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ORNL-3403
UC-25 -- Metals, Ceramics, and Materials
TID-4500 (18th ed., Rev.)

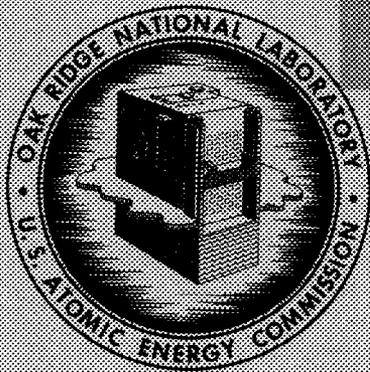
THE EFFECT OF IRRADIATION ON THE HYDROLYSIS
OF URANIUM CARBIDES. I. PREPARATION OF
URANIUM MONOCARBIDE PELLETS FOR IRRADIATION

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Printed in USA. Price: \$0.75 Available from the
Office of Technical Services
U. S. Department of Commerce
Washington 25, D. C.

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ORNL-3403

Contract No. W-7405-eng-26
CHEMICAL TECHNOLOGY DIVISION
Chemical Development Section B

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I. PREPARATION OF URANIUM MONOCARBIDE PELLETS FOR IRRADIATION

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DATE ISSUED

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ABSTRACT

The effect of irradiation on the hydrolytic behavior of uranium monocarbide as related to the aqueous chemical processing of these reactor fuels is being investigated. This report describes the fabrication, evaluation, and encapsulation of the UC in niobium prior to irradiation.

Uranium monocarbide buttons were prepared by arc-melting 1.51%-enriched uranium metal with the stoichiometric quantity of spectroscopic-grade carbon. The buttons were then remelted and drop-cast into graphite thimbles $3/8$ in. in diameter, and 3 in. long. The cylindrical castings were surface ground and cut into 0.17 in.-thick pellets, which were then polished.

Preirradiation evaluation included metallography, chemical analysis, and hydrolysis studies. Microstructures of the final pellets indicated that the monocarbide was nearly stoichiometric; however, some α -uranium was present along with carbon contamination from the graphite thimbles. The use of tungsten-tipped electrodes in the arc-melting operation introduced about 0.3% of tungsten impurity. The composition of the pellets, as estimated from hydrolysis studies and chemical analyses was 92% UC, 3.5% UC₂, 4.0% uranium metal, and 0.3% WC.

The UC pellets were encapsulated in niobium and subsequently shrunk-fit into a stainless steel cladding. The capsules were sealed by heliarc welding in a helium atmosphere chamber.

1. INTRODUCTION

A program has been initiated to determine the effects of irradiation on the chemical reactions of interest in the aqueous processing of spent uranium carbide reactor fuels. Fuels under investigation include both massive carbide slugs clad in stainless steel or zirconium, and carbide particles dispersed throughout a graphite matrix.¹ The metal-clad elements probably will require mechanical chopping prior to either hydrolysis followed by nitric acid dissolution of the residue or direct nitric acid dissolution of the carbide core in preparation for uranium recovery by solvent extraction. Nitric acid leaching is also applicable to uncoated graphite-base fuels. Therefore, the reactions of particular interest to fuel processing are: hydrolysis in nonoxidizing aqueous reagents such as water, HCl, and H₂SO₄, which yields hydrocarbons^{2,3}, and direct dissolution in nitric acid, which results in the production of some soluble organic acids.

If the hydrolysis of uranium monocarbide proceeds through a free-radical mechanism, irradiated specimens could yield significant quantities of nonvolatile carbon-containing polymers even though unirradiated UC yields principally methane.³ These polymers, in turn, after their dissolution in nitric acid along with the uranium oxide residue might adversely affect solvent extraction behavior and/or, if unsaturated, form potentially explosive nitro-organics.

Since most uranium monocarbide preparations contain UC₂ as an impurity which yields higher hydrocarbons upon hydrolysis, extensive evaluation of unirradiated specimens from the UC batch intended for irradiation is required to distinguish between impurity and radiation effects. This initial phase of the program, which is summarized in this report, involved the fabrication, encapsulation, and preirradiation chemical and metallographic evaluation of uranium monocarbide pellets. The resulting capsules will shortly be irradiated to several burnup levels.

The authors thank A. D. Horton of the ORNL Analytical Chemistry Division for analysis of the gaseous hydrolysis products, L. Queener

of the Metals and Ceramics Division for the arc-melting and casting of the uranium carbide, and P. P. Haydon, Chemical Technology Division, for supervising the cutting and polishing of the pellets. Other chemical analyses and the metallographic examinations were provided by the groups of W. R. Laing, Analytical Chemistry Division, and R. J. Gray, Metals and Ceramics Division.

