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IRRADIATION EFFECTS ON MASSIVE URANIUM MONOCARBIDE

by

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The results of examinations of UC compounds having nominal compositions of uranium-4.6, –4.8, and –5.0 w/o carbon are encouraging after irradiation to burnups of from 400 to 15,000 MWD/T of uranium. Density changes were small, varying from a minimum of 0.3 per cent to a maximum of about 2.5 per cent. Cracking occurred in all specimens; however, it can probably be largely attributed to thermal stresses and to oxidation, after decapsulation, of NaK entrapped in microcracks. Depletion of carbon appears to be occurring in the specimens having the nominal uranium-5 w/o carbon composition. Metallographic examination shows that the UC_2 phase disappears at high temperature and high burnup. The fission-gas-retention properties of the compounds appear quite good. In all cases, the amount of fission gas released was comparable with the calculated amount released by recoil.

INTRODUCTION

A research program which is designed to evaluate massive uranium carbide as a possible fuel material for sodium-, organic-, and gas-cooled reactors is being conducted at Battelle Memorial Institute. Atomics International, acting for the U. S. Atomic Energy Commission, is directing and supporting this program. Uranium monocarbide was chosen for study because of its high thermal conductivity, its high uranium density, its refractory character, and its potential adaptability to fabrication of high-density shapes by casting techniques.(1) Uranium monocarbide is quite reactive with water; therefore, if it proves to be a successful reactor fuel, its initial use will probably be for the reactor types mentioned above.

The program herein described is concerned with the effects on cast uranium carbide specimens of irradiation in the Battelle Research Reactor at Columbus, Ohio, and at the Materials Testing Reactor at the AEC testing site in Idaho. To obtain initial irradiation information on massive uranium carbide, 12 specimens were irradiated at the BRR in a simple NaK-cooled capsule to a burnup of about 400 MWD/T of uranium. Five test capsules were irradiated in the MTR. One capsule is currently being irradiated in the MTR. The specimens experienced burnups from 400 to a maximum of about 15,000 MWD/T of uranium at temperatures which ranged from a minimum core temperature of about 660 F to a maximum of about 1600 F.

PREPARATION OF THE URANIUM CARBIDE SPECIMENS

Cylindrical uranium monocarbide castings were made by directly alloying the elements by arc-melting techniques. Cast buttons of the material were made by arc melting freshly pickled enriched uranium with crushed spectrographic-grade carbon in a water-cooled copper crucible under a helium atmosphere of 10 to 20 in. of mercury

(1) References at end.

absolute. The melting was done in a charge-flipping arc furnace with a carbon-tipped electrode. Each button was flipped over and remelted six times. In the cases where previously melted uranium monocarbide was used, only four remelts were necessary to achieve homogeneity. A low cooling rate on the final melt was used to prevent the buttons from fracturing.

The cast UC specimens were made from these buttons by using a "drop-casting" technique. This involved placing a button of uranium monocarbide over the opening of a graphite thimble mold and heating sufficiently with an electric arc to cause all of the UC to melt and drop into the cylindrical mold. With this technique, relatively sound homogeneous castings were produced. These castings were then ground on a diamond wheel to final size. Figure 1 shows the type of copper mold used, the size of the button casting, and the resultant cylindrical casting which was produced.

Evaluation of the finished castings included carbon analyses, density measurements, dimension measurements, and metallographic and radiographic examinations. These tests were conducted on only those machine-ground specimens which visually appeared to be sound and were of sufficient length to be possible candidates for the irradiation tests.

Twelve cast specimens of uranium monocarbide were prepared for the initial irradiation in the BRR. Each casting was 3/8 in. in diameter and was subsequently ground to 1/4 in. in diameter. The specimens were about 1/2 in. long. The carbon content of the uranium carbide was 5 w/o, slightly above the stoichiometric 4.8 w/o. This composition was used in the initial work to avoid having to consider possible effects of alpha uranium, present in substoichiometric UC, in evaluating the data to be obtained.

Twelve castings, about 1/2 in. in diameter and about 3 in. long, were prepared for the capsules that were to be irradiated in the MTR. Specimens 3/8 in. in diameter and 2 in. long were machined from the castings. Also, 1/2- and 1/4-in.-long specimens of the same diameter were prepared. The 1/2-in.-long specimens were used to separate the actual test specimens during the irradiation, and the 1/4-in.-long pieces were placed at the top of the top specimen and the bottom of the bottom specimen to reduce end effects during the tests. Aside from visual examination, these specimens were not evaluated in the study.

To study the effects of carbon content upon irradiation behavior of this material, test specimens containing nominally 5, 4.8, and 4.6 w/o carbon were made. Table 1 summarizes capsule and specimen identification data.

N51387

FIGURE 1. ASSEMBLED MOLD WITH ARC-MELTED UC BUTTON CHARGE AND FINISHED CASTING

TABLE 1. CAPSULE AND SPECIMEN IDENTIFICATION AND SPECIMEN COMPOSITION

Capsule	Specimen	Description	Carbon Content[a], w/o	Design Burnup, MWD/T of uranium
BRR	12	All solid	5.0-5.57	700
BMI-23-1	2	Top drilled for thermocouple	5.2	1,000
		Bottom solid	5.2	
BMI-23-2	2	Top drilled for thermocouple	5.3	5,000
		Bottom solid	5.1	
BMI-23-3	2	Top drilled for thermocouple	5.0	10,000
		Bottom solid	5.0	
BMI-23-4	2	Top drilled for thermocouple	5.2	20,000[b]
		Bottom solid	5.3	
BMI-23-5	2	Top drilled for thermocouple	4.6	5,000
		Bottom solid	4.7	
BMI-23-6	2	Top drilled for thermocouple	4.8	5,000
		Bottom solid	4.6	

(a) Determined by metallography, density, and carbon analysis from top of cast ingot.
(b) Capsule BMI-23-4 is still being irradiated in the MTR.

PREIRRADIATION EXPERIMENTS

Compatibility Experiments With NaK

At the outset of this irradiation program, NaK could not be used as the heat-transfer medium in the test capsules until there was assurance that there would be no interaction with the UC or stainless steel. Also, it was unknown whether common solvents like acetone, carbon tetrachloride, butyl alcohol, etc., would react with the UC during the pre- and postirradiation cleaning operations. Since little information could be found in the literature, several simple compatibility experiments were conducted.

Uranium carbide specimens were placed in molybdenum and in Type 304 stainless steel or molybdenum baskets, sealed in NaK-loaded stainless steel capsules under a helium atmosphere, and heated for extended periods in an electric furnace. The test conditions and results are summarized in Table 2.

TABLE 2. UC-NaK COMPATIBILITY TESTS

Test	Basket Material	Exposure, weeks	Temperature, F	Weight Change of 10-G Specimen, mg	
				Basket	Specimen
1	Type 304 stainless steel	2	1100	-0.4	-3.6
2	Molybdenum	2	1100	-0.3	-4.5
3	Molybdenum	2	1300	+0.3	-5.5
4	Molybdenum	6	1300	0.0	-5.6
5	Type 304 stainless steel	12	1100	(a)	-13.6
6	Molybdenum	12	1100	-2.0	-22.4

(a) Stainless steel baskets became brittle and fragments were lost so that weight changes could not be assessed. Except for embrittlement, the baskets appeared to be unchanged.

The specimen baskets were essentially unchanged except for the embrittlement of the Type 304 stainless steel basket during the 12-week exposure at 1100 F. To explore this embrittlement, metallographic examinations were made of the UC specimens from the 12-week experiments and of samples from the steel basket. Results indicated that the embrittlement of the steel was probably caused by a carbide precipitate (Cr_4C) at the grain boundaries and possibly by the formation of an unknown phase at these boundaries and along strain lines within the grains. It is possible that the unknown

phase may have been sigma or some other embrittling phase resulting from diffusion of one of the elements from the testing environment. However, the possibility that the UC contributed to the situation was tempered somewhat by the fact that the microstructure of the surface of the UC specimens examined appeared to be normal.

The UC specimens exposed in these experiments remained intact but showed slight discoloration at points of contact with the baskets. They exhibited small weight losses (a few milligrams in 10-g samples), as shown in Table 2. However, no measurable change in specimen density was observed, and there was no evidence of NaK penetration into the specimen surfaces.

Embrittlement of unstabilized stainless steel is not surprising since the test temperature of 1100 F is in the middle of a sensitizing (carbide precipitation) range, 900 to 1400 F. The use of a stabilized stainless steel such as Type 347 and the elimination of cold working would, of course, minimize such a tendency; however, Type 304 was used throughout the program.

Compatibility Experiments With Solvents

In addition to the experiments with NaK, the compatibility of UC with various liquids was examined to determine those suitable for use in preirradiation and postirradiation examination. Compatibility was determined by observing UC specimens, before, during, and after exposure and by comparing pretest and posttest specimen weights. Tests were run at room temperature for periods of 1, 3, and 5 hr with water, acetone, methyl alcohol, ethyl alcohol, butyl alcohol, benzene, xylene, kerosene, CCl_4, and Zyglo.

Reactions were observed only with water. During the 1-hr exposure gas bubbles were evolved from the specimen, and during the 3-hr exposure some flaking occurred. The change in specimen weight with liquids other than water was insignificant (less than 1 mg in 10-g samples). Also, weight changes were insignificant after allowing some UC specimens to stand in a Drierite desiccator for 2 days and for 2 months. Also, no change was observed after heating for 2 and 17 hr in a vacuum oven at 180 F and 550 μ of pressure.

