, and approximately 32 percent was in a plenum at the end of the fuel region. The remainder was in the clearance between the fuel and the clad.

After irradiation the fuel pin was measured to check dimensional stability and examined metallographically to determine the effect of the irradiation. Burnup was determined in several sections of the fuel pin.

The results of the postirradiation examination indicated

1. A transverse, intergranular failure of the fuel pin occurred when the fuel pin reached 2.0-percent burnup. This corresponds to 1.33×10^3 kilowatt-hours per cubic centimeter, where the volume is the sum of the fuel, clad, and void volumes in the fuel region.

2. The maximum swelling of the fuel pin was less than 1.5 percent on the fuel-pin diameter.

3. There was no visible interaction between the TZM clad and the UO_2.

4. Irradiation at 1700 K produced a coarse-grained structure, with an average grain diameter of 0.02 centimeter and with some of the grains extending one-half of the thickness of the clad.

5. Below approximately 1500 K, the irradiation of the clad produced a moderately fine-grained structure, with an average grain diameter of 0.004 centimeter. The maximum grain size was less than 0.1 of the clad thickness. Irradiation at this temperature probably would have resulted in increased fuel-pin life.

INTRODUCTION

The use of helium as a coolant for high-temperature nuclear reactors in the temperature range 1400 to 1700 K (ref. 1) permits the use of molybdenum-base alloys as a fuel-pin clad, since molybdenum has a melting temperature of 2883 K and a low thermal cross section and is compatible with uranium dioxide (UO_2) fuel. In this irradiation it was desired to test the irradiation behavior of the molybdenum-base alloy TZM (molybdenum with 0.5 percent titanium and 0.08 percent zirconium).

The design for the lightweight, compact, helium-cooled reactor for air-cushion vehicles requires high burnups and high total energy release density. The irradiation was planned to determine the overall performance and stability of TZM - solid-pellet-UO_2 fuel pins when irradiated in a helium coolant at a maximum clad surface temperature of 1700 K and with an axial temperature profile simulating the reactor profile. To simulate a gas-cooled reactor, the TZM-UO_2 fuel pin was irradiated in a capsule with natural convective cooling by helium.

APPARATUS

Fuel Pin

The fuel pin irradiated in this test had a TZM clad (molybdenum, 0.5 percent titanium, and 0.08 percent zirconium) and a 5-centimeter stack of 18 solid pellets of UO_2 of 93.15 percent enrichment. The assembly and dimensions of the fuel pin are shown in figure 1.

The clad was fabricated from solid TZM alloy bar stock by gun drilling the inside diameter and grinding the outside diameter. End caps were machined from solid molybdenum bar stock. The plenum sleeves and support rings were machined from swaged molybdenum tubing having a 0.64-centimeter (outside) diameter and a 0.12-centimeter wall thickness. Electrical discharge machining was used to fabricate the tungsten wafers and spring from unalloyed tungsten foil. The spectral analysis of the TZM is shown in table I.

The UO_2 pellets were prepared by cold-pressing and sintering techniques. The density of the sintered pellets was 95 percent of theoretical density. The fuel column was fabricated to exact length by grinding the final pellet. The analysis of the UO_2 is shown in table II.

The fuel pin was assembled in a welding box, evacuated to 1×10^{-4} torr, backfilled with helium, and tungsten-inert-gas (TIG) welded closed. Molybdenum conduction insulators on the fuel pin end pieces were used to minimize end heat conduction losses and to flatten the fuel-pin axial temperature profile during irradiation. The fuel-pin end

pieces had a thermocouple well for measurement of the fuel-pin temperature during irradiation.

Irradiation Facility

The irradiation was performed in a capsule with static helium. A schematic of the irradiation capsule is shown in figure 2. The heat was transferred from the fuel pin to the capsule walls by natural convective heat transfer in the helium. The capsule was 304 stainless steel with a shrunk-on outer jacket of 606 1-T6 aluminum. During irradiation a slow bleed flow of helium was continuously monitored to detect the first release of fission products. The fuel pin was held in the capsule in a horizontal position by 0.05-centimeter-thick molybdenum plates. Temperatures on the fuel pin were measured with tungsten-5-percent-rhenium - tungsten-26-percent-rhenium thermocouples banded to the surface of the fuel pin.

PROCEDURE

Irradiation

Prior to insertion of the capsule into the reactor, the capsule was pressurized to the required helium pressure for design cooling at the design heat generation rate. Then the capsule was inserted into the reactor flux until the desired temperature was obtained. Any effects on temperature of small changes in reactor flux were compensated for by adjustment of the helium coolant pressure. When the reactor was shut down, the capsule was removed from the reactor flux; after reactor startup the capsule was reinserted.

Postirradiation Examination

The fuel pin was first examined as it was removed from the capsule in order to record gross changes in appearance. The fuel pin was then measured to detect changes in dimensions and sectioned into nine pieces (fig. 3). Pieces 4 and 8 were used for burnup determination, and the remaining pieces were used for metallographic samples. The samples were mounted in epoxy resin, ground through 600 grit, and polished with Linde B alumina.

RESULTS AND DISCUSSION

Fuel-Pin Measurement

The irradiation of the fuel pin was terminated after 937 hours at temperature when a clad failure was indicated by the release of fission products. When the capsule was opened, visual examination showed that the fuel pin was ruptured transversely. The irradiated pin is shown in figure 4. Measurement of the pin diameter along the length of the pin showed that the swelling of the fuel pin was less than 1 percent.

Burnup

Burnup was measured on sections 4-BU and 8-BU by change in uranium isotopes. The burnup in 937 hours is recorded in table III. The burnup results indicate that there was a uniform burnup and a uniform power per cubic centimeter of fuel along the length of the fuel pin.

Irradiation Temperature

Thermocouples were fixed to the fuel pin at the four locations shown in figure 5. The axial temperature profile of the fuel pin is also shown in figure 5. The temperature was lower at the ends because of heat conduction from the ends of the fuel pin. The average temperature variation during irradiation was ±38 K.