2. PREPARATION OF CYLINDRICAL CASTINGS OF URANIUM MONOCARBIDE

Eleven cylinders of 1.51%-enriched uranium monocarbide were arc-cast for the irradiation studies as nominally 3/8-in.-diameter and 3-in.-long rods. The arc-melting and drop-casting method was selected for preparing the carbide because of its demonstrated ability to produce high-density material of controlled composition. The UC was prepared by alloying purified uranium derby metal with spectroscopic-grade carbon in an arc-button furnace (Fig. 1) that had a water-cooled tungsten-tipped electrode and a water-cooled copper hearth. The tungsten-tipped electrodes permitted close control of carbon composition, although the final castings contained some tungsten as an impurity. The UC buttons were remelted and cast into graphite thimbles, 0.394 in. in inner diameter, by a drop casting-method similar to that originally developed at Battelle Memorial Institute⁴. The procedure used here was developed under the direction of D. T. Bourgette⁵. Graphite mold liners were used to decrease the cooling rate of the castings in order to decrease thermal-stress cracking; this resulted in the pickup of carbon which appeared as UC₂ precipitated along the surface.

2.1 Purification of Starting Metal

The starting metal, 1200 g of 1.51%-enriched uranium derby stock, which contained 19 ppm of carbon, was pickled in 13 N HNO₃ to remove surface scale and then was divided into two approximately equal portions. The chunks of derby metal in each portion were charged to a slab cavity in the water-cooled copper hearth. The furnace was sealed and evacuated to a pressure less than 2×10^{-5} torr. High-purity argon was introduced and gettered by melting a zirconium button. The uranium was melted several times with an arc power of 450 to 500 amp at 25 to 30 volts, the