Drop Tests

In drop tests and thermal-shock tests, UC specimens were spring loaded in a stainless steel expanded-metal basket and placed in a stainless steel capsule filled with a liquid medium, as they would be during irradiation in the MTR. Acetone at room temperature was the liquid medium in the drop test because its density and its viscosity are similar to those of NaK at 1100 F. The capsule was dropped with its axis horizontal onto a concrete floor from heights of 1, 2, and 3 ft. Then the capsule was dropped with the axis vertical. The specimens did not fracture during any of these mechanical shocks.

Thermal-Shock Tests

In the thermal-shock test, good specimen shock resistance was demonstrated. Two capsules containing specimens immersed in NaK were heated in an electric furnace to 1300 F and then plunged into room-temperature water. This cycle, during which the capsules cooled to room temperature in about 2 min, was repeated ten times. Radiographs taken after the first, fifth, and tenth cycles indicated that the specimens did not fracture.

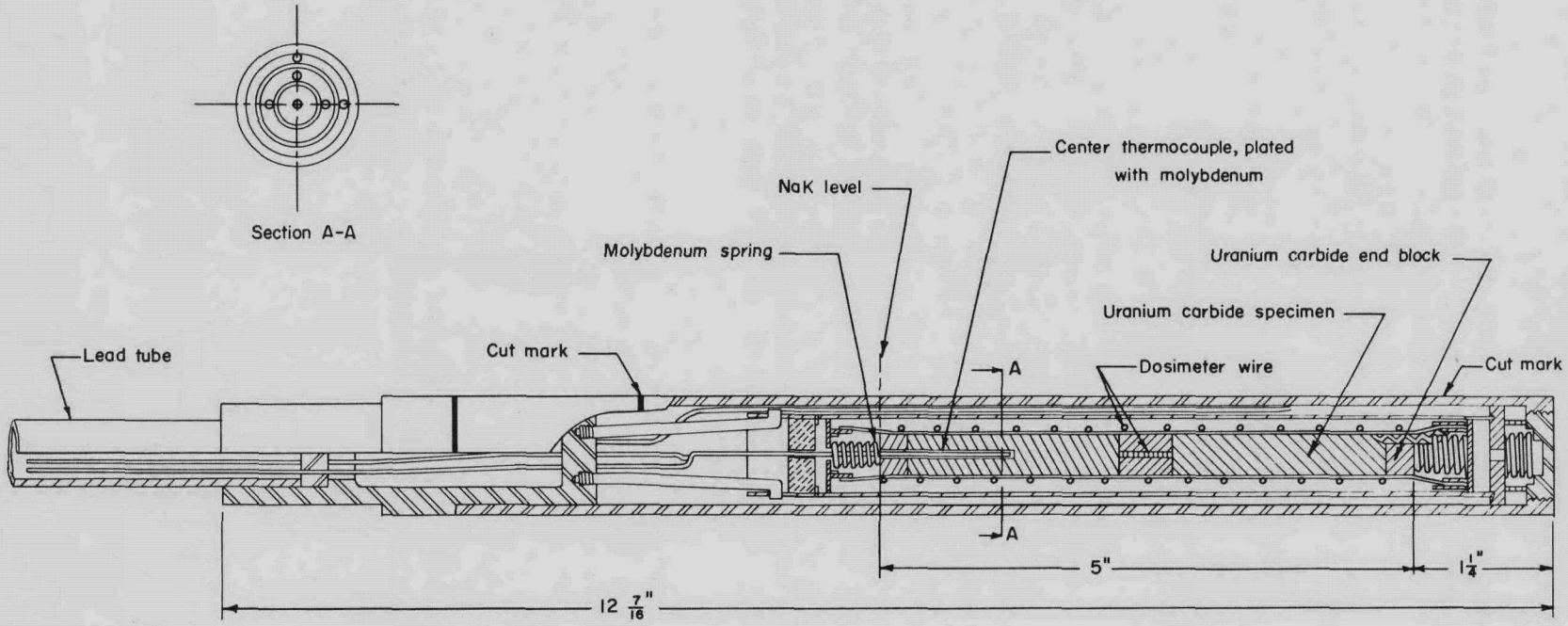
CAPSULE-DESIGN INFORMATION

The capsule design for the MTR capsules is shown in Figure 2. Figures 3 and 4 are views of the BRR capsule and the MTR capsule, respectively. The BRR capsule consisted of a simple cylinder which contained sufficient NaK to cover the 12 specimens. The top cap was provided with a spring which kept the specimens in close contact during the irradiation.

The fuel stack in each MTR capsule consisted of five unclad 3/8-in.-diameter UC specimens. The two test specimens were about 2 in. long and were located in the central zone of the capsule. A 1/4-in.-long spacer was placed at each end of the stack to minimize external and nuclear end effects and a 1/2-in.-long spacer separated the actual test specimens. The top 1/4-in. specimen and the top half of the upper 2-in.-long specimen were drilled longitudinally down the center to accommodate a 1/16-in.-OD thermocouple. The hole was drilled ultrasonically.

An expanded stainless steel basket retained the specimens in a stack. Adequate space was provided to permit unrestrained radial growth of the specimens during irradiation. Molybdenum springs were placed at the ends of the stack to keep the specimens in a firm position yet allow axial growth. Six thermocouples were provided to record temperature data at various positions within the capsule. Dosimeters were provided inside and outside of the capsule body.

After the capsule body was filled with NaK, the capsules were sealed by Heliarc welding the end caps to the capsule wall. The thermocouples were brazed into the headers with a nickel-alloy braze with a melting temperature of about 1800 F.



All materials of construction to be Type 304 stainless steel unless otherwise specified