The temperature of irradiation was sufficiently high to vaporize and redistribute the UO_2. Longitudinal and transverse sections of the fuel are shown in figure 6. The radial symmetry of the fuel in the transverse sections indicates that the radial temperature and radial heat transfer were symmetrical.

Postirradiation Fuel Characteristics

Figure 3 shows the fuel profile in the postirradiated fuel pin. Temperatures from sections 3-L to 9-L were sufficiently high for sintering of the fuel to full density. The calculated smeared density of the postirradiated fuel - center-void combination was 93 percent, and the measured preirradiated fuel density was 95 percent. The center void in the fuel resulted from the redistribution during irradiation of the 95-percent-dense preirradiated fuel.

Longitudinal and transverse sections of the fuel for specimens 2-T, 7-T, and 9-L are shown in figure 7. The fuel has the characteristic columnar grains, lenticular voids, and extensive cooldown thermal cracking that is characteristic of irradiation at this high temperature (ref. 2).

A transverse section of the fuel pin at position 5-T, close to the fuel-pin maximum temperature, is shown in figure 8 at a magnification of 75. There was no visible reaction between the fuel and clad, and there was no bonding between the fuel and clad. The same fission gas porosity and thermal cracks are present as in the sections in figure 7. In addition, the higher magnification shows the beginning of the deposition of solid metallic fission products. The 2-percent burnup at the irradiation temperature of 1700 K marks the beginning of the agglomeration of solid metallic fission products into particles large enough to observe at a magnification of 75.

Figure 9 shows, in section 1-L, the boundary between the depleted UO_2 pellet and the enriched UO_2. In the preirradiated pin the depleted UO_2 and the adjacent enriched UO_2 were separate pellets. During the irradiation the enriched UO_2 diffused into the depleted UO_2 to approximately one-third the depth of the depleted UO_2 pellet (approximately 0.1 cm).

The characteristics of the postirradiated UO_2 are as expected from irradiation to 2-percent burnup at 1700 K. The structure indicates that the fission gas can escape from the UO_2 to the center plenum formed by the fuel redistribution. The fuel cracking and separation of the fuel from the clad provide pathways for movement of the fission gas to the plenum at the cold end of the fuel pin. As expected from previous work (ref. 2), the UO_2, even though extensively cracked, remains locked in position in the fuel pin.

Postirradiation Clad Characteristics

When the capsule was opened, it was observed that the fuel pin was broken transversely at section 6-L (figs. 4 and 6). A photograph of the broken surface is shown in figure 10. The photograph shows coarse-grained intergranular fracture in the TZM clad.

Photomicrographs at a magnification of 75 across the clad at sections 2-T, 5-T, and 7-T are shown in figures 11(a), (b), and (c). The TZM clad of sections 5-T and 7-T at the high temperature (1700 K) has formed large grains during the 937-hour irradiation, some of which extend approximately one-half of the thickness of the clad. Sections 5-T and 7-T were adjacent to the maximum fuel-pin temperature at section 6-L. The fuel-pin rupture was at the point of maximum fuel-pin temperature. Section 3-L, shown in figure 11(d), is moderately fine-grained. There is no grain which extends one-tenth the thickness of the clad. The operating temperature of this section was approximately

1500 K. At 2-percent burnup and 937 hours, the temperature of 1500 K is sufficiently low to preclude an extremely coarse-grained and weak structure which could lead to transverse rupture.

The helium in the capsule was monitored continuously for fission products. Fission products were detected during operation and indicated that the failure occurred during irradiation and while at temperature. The rupture occurred at the position of maximum temperature and in a region of large grains.

From figures 11(a), (b), and (c) it is seen that there was no visible reaction between the clad and the fuel during the 937-hour irradiation at any temperature up to and including the maximum temperature of 1700 K (ref. 3).

SUMMARY OF RESULTS

A solid-pellet uranium dioxide (UO_2) fuel pin with a TZM clad was fission heated in a capsule for 937 hours to achieve 2-percent burnup. The fuel pin was helium-cooled by natural convection. The UO_2 was 93.15 percent enriched. The results of the irradiation were as follows:

1. After reaching 2-atom-percent burnup of the uranium-235 and while at temperature, the fuel pin ruptured transversely at the position of maximum temperature. The irradiation of TZM at 1700 K resulted in intergranular rupture in the coarse-grained clad, in which some large grains extended approximately one-half the thickness of the clad.

2. Diametral swelling of the fuel pin was less than 1.5 percent. This increase in diameter was within the design limits.

3. There was no visible reaction between the TZM clad and the UO_2 or the fission products.

4. Below approximately 1500 K the irradiated clad had a moderately fine-grained structure with an average grain extending only approximately 1/30 of the clad thickness and with a maximum grain size less than 1/10 of the clad thickness. Irradiation at this temperature probably would have resulted in increased fuel-pin life.

Lewis Research Center,
National Aeronautics and Space Administration,
Cleveland, Ohio, January 12, 1973,
501-24.

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1. Rom, Frank E.: Status of the Nuclear Powered Airplane. J. Aircraft, vol. 8, no. 1, Jan. 1971, pp. 26-33.

2. McDonald, Glen E.: Irradiation of Mo-UO_2 Fuel Pin at 2700° F to 8.5 Percent Burn-up. Trans. Am. Nucl. Soc., vol. 13, no. 2, Nov. 1970, pp. 564-565.

3. McDonald, Glen E.: Irraidation of TZM-UO_2 and TZM-UN Pins. Trans. Am. Nucl. Soc., vol. 15, no. 1, June 1972, pp. 190-191.

TABLE I. - SPECTRAL ANALYSIS OF TZM CLAD

Element	Concentration[a]	Element	Concentration[a]	Element	Concentration[a]
Silver	ND	Mercury	ND	Rubidium	NA
Aluminum	<M[b]	Indium	ND	Rhenium	NA
Arsenic	ND	Iridium	ND	Rhodium	ND
Gold	ND	Potassium	NA	Ruthenium	ND
Boron	ND	Lanthanum	ND	Antimony	ND
Barium	ND	Lithium	NA	Scandium	NA
Beryllium	ND	Magnesium	ND	Silicon	T
Bismuth	ND	Manganese	<T	Tin	ND
Calcium	NA	Molybdenum	SS	Strontium	ND
Cadmium	ND	Sodium	ND	Tantalum	ND
Cobalt	ND	Niobium	ND	Thorium	ND
Chromium	ND	Nickel	ND	Titanium	M
Cesium	NA	Osmium	NA	Uranium	ND
Copper	<T	Phosphorus	ND	Vanadium	ND
Iron	M	Lead	ND	Tungsten	ND
Gallium	ND	Palladium	ND	Yttrium	ND
Germanium	NA	Platinum	ND	Zinc	ND
Hafnium	ND	Plutonium	NA	Zirconium	M

[a]Not analyzed for, NA; moderate, M; analyzed for but not detected, ND; trace, T; major element, SS.

[b]Interference.

TABLE II. - ANALYSES OF URANIUM DIOXIDE

(a) Powder (spectral analysis)

Element	Concentration, ppm	Element	Concentration, ppm
Silver	0.1	Manganese	<5
Aluminum	<25	Molybdenum	<10
Boron	<.25	Nitrogen	81
Barium	<2.5	Sodium	<10
Beryllium	<.5	Nickel	46
Bismuth	<1	Phosphorus	<12.5
Calcium	<25	Lead	<1
Cadmium	<.2	Rubidium	<10
Chlorine	<10	Antimony	<5
Cobalt	<2.5	Silicon	<25
Chromium	<17	Tin	<2.5
Cesium	5	Strontium	<10
Copper	<10	Thorium	<8
Fluorine	10	Thallium	<10
Iron	40	Titanium	<25
Indium	<2.5	Vanadium	<25
Potassium	<10	Zinc	<25
Lithium	<1	Zirconium	<25
Magnesium	<17		

(b) Pellets (chemical analysis)

Element[a]	Concentration, ppm
Water	27
Fluorine	<2
Chlorine	<5
Carbon	30

[a]Oxygen-uranium ratio, 2.019.

TABLE III. - BURNUP ANALYSIS

Section	Burnup, at. %
4	1.94
8	1.89

TABLE II. - ANALYSES OF URANIUM DIOXIDE

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Silver	0.1	Manganese	<5
Aluminum	<25	Molybdenum	<10
Boron	<.25	Nitrogen	81
Barium	<2.5	Sodium	<10
Beryllium	<.5	Nickel	46
Bismuth	<1	Phosphorus	<12.5
Calcium	<25	Lead	<1
Cadmium	<.2	Rubidium	<10
Chlorine	<10	Antimony	<5
Cobalt	<2.5	Silicon	<25
Chromium	<17	Tin	<2.5
Cesium	5	Strontium	<10
Copper	<10	Thorium	<8
Fluorine	10	Thallium	<10
Iron	40	Titanium	<25
Indium	<2.5	Vanadium	<25
Potassium	<10	Zinc	<25
Lithium	<1	Zirconium	<25
Magnesium	<17		

(b) Pellets (chemical analysis)

Element[a]	Concentration, ppm
Water	27
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Chlorine	<5
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4	1.94
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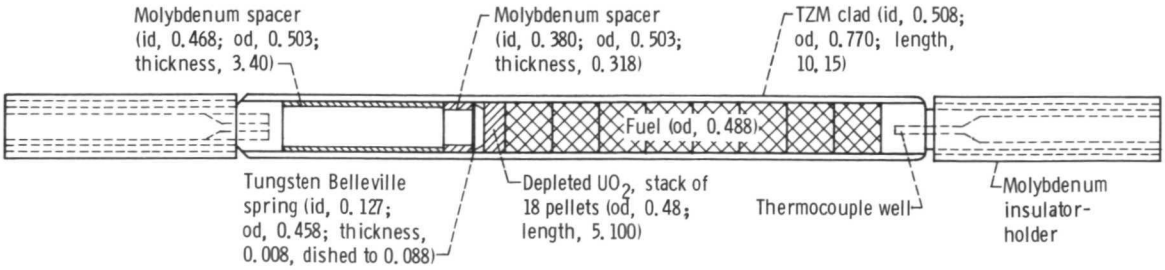


Figure 1. - Fuel pin. (Dimensions in centimeters.)

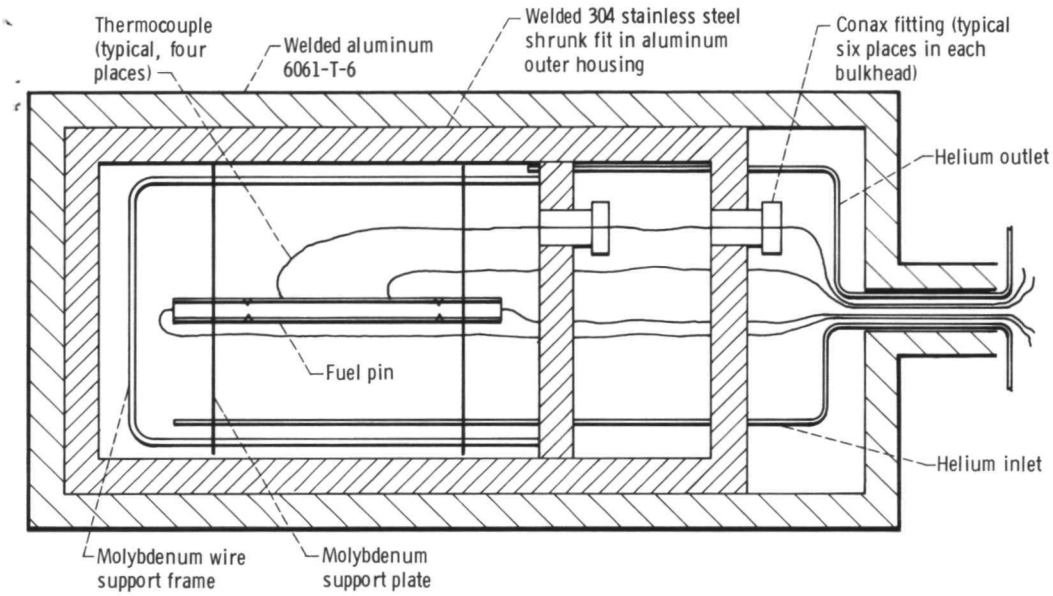


Figure 2. - Irradiation capsule.