FIGURE 2. BASIC DESIGN OF THE MTR CAPSULES FOR THE IRRADIATION OF URANIUM MONOCARBIDE

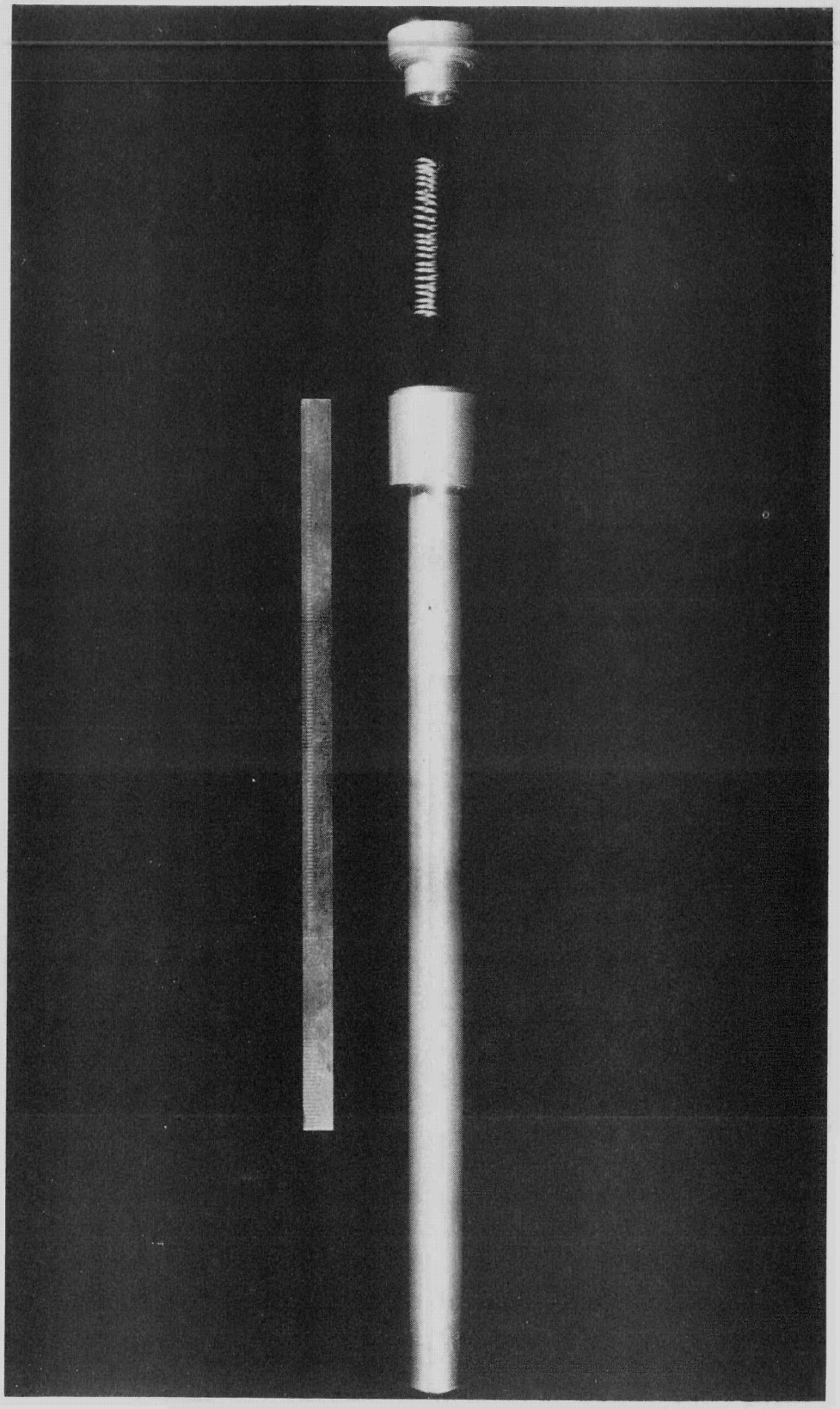


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FIGURE 3. VIEW OF THE PARTIALLY ASSEMBLED BRR CAPSULE

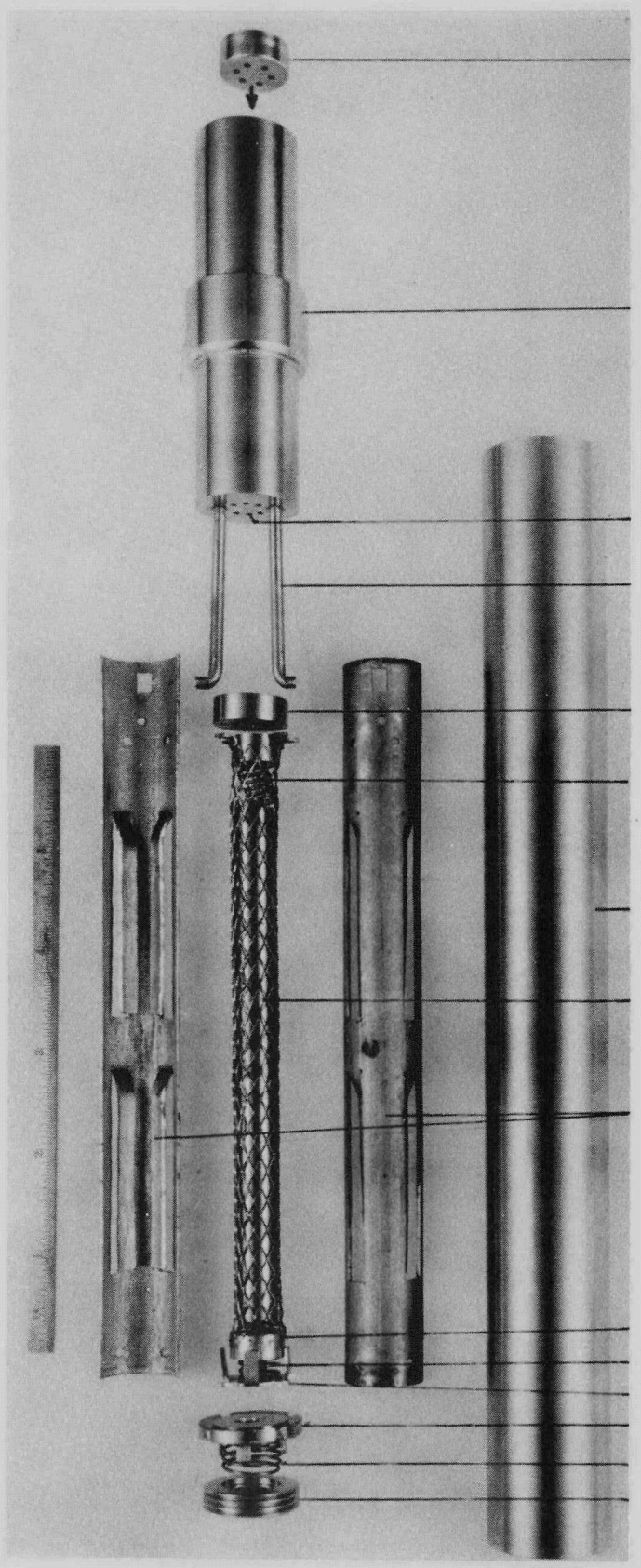

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FIGURE 4. VIEW OF THE PARTIALLY ASSEMBLED MTR CAPSULE

IRRADIATION HISTORY AND POSTIRRADIATION EXAMINATIONS

The irradiation history and postirradiation data of the specimens are summarized in Table 3. As can be seen in Table 3, the diameter measurements showed a change of only 0.16 to 0.28 per cent for the BRR specimens. For the MTR specimens, the diameter measurements increased from a minimum of 0.3 to a maximum of 1.2 per cent. A comparison of these data indicates that no correlation can be observed between the burnup levels and the diameter change. Some cracking did occur in all of the MTR specimens, and it is believed that this cracking could easily have caused these dimensional changes.

The density changes for all of the irradiated specimens were relatively quite small. Again, no correlation can be made between the burnup data and the density data. Because of the cracks present in the specimens during these measurements, it is felt that the actual densities of the whole pieces of irradiated specimens were probably somewhat higher than the data indicate.

Figures 5, 6, and 7 show the typical appearance of the specimens from the MTR irradiation prior to and after irradiation. The various cracks in each irradiated specimen are quite obvious. The cracks were both radial and transverse. Some spalling of the surface occurred in several specimens and an example of this is shown in Figure 6. Unfortunately, the exact determination of the amount of cracking which occurred in a specimen during irradiation is somewhat difficult to determine. Further cracking of the specimens occurred immediately upon removal of the specimens from the butyl alcohol bath. (Butyl alcohol was used to dissolve the NaK heat-transfer medium in the capsules.) This further cracking is apparently caused by the hydrolysis and subsequent expansion of the NaK trapped in the microcracks. In a comparison of all the specimens from all the MTR capsules except BMI-23-4, which is still in-pile, the specimens from Capsule BMI-23-3 appeared to exhibit a somewhat greater degree of cracking. However, while specimen burnup in Capsule BMI-23-3 was about two times that of the other capsules, the amount of specimen cracking was only about 25 per cent more than in the others.

The radial cracking is, of course, attributed to the thermal stresses caused by the temperature gradients between the core and surface. Further cracking is induced by the number of thermal cycles the specimens experienced during the irradiation. The specimens which contained alpha uranium in the grain boundaries – for example, Specimens 42 and 46 (both uranium-4.6 w/o carbon) – did not appear to experience any more or less cracking than did the specimens of stoichiometric composition or above which received about the same level of burnup.

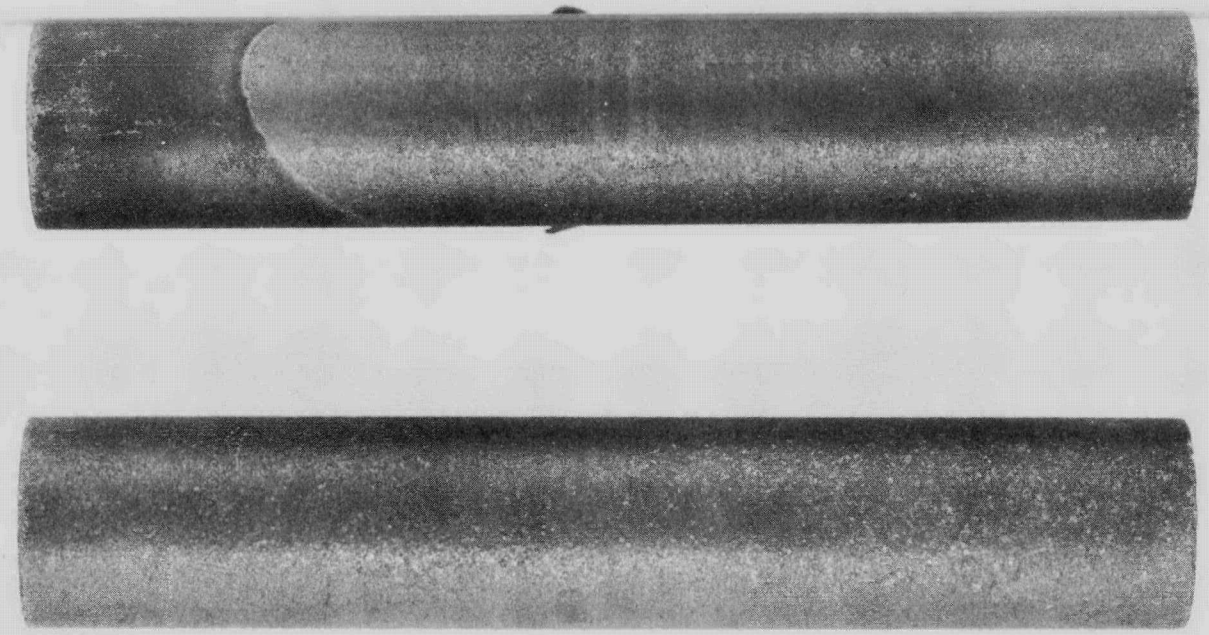
The core temperatures of the top specimens in all of the MTR capsules were determined by readings taken from a thermocouple located in the center of each top specimen. The surface temperatures for both the top and bottom specimens were determined by readings taken from thermocouples located near the surface of the specimens. The core temperatures for the bottom specimens were estimated from the readings of the thermocouples on the surfaces of the bottom specimens.

As can be seen from the data in Table 3, the average surface temperature for all six capsules, including the BRR capsule, ranged from 540 to a maximum of about 915 F. Core temperatures were from 660 to about 1600 F. Except for the BRR specimens

TABLE 3. IRRADIATION HISTORY AND POSTIRRADIATION DATA FOR UC SPECIMENS FROM CAPSULES BRR, BMI-23-1, BMI-23-2, BMI-23-3, BMI-23-5, AND BMI-23-6

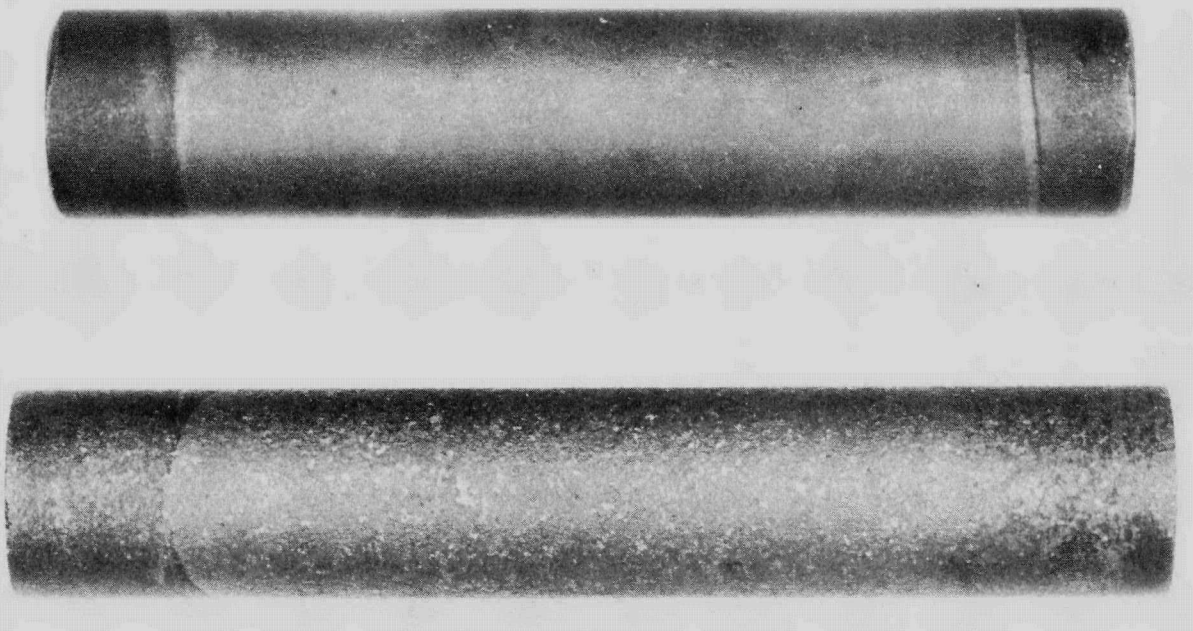
	BRR			BMI-23-1		BMI-23-2		BMI-23-3		BMI-23-5		BMI-23-6	
				Top	Bottom	Top	Bottom	Top	Bottom	Top	Bottom	Top	Bottom
	12A	15A	25A	3	2	26	37	28	29	42	50B	41	46
Nominal Carbon Content, w/o	5.0	5.0	5.0	5.0	5.0	5.0	5.0	5.0	5.0	4.6	4.6	4.8	4.8
Carbon Content by Determination[a], w/o	5.00	5.4	5.57	5.2	5.2	5.3	5.1	5.0	5.0	4.6	4.7	4.8	4.6
Diameter Change, per cent	0.16	0.28	0.16	0.4	0.3	0.9	0.9	--	0.8	0.8	1.2	0.48	0.48
Density Change, per cent	0.29	0.45	0.44	0.7-0.9	2.5	0.6-2.0	2.5	1.8	1.8	2.4	1.6	1.2	1.6
Average Temperature, F													
Surface	600	600	600	770	540	915	700	765	623	882	772	850-780	500-700
Core	660	660	660	1300	880	1375	1060	1178	1086	1326	1248	1300-1600	800-1100
Fission-Gas Release, cm^3 of krypton-85													
Estimated Total Produced				5.9×10^{-2}		20.9×10^{-2}		--		20.4×10^{-2}		21.9×10^{-2}	
Estimated Released by Recoil				2.5×10^{-5}		8.8×10^{-5}		--		8.7×10^{-5}		9.4×10^{-5}	
Measured Release	Negligible			2.4×10^{-5}		9.7×10^{-5}		(b)		8.22×10^{-5}		8.37×10^{-5}	
Burnup, MWD/T of uranium[c]													
Nominal	700	700	700	1,000		5,000		10,000		5,000		5,000	
Dosimetry	400	400	400	1,420	1,780	5,600	7,800	12,200	12,800	6,500	6,400	--	--
Radiochemical[d]	--	--	--	--	--	--	--	15,650	--	8,600	--	6,500	--
Isotopic[e]	--	--	--	--	--	--	--	13,750	--	8,100	--	5,900	--
Estimated Average Heat-Generation Rates, 10^3 Btu/(ft^2)(hr)													
Based on Dosimetry Burnup Data	--	170	--	680	850	880	940	--	695	--	710	--	--
Based on Isotopic Burnup Data	--	--	--	--	--	--	--	750	--	900	--	600	--

(a) The estimated composition is based on metallography, density, and a carbon analysis from the top of the cast ingots.
(b) Fission-gas sample was lost, apparently by leakage from capsule.
(c) Based on a 2200-lb ton and a fission energy of 195 Mev.
(d) Based on an analysis for cesium-137.
(e) Based on an analyses for uranium-236 by mass spectrometry.



3X RM11164

a. Specimens in Capsule BMI-23-3



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b. Specimens in Capsule BMI-23-6

FIGURE 5. TYPICAL SPECIMENS FROM THE MTR CAPSULES PRIOR TO IRRADIATION

4X HC3897

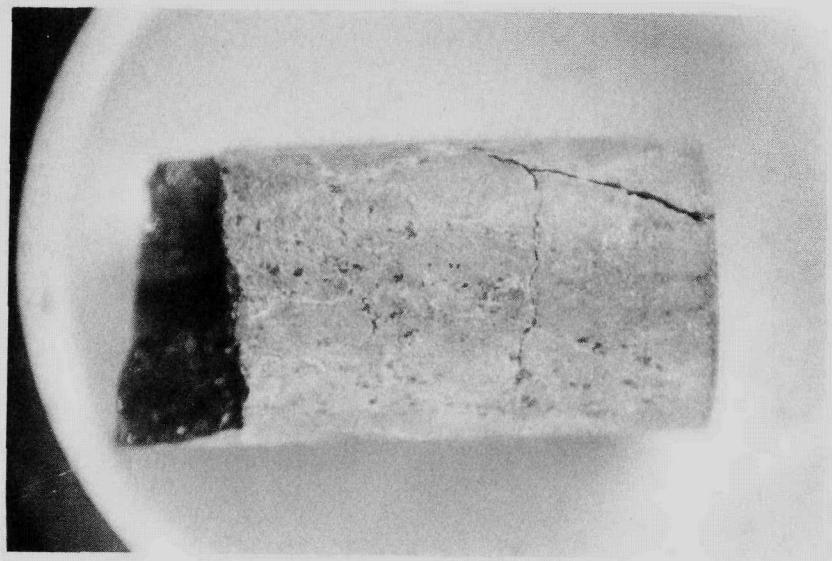
a. Part of Bottom Specimen

Note spalling at right.

4X HC3892

b. Part of Top Specimen

FIGURE 6. SPECIMENS OF IRRADIATED UC FROM CAPSULE BMI-23-3



4X HC3884

4X HC3882

FIGURE 7. TOP SPECIMEN OF IRRADIATED UC FROM CAPSULE BMI-23-6

which experienced a temperature differential of only about 60 F, the specimens experienced a temperature differential from surface to core in the range of about 400 to 550 F for the top specimens and about 350 to about 500 F for the bottom specimens. These temperature differentials are probably of a magnitude which would cause the circumferential and normal stresses within the specimens necessary to cause cracking. The elevated-temperature properties of UC are not well known, however, and, therefore, an accurate estimate cannot be made of the stresses required to cause fracturing.

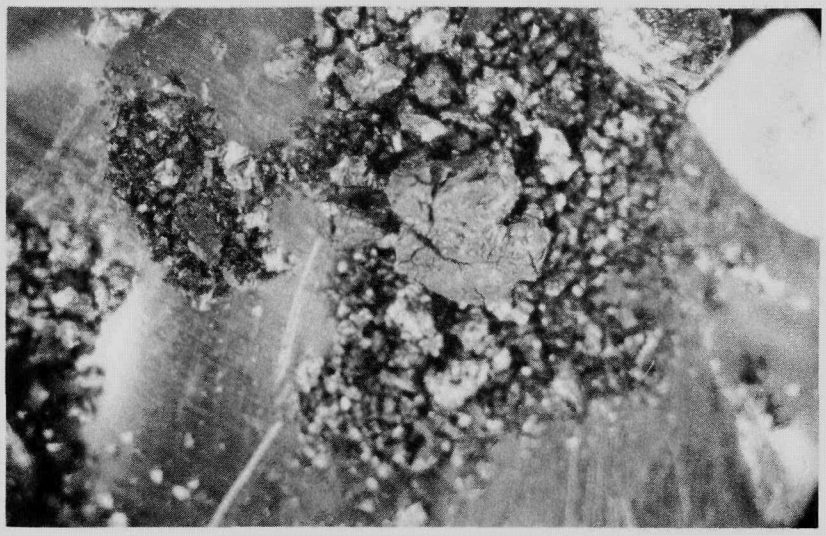
Prior to the actual opening of the capsules, each capsule was punctured and sampled for fission gas. The measured fission gas recovered from all the capsules is shown in Table 3. The gas sample from the BRR was so small that it was negligible. The sample from Capsule BMI-23-3 was inadvertently lost during the sampling operation, or the gas may have leaked from the capsule body.

It is well known that fission products will escape from the surfaces of irradiated unclad fuel specimens by recoil; therefore, a calculation was made to determine an estimate of the amount of fission gas released by this mechanism. Table 3 shows the estimated total krypton-85 produced in the UC specimens, the estimated amount released by recoil, and the actual amount measured after irradiation. For Capsules BMI-23-1, BMI-23-2, BMI-23-5, and BMI-23-6, this actual measured amount is essentially the same as that estimated to be released by recoil.