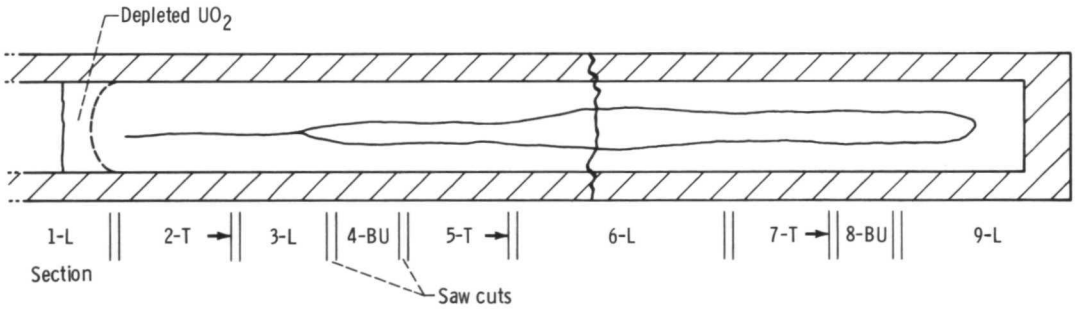


Figure 3. - Metallographic and burnup specimen location and postirradiation fuel profile. x5.

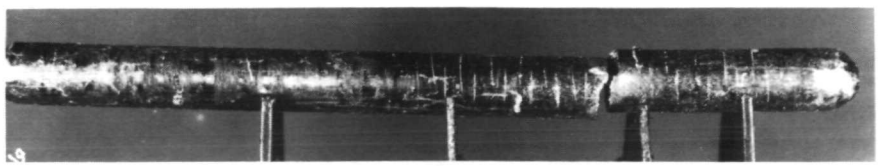


Figure 4. - Irradiated fuel pin.

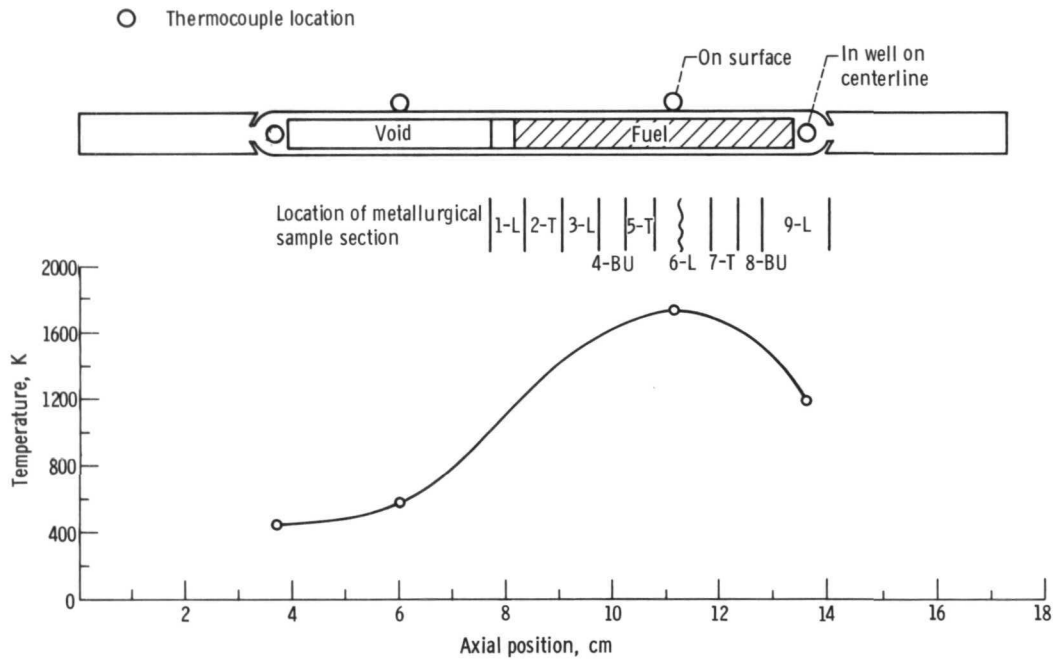


Figure 5. - Temperature distribution.