As part of this irradiation-effects study an experiment was conducted with an irradiated and an unirradiated UC specimen to determine the effect of a heat treatment on the irradiated material. The experiment was conducted to determine if any changes would occur in the density, the dimensions, or the microstructure of the specimens and also to measure and analyze any fission gas released during the heat treatment. The irradiated specimen which was used for this study was from Capsule BMI-23-2.

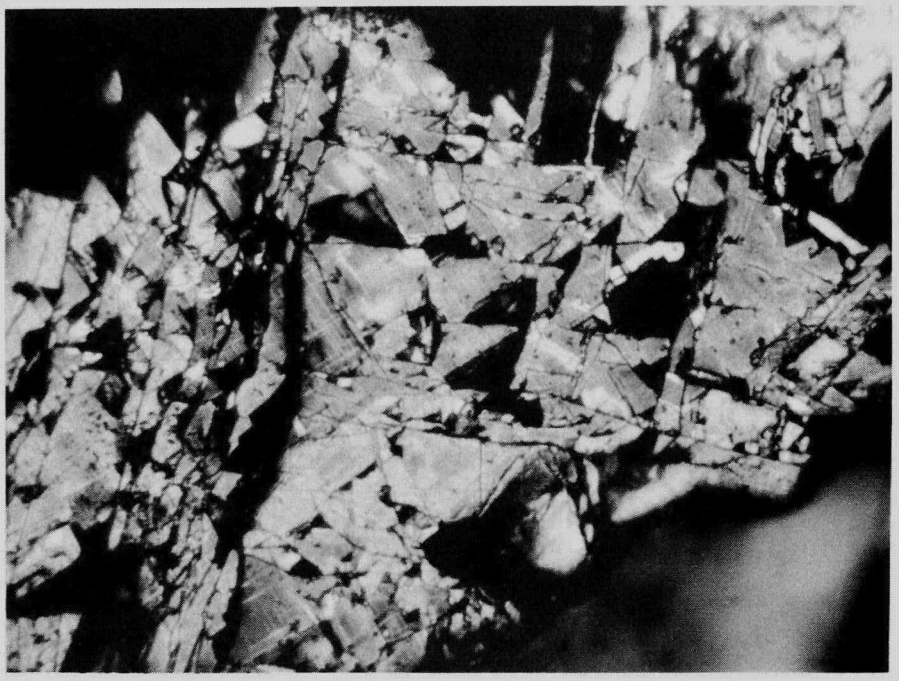
The two specimens were heat treated for 8 hr at 2000 F under vacuum in a quartz tube. After the heat treatment, it was found that the unirradiated specimen experienced no appreciable change; however, the irradiated specimen disintegrated into particles, as shown in Figure 8a. Figure 8b shows the microstructure of the fine particles. The UC structure is quite evident, as is the severe disintegration of the specimen during the heat treatment. Prior to the opening of the quartz tube, it was first punctured and the fission gas was collected. The amount of fission gas was determined by measuring the amount of krypton-85 present. A total of 2.04×10^{-6} cm^3 of krypton-85 was released during heat treatment. Based on the weight of this specimen prior to heat treatment, it was calculated that the total amount of krypton-85 that was produced in the specimen was 8.0×10^{-2} cm^3. The amount of krypton-85 released after irradiation was only 12.0×10^{-5} cm^3. It can be seen that the amounts of krypton-85 released after irradiation and after the heat treatment were negligible when compared with the calculated amount produced during irradiation. These data were based on average calculated values which included all of the UC contained in the capsule.

Burnup of the specimens from the BRR capsules and Capsules BMI-23-1 and BMI-23-2 were estimated from dosimeter analyses. In the dosimeter analyses, a dosimeter wire was taken from the center 1/2-in. spacer in Capsules BMI-23-1 and BMI-23-2 and three sections were cut from the 12-in. dosimeter wire which was wrapped around each capsule basket. The analyses for the top and center sections of the 12-in. dosimeter wire were averaged to calculate the burnup for the bottom specimens. For these analyses, the Brad Lewis factor[3] was used in estimating the



6X HC2663

a. UC Specimen After Heat Treatment



500X HC3117

b. Typical Microstructure of the Particles of the Heat-Treated UC Specimen

FIGURE 8. EFFECT OF 8-HR HEAT TREATMENT IN VACUUM AT 2000 F ON IRRADIATED UC FROM CAPSULE BMI-23-2

effective thermal-neutron flux in the specimens. This method was used with a reasonable amount of confidence, since at the time of the postirradiation examination of these capsules Battelle experience indicated that this method for determining burnup data was as reliable as any other method and appeared to be relatively consistent.

During the postirradiation examination of Capsules BMI-23-3, BMI-23-5, and BMI-23-6, burnups were determined by three methods: (1) dosimetry data, (2) radiochemical analyses by the cesium-137 techniques, and (3) a mass isotopic analysis. These data are shown in Table 3. It is estimated that the uncertainty in the radiochemical burnup data is about 25 per cent, and in the burnup level based on the isotopic analyses it is about 12 per cent. With these uncertainties, the data can be considered to be in good relative agreement.

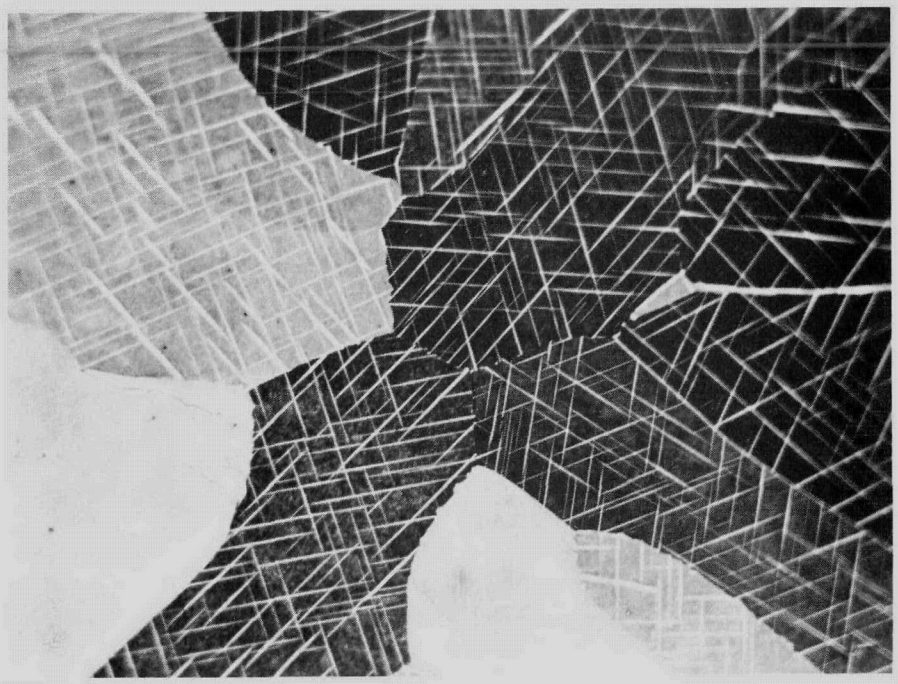
In an effort to further establish reliable burnup data, a specimen of NaK coolant from Capsules BMI-23-1, BMI-23-2, BMI-23-3, and BMI-23-5 was analyzed for cesium-137 content to determine if perhaps the radiochemical analyses on the UC specimens themselves were low because of diffusion and subsequent loss of cesium-137 during the irradiations. These analyses indicated only a negligible amount of cesium-137 in each NaK specimen.

The average surface heat fluxes are also tabulated in Table 3. The heat fluxes are based on dosimetry data except for the capsules for which isotopic burnups were determined. The average surface heat fluxes for the specimens vary from about 600,000 to about 940,000 Btu/(ft^2)(hr). The maximum heat fluxes obtained during the irradiations were probably higher.

A metallographic examination was made on several specimens from the BRR capsule and also on the top and bottom specimen from each MTR capsule. The metallographic specimens were mounted in cold-setting plastic, ground through 600-grit paper, and then polished and etched. The specimens were swab etched with a solution of equal parts of nitric acid, acetic acid and water. Figures 9 through 16 show the typical microstructures of the unirradiated and the irradiated specimens from all of the test capsules. The matrix phase in all of the photographs is stoichiometric uranium monocarbide, and, where shown, the platelike structure is UC_2.

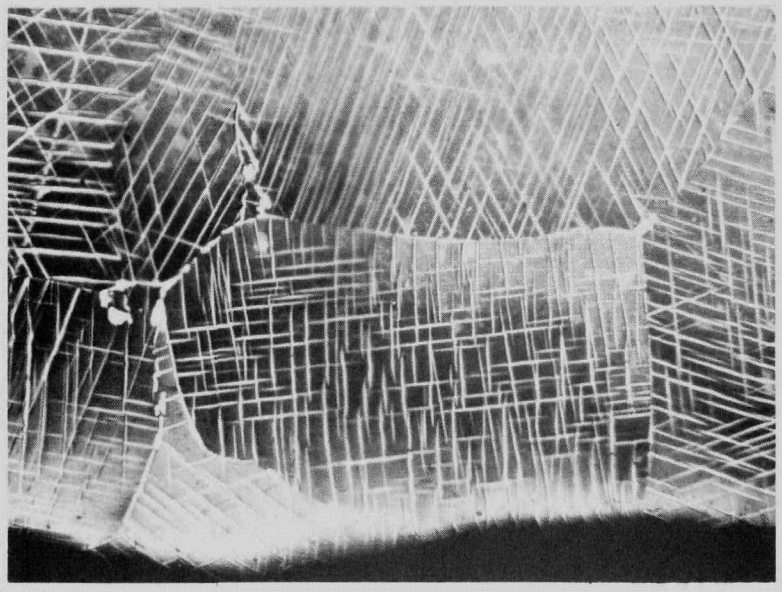
Figures 9 and 10 show the microstructures of the specimens from the BRR capsule and Capsule BMI-23-1. As can be seen, the material appeared to be unchanged after irradiation. However, Figures 11, 12, and 13, which illustrate specimens from Capsules BMI-23-2 and BMI-23-3 show that the UC_2 structure has apparently disappeared from the matrix phase in the irradiated specimens. In the top specimens of BMI-23-3, the UC_2 structure completely disappeared. The bottom specimens showed a gradual disappearance of the UC_2 structure from the edge of the specimen, where it was most prominent, to the center, where it completely disappeared. The remaining structure clearly resembled the 4.8 w/o carbon stoichiometric composition.

The mechanism of this apparent change in the microstructure of these 5 w/o carbon specimens is not fully understood. Perhaps the UC_2 is actually put into solution in the UC matrix as a result of neutron bombardment. Further investigation of this phenomenon is needed.



250X RM10615

a. Center of Specimen Before Irradiation



250X HC2068

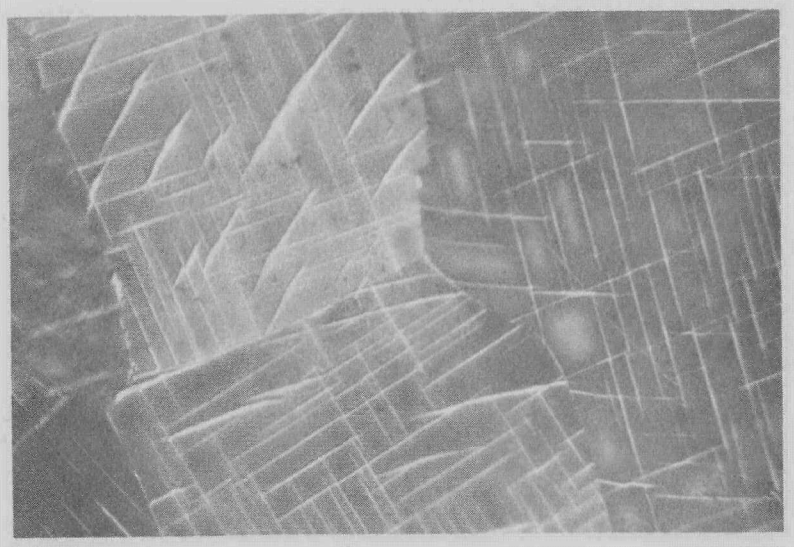
b. Edge of Specimen After Irradiation

FIGURE 9. NOMINAL URANIUM-5 w/o CARBON SPECIMEN FROM BRR CAPSULE BEFORE AND AFTER IRRADIATION

Dosimetry data indicate that the burnup was about 400 MWD/T of uranium.

250X RM10452

a. Preirradiation

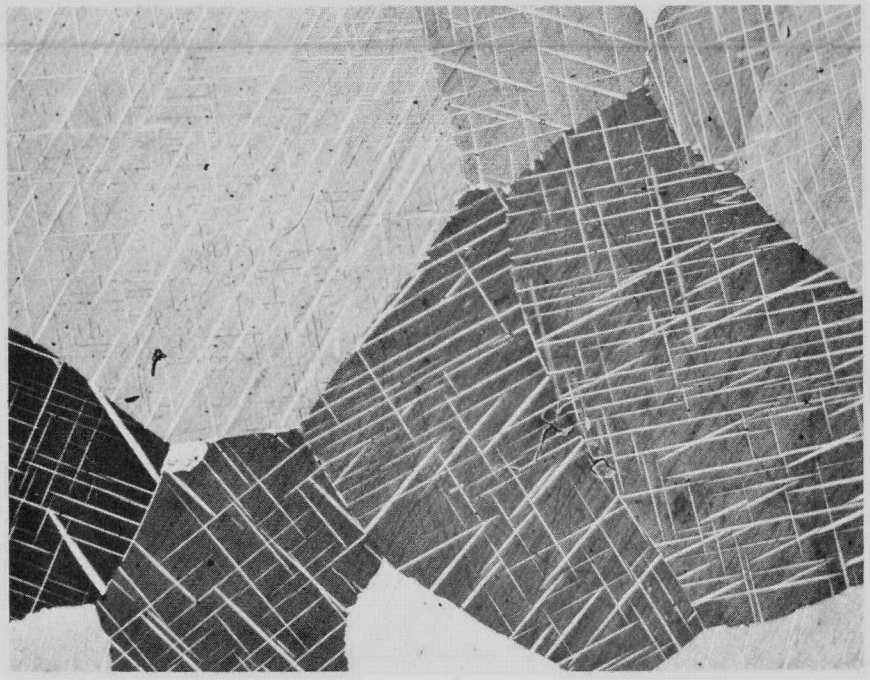


250X HC2066

b. Postirradiation

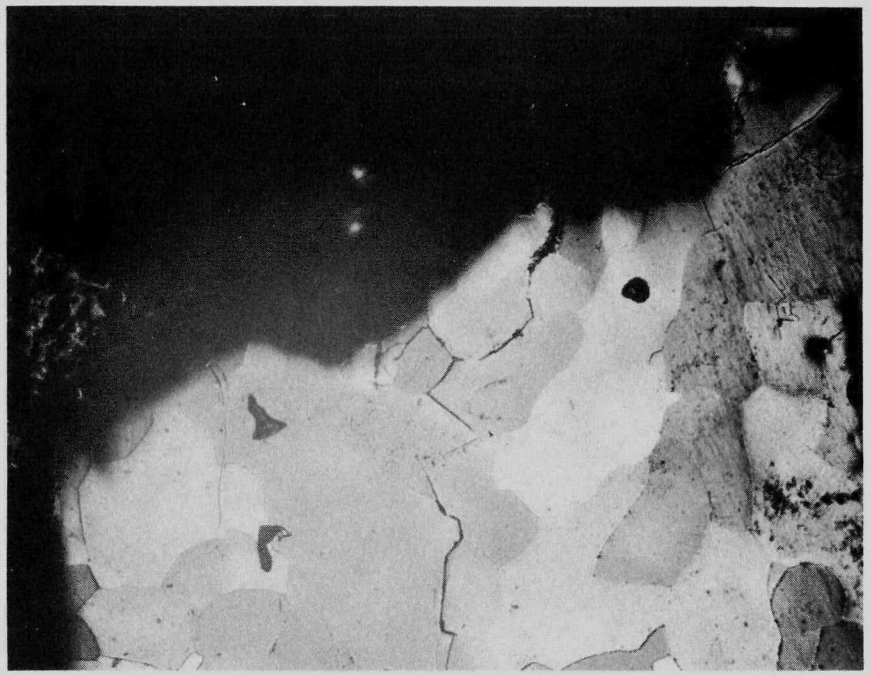
FIGURE 10. CENTER OF TOP SPECIMEN IN CAPSULE BMI-23-1 BEFORE AND AFTER IRRADIATION

Dosimetry data indicate that the burnup was about 1420 MWD/T of uranium for this specimen. The analyzed composition was uranium-5.2 w/o carbon.