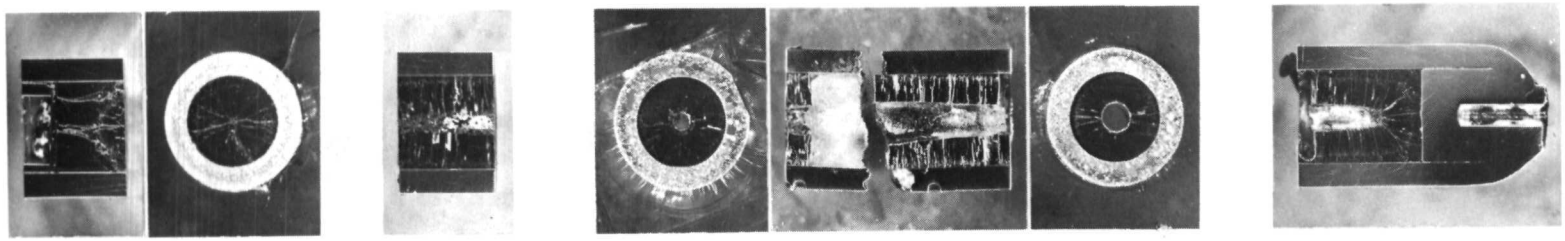
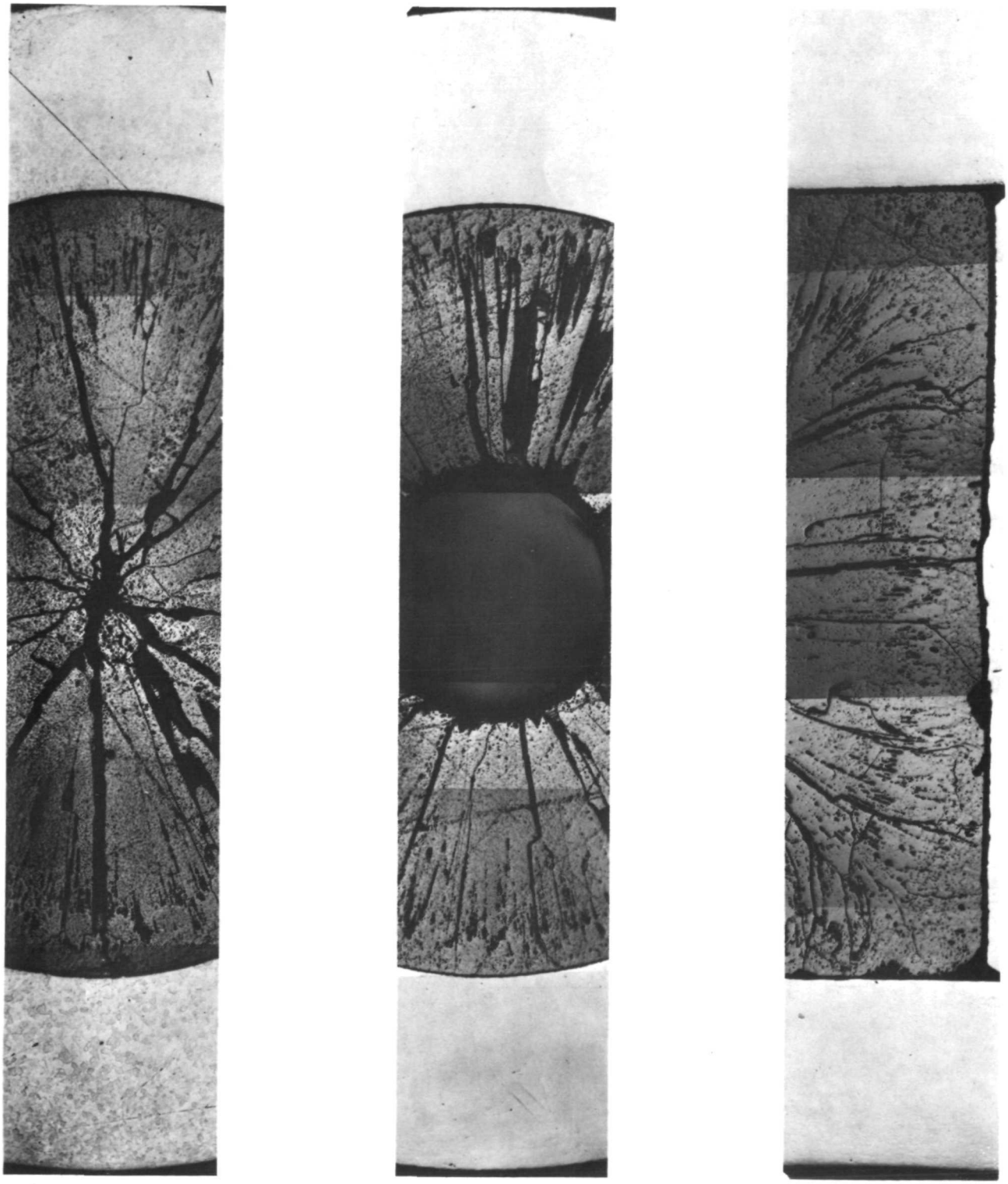


Figure 6. - Irradiated fuel-pin sections.



2-T 7-T 9-L

Figure 7. - Photomicrographs of irradiated fuel-pin sections. x28.

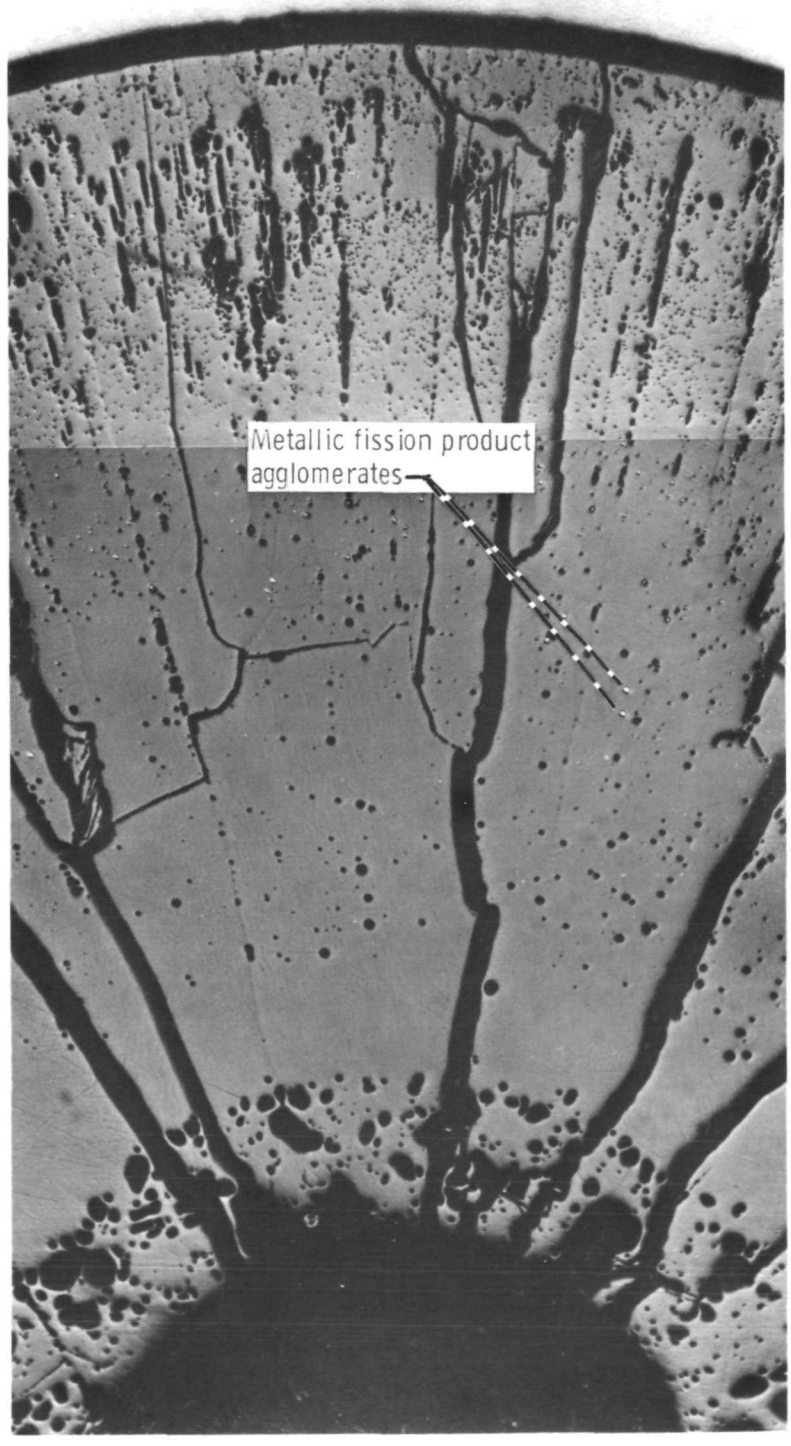


Figure 8. - Photomicrograph of irradiated fuel-pin section 5-T. x75.

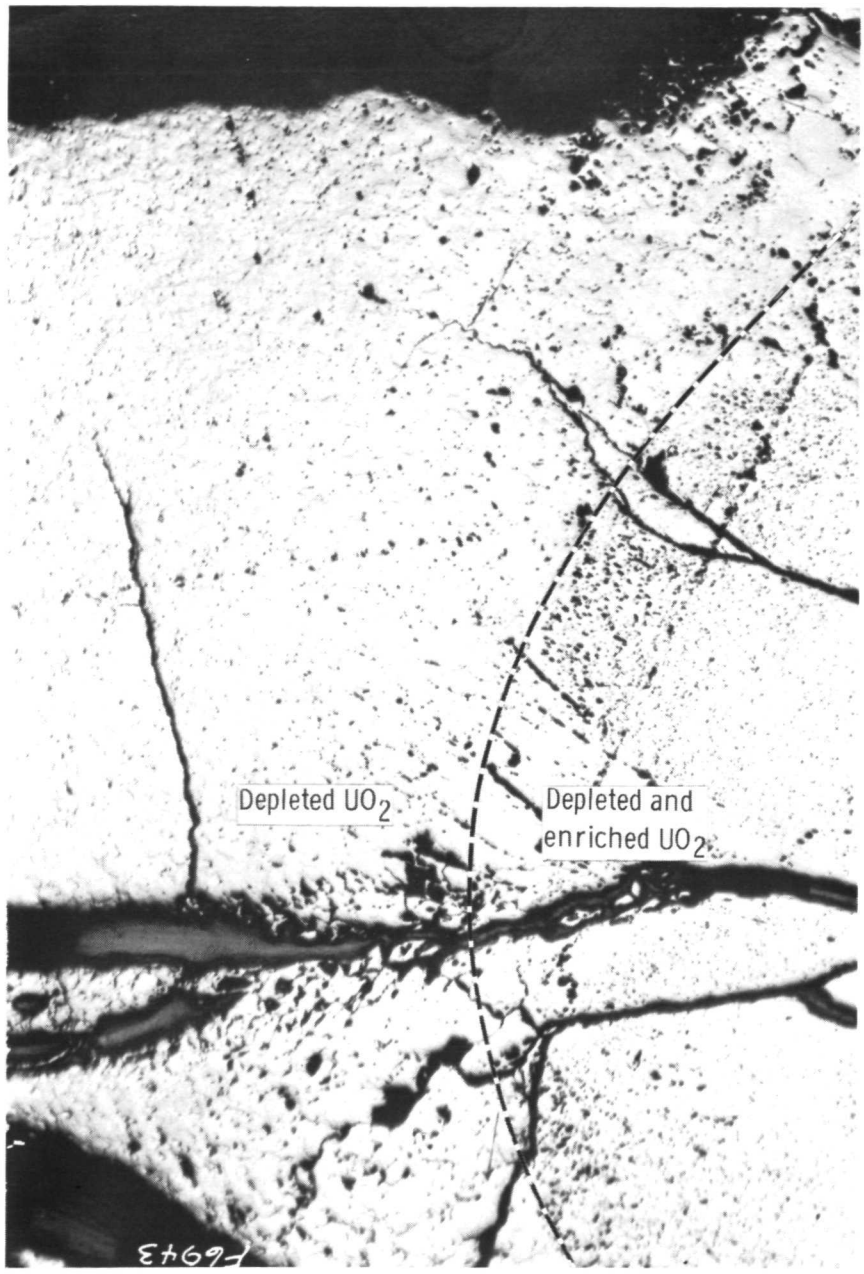


Figure 9. - Boundary between depleted UO_2 and depleted and enriched UO_2 in section 1-L. x75.