250X RM10942

a. Preirradiation

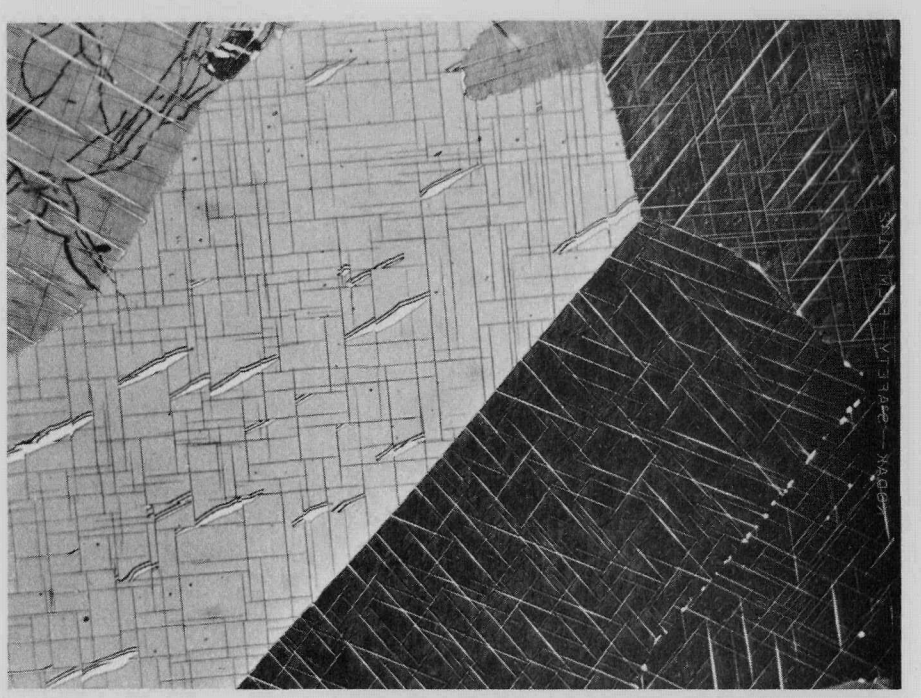


250X HC2621

b. Postirradiation

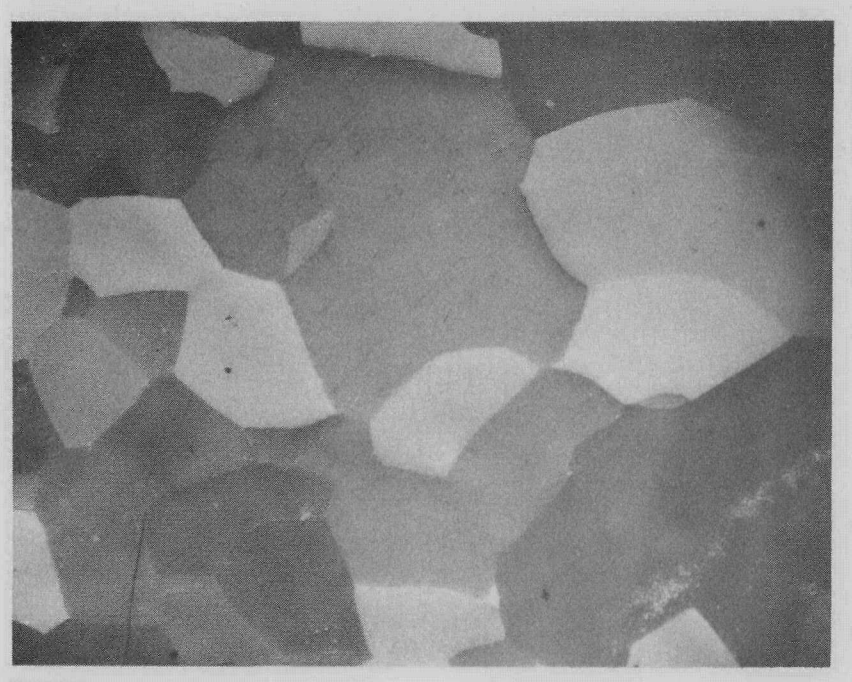
FIGURE 11. CENTER OF BOTTOM SPECIMEN FROM CAPSULE BMI-23-2 BEFORE AND AFTER IRRADIATION

Dosimetry data indicate that the burnup was about 7800 MWD/T of uranium for this specimen. The analyzed composition was uranium-5.1 w/o carbon.



250X RM10745

a. Preirradiation



250X HC4190

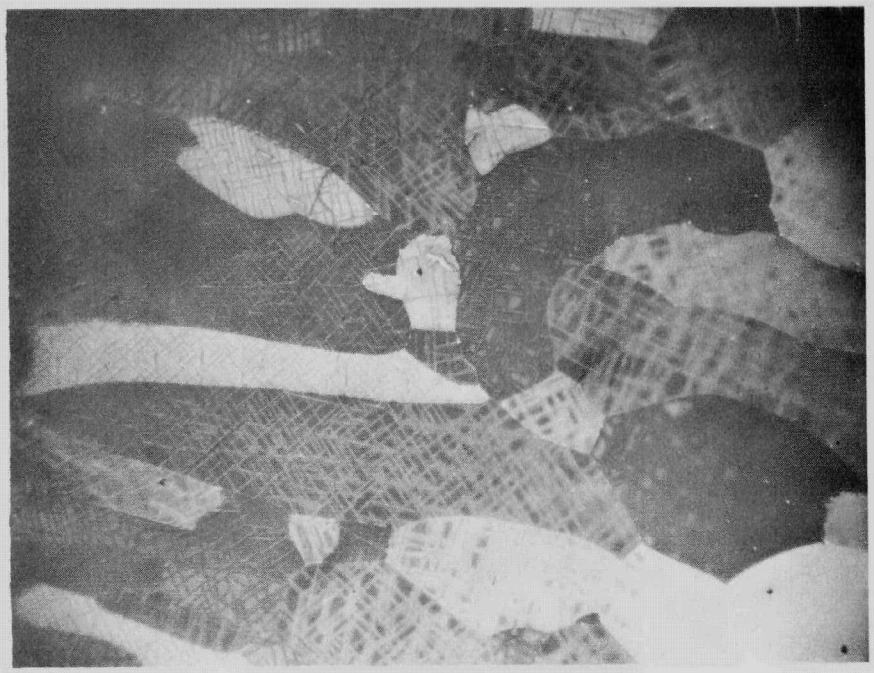
b. Postirradiation

FIGURE 12. MICROSTRUCTURE AT CENTER OF SPECIMEN 29, THE BOTTOM SPECIMEN FROM CAPSULE BMI-23-3, BEFORE AND AFTER IRRADIATION

Dosimetry data indicate that the burnup was about 12,800 MWD/T of uranium. The analyzed composition was 5.0 w/o carbon.

100X HC4187

a. Depleted Areas in Center and Areas With UC_2 Toward Edge of Specimen

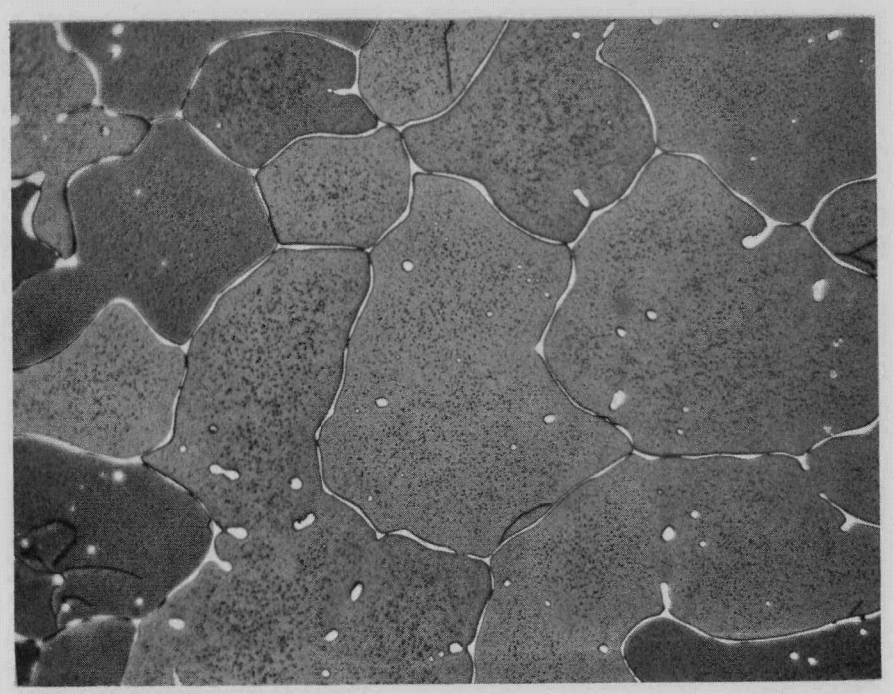


100X HC4189

b. UC_2-Containing Areas Near Edge of Specimen

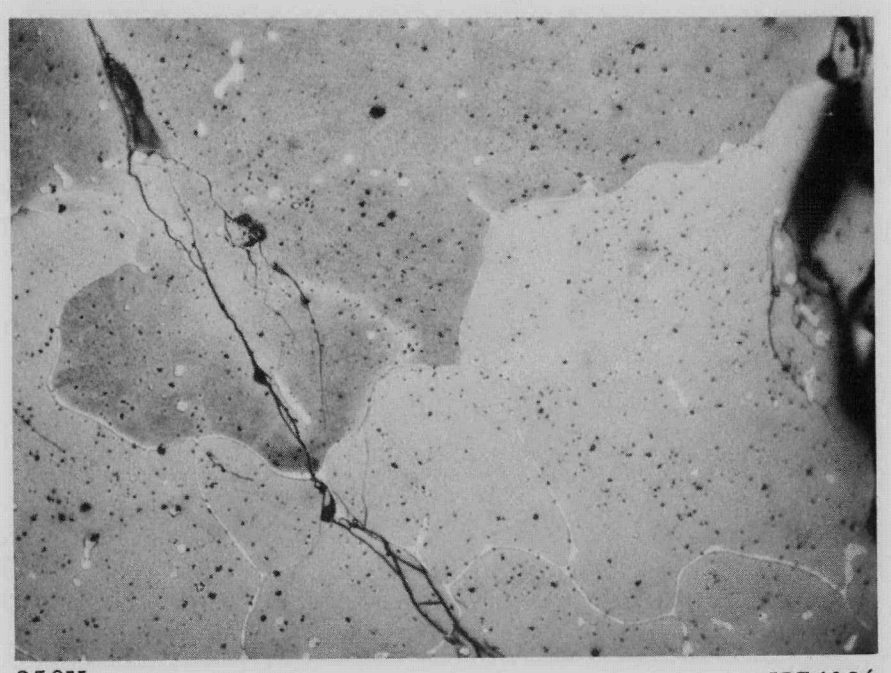
FIGURE 13. DISAPPEARANCE OF UC_2 STRUCTURE WITH DISTANCE FROM SURFACE IN BOTTOM SPECIMEN IN CAPSULE BMI-23-3

Dosimetry data indicate that the burnup was about 12,800 MWD/T of uranium. The analyzed composition was 5.0 w/o carbon.



250X RM1193

a. Preirradiation

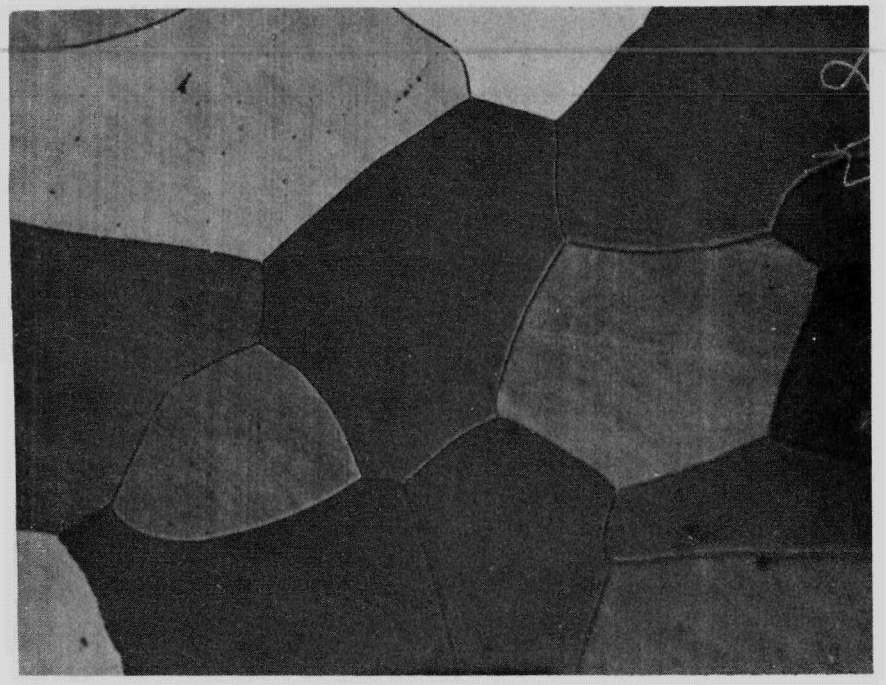


250X HC4196

b. Postirradiation

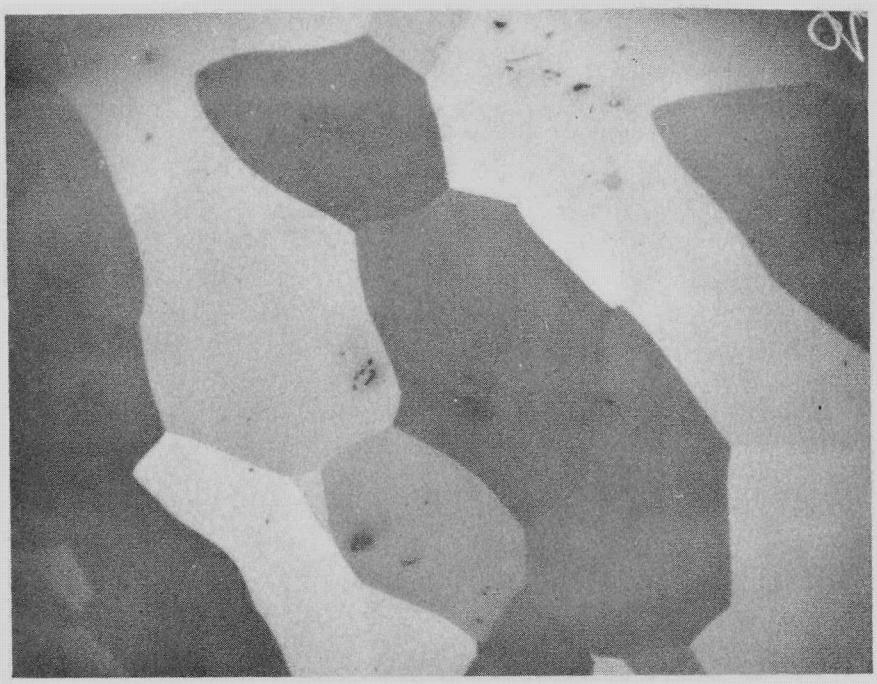
FIGURE 14. TYPICAL MICROSTRUCTURE BEFORE AND AFTER IRRADIATION OF SPECIMEN 50B FROM CAPSULE BMI-23-5

Dosimetry data indicate that the burnup was about 6400 MWD/T of uranium. The analyzed composition was 4.7 w/o carbon.



250X RM11149

a. Preirradiation

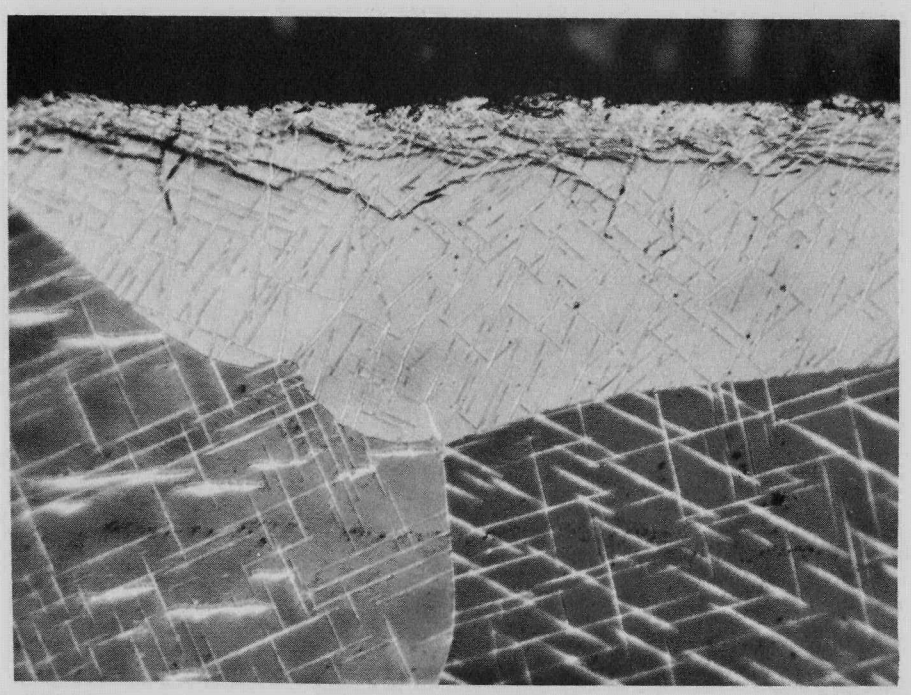


250X HC4385

b. Postirradiation

FIGURE 15. MICROSTRUCTURE OF TOP SPECIMEN FROM CAPSULE BMI-23-6 BEFORE AND AFTER IRRADIATION

Dosimetry data indicate that the burnup was about 6500 MWD/T of uranium. The analyzed composition was 4.8 w/o carbon.



250X HC2615

FIGURE 16. MICROSTRUCTURE OF SPECIMEN FROM CAPSULE BMI-23-2 SHOWING SURFACE DAMAGE

Figure 14 shows the typical microstructure of the nominal 4.6 w/o carbon bottom specimen from Capsule BMI-23-5 before and after irradiation. Alpha uranium can be seen in the grain boundaries and also within the grains. No apparent change because of irradiation was observed. Figure 15 shows the typical microstructure of the 4.8 w/o specimen from Capsule BMI-23-6. The pre- and postirradiation microphotographs show identical structures.

In Figure 16 the corrosive effect typical of the NaK is evident at the surface of the specimen. Apparently, this is a reaction of the UC with the Na_2O in the triple-distilled NaK which was used in the capsules. Thermodynamically, it can be shown that such a reaction is possible at room temperature and becomes more likely at increasing temperatures. The NaK in the test capsules contained about 50 ppm oxygen and it was felt that this is enough to cause the attack on the surface of the specimens. Perhaps this is the area which tends to spall off during the irradiation. This surface attack was evident on all of the specimens examined.

CONCLUSIONS

The results of the examinations on the specimens described above are encouraging for the use of UC as a fuel material for sodium-, organic-, or gas-cooled reactors. The density and dimension changes experienced at burnups as high as about 13,750 MWD/T of uranium are negligible, and, if microcracks and surface damage are discounted, the radiation stability of UC can be considered excellent.

The amount of cracking could probably be reduced if the UC could be divorced from actual contact with NaK or if it could be irradiated in ultrahigh-purity NaK. The cracking appears to be somewhat related to burnup or the thermal cycles correlated with the burnup. Some cracking is no doubt due to thermal stresses caused by the thermal gradients across the specimen during the irradiations.

Although the UC_2 structure tends to disappear during irradiation of the 5 w/o carbon specimens, the change in microstructure does not appear to have any deleterious effects on the specimens. The behavior of the 4.6, 4.8, and 5 w/o carbon specimens was comparable at relative burnup levels.

The average surface heat-generation rates obtained during these irradiations are encouraging. They ranged from about 600,000 to about 940,000 Btu/(ft^2)(hr) for the MTR capsule specimens. Instantaneous heat-generation rates probably reached values in excess of 1,000,000 Btu/(ft^2)(hr).

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REFERENCES

(1) Secrest, A. C., Foster, E. L., and Dickerson, R. F., "Preparation and Properties of Uranium Monocarbide Castings", BMI-1309 (January 2, 1959).

(2) Price, R. B., Stahl, D., Stang, J. H., and Simons, E. M., "Irradiation-Capsule Study of Uranium Monocarbide", BMI-1425 (March 2, 1960).

(3) Lewis, W. B., "Flux Perturbations by Material Under Irradiation", Nucleonics, 13 (10), 82 (October, 1955).

AWH:FAR/ims