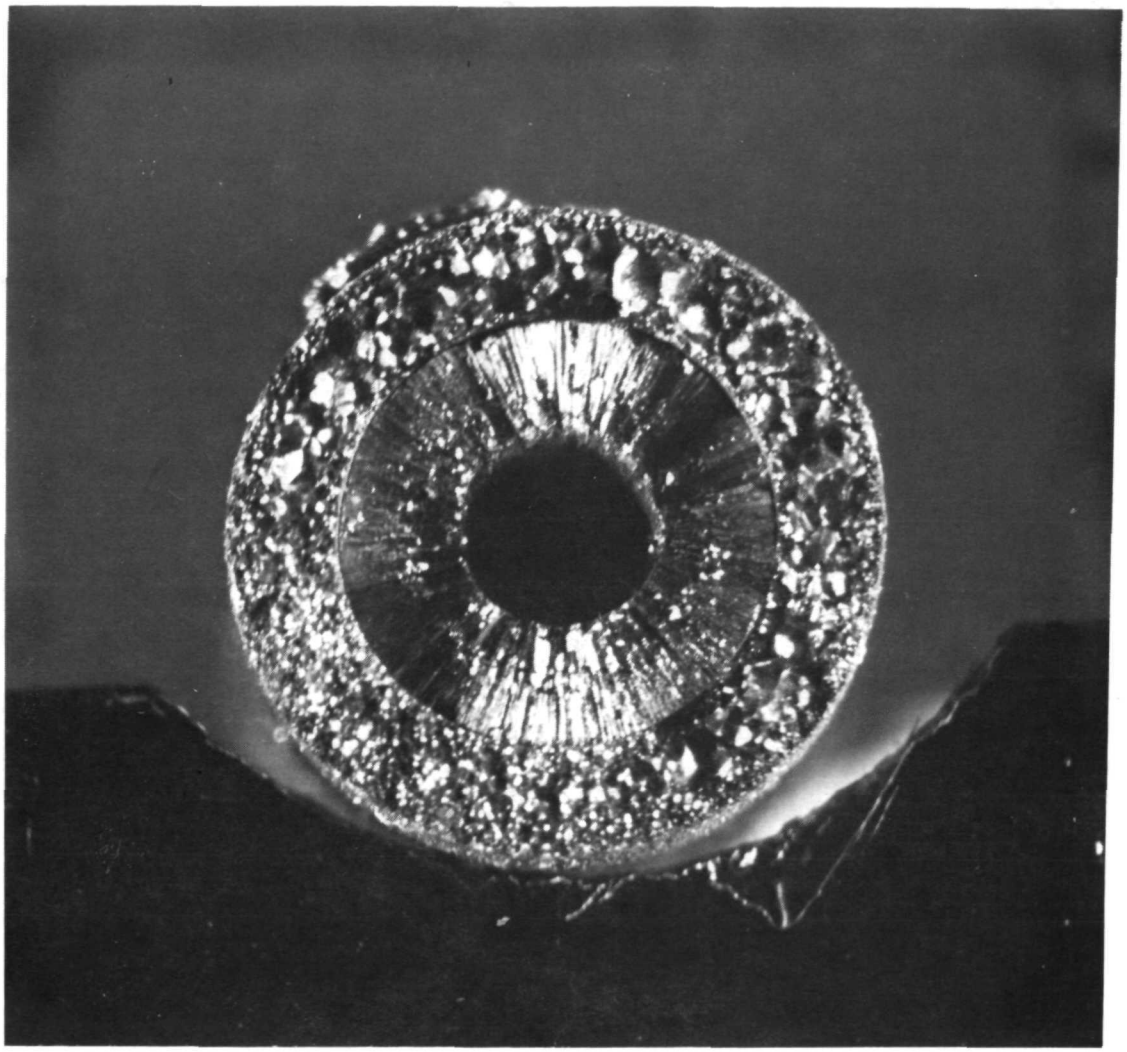
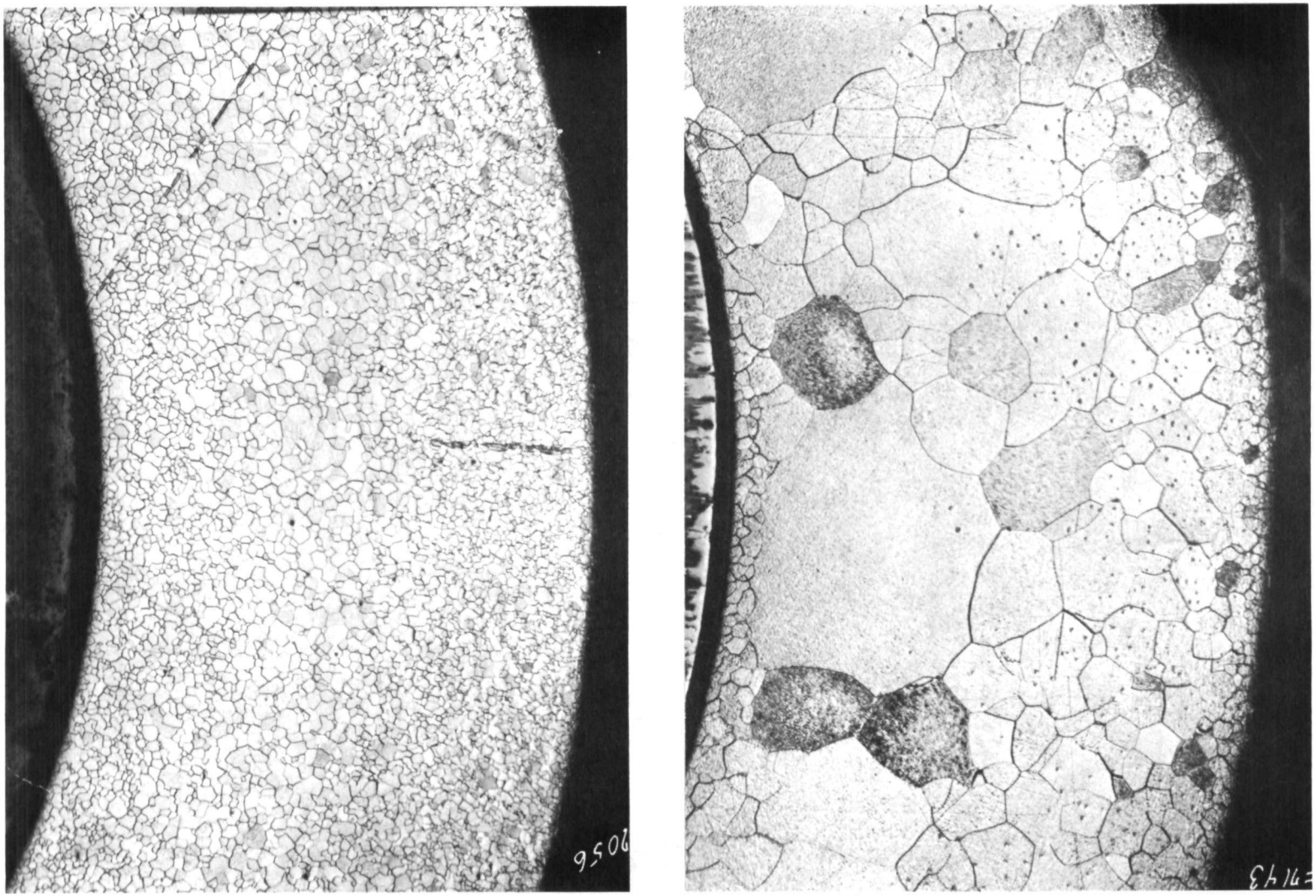


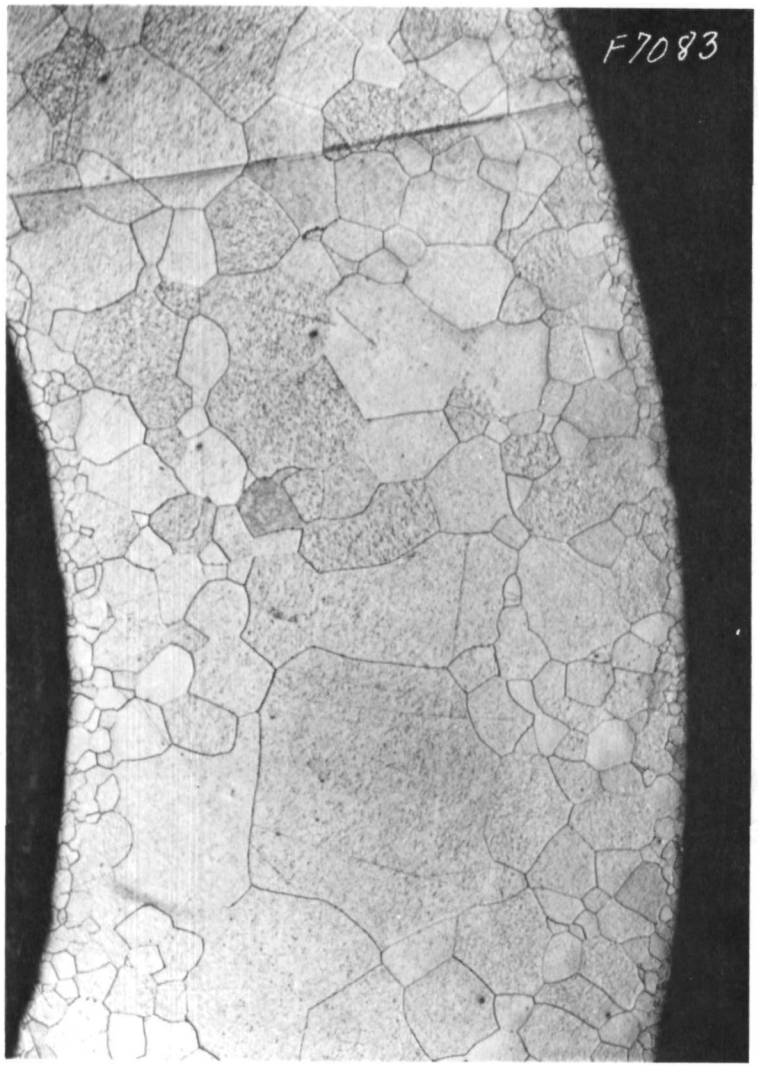
Figure 10. - Fractured surface of fuel pin. x12.



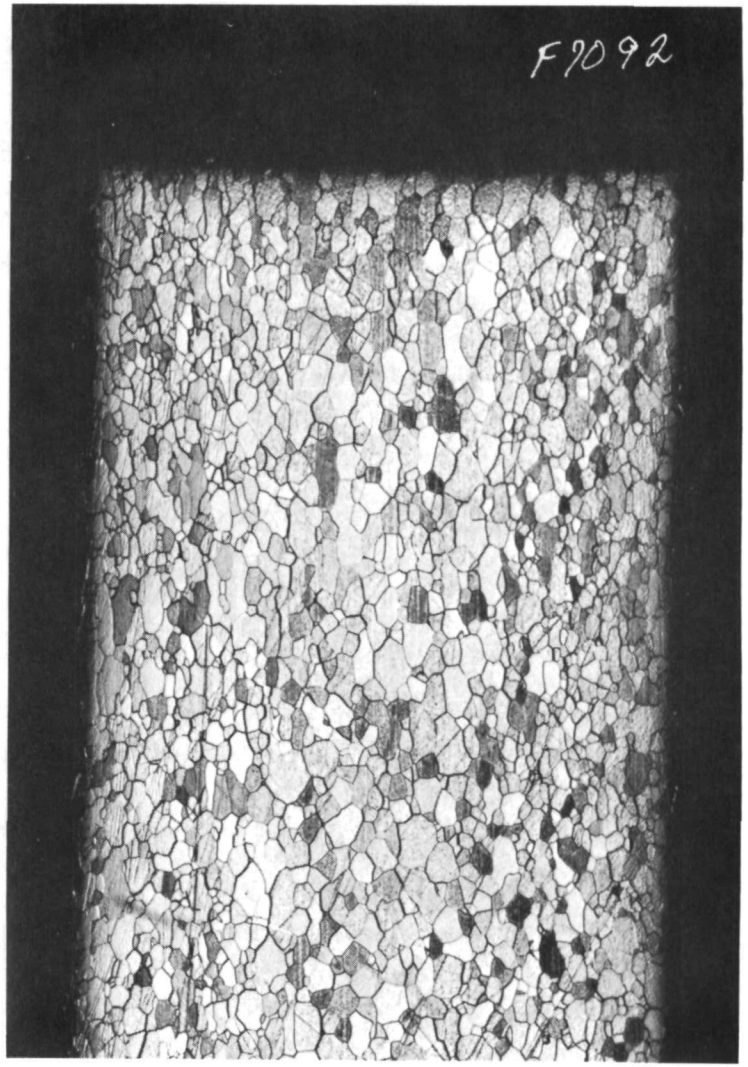
(a) Section 2-T.

(b) Section 5-T.

Figure 11. - Photomicrographs of clad. Etched; x75.



(c) Section 7-T.



(d) Section 3-L.

Figure 11. - Concluded.