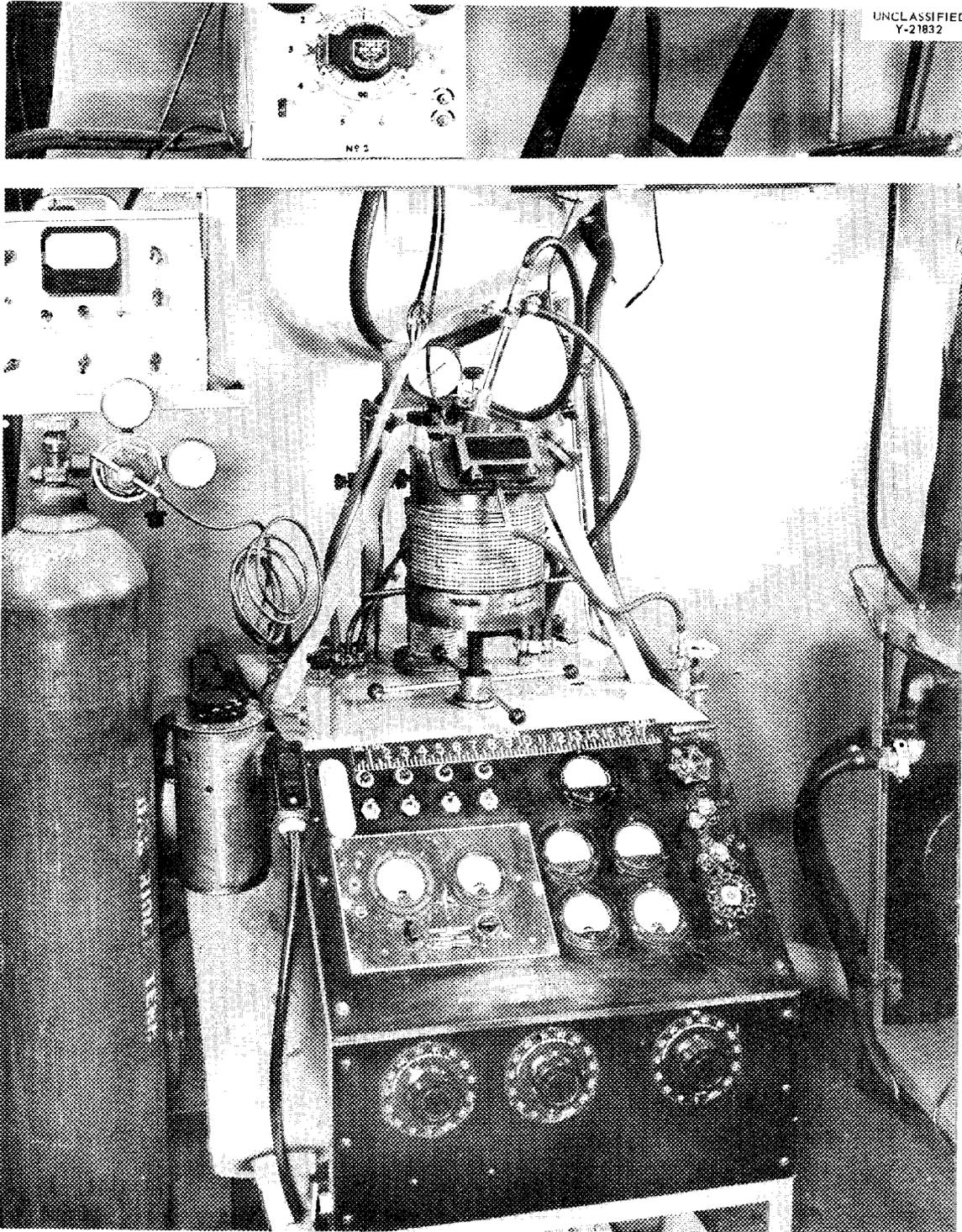


Fig. 1. Arc button furnace used for casting UC.

uranium slab being turned over with the electrode between melts. The argon pressure during melting was 20 to 25 psia. The slabs were then pickled in 13 N HNO_3 to remove most of the surface scale and then were remelted. Less surface scale was observed after the second melt, indicating that most of the removable impurities had been eliminated during the first melting. The slabs were hot rolled from a salt bath at 625°C into 0.022-in.-thick strips; then, the strips were pickled in 13 N HNO_3 and sheared into small pieces.

2.2 Preparation of Uranium Carbide Buttons

Weighed quantities (± 1 mg) of the purified uranium metal strips and small pieces of spectroscopic-grade carbon in the ratio for stoichiometric UC (4.80 wt % carbon) were alloyed by arc-melting as buttons (about 150 g). No adjustment for carbon in the uranium metal was necessary since it contained only 19 ppm. Starting buttons for the last four castings were prepared by remelting the skulls from the first seven.

Four uranium-carbon charges and a zirconium getter button were loaded into hemispherical cavities in the water-cooled copper hearth of the arc-button furnace. The furnace was evacuated to 1 to 2×10^{-5} torr and then high-purity argon was admitted to establish a pressure of about 11 psia. The zirconium getter button was melted first and then the uranium-carbon charges. The first melt of each charge was continued until particles of carbon were no longer visible. Each UC button was then melted an additional six or seven times to complete the reaction of the uranium and carbon and to achieve homogeneity. The argon pressure rose to about 30 psia during the melting with a 500-amp 30-v arc.

2.3 Drop Casting of the UC Cylinders

A drop-casting assembly, consisting of a copper block mold with a copper insert and a graphite thimble (Fig. 2 a, b, and c) was installed in the arc-button furnace. The basic copper mold had been used in a previous development study⁵. It was modified by machining the bottom of the block mold and flame-spraying the sides with Al_2O_3 to further decrease heat loss, and reduce thermal-stress cracking of the castings

(Fig. 2b). In operation, the flanged portion of the graphite thimble rested snugly in a flat recess in the copper, but the thimble sidewalls were not in contact with copper (Fig. 2 a and 2 c).

The UC button was placed over the graphite thimble; the furnace was evacuated and back-filled with argon to about 11 psia and the arc struck on the tungsten peg on the upper rim of the copper mold. The casting operation was conducted according to the following schedule:

1. The arc was played on the copper mold in a circular motion for 60 sec at low power (about 200 amp) to preheat the mold.
2. The insert was then heated for 30 sec. During the last 10 sec of this interval, the arc was directed intermittently on the edge of the UC button.
3. The arc was then directed on the UC button itself, and the power was gradually increased from the initial 200 amp to about 600 amp until the button, except for a thin layer at the bottom, was molten.
4. The power was then increased until the bottom of the button melted through and the molten UC dropped into the thimble. With the first four castings this final power increase (up to almost 800 amp) was applied rapidly. For the rest of the castings, the terminal power increase was more gradual, and the maximum power did not exceed 700 amp. The surfaces of these latter castings were brighter than those of the first.
5. After the UC had dropped into the thimble, the vacuum valve was opened to remove the argon, and thereby decrease the rate of heat loss from the thimble in an attempt to decrease the thermal stresses on the casting.

The total elapsed time from arc initiation to the moment the molten UC dropped varied from 2 min 56 sec to 3 min 51 sec for castings 2 to 7, and from 3 min 10 sec to 3 min 15 sec for the other five castings.

The as-cast appearance and measurements of the diameters of the eleven castings are illustrated in Fig. 3 a and b.

The castings were radiographed by the Nondestructive Testing Laboratory, Y-12 Technical Division, with a million-volt x-ray unit. Casting 1 appeared to have a large cavity, while numbers 6 and 8 had small cavities. The other castings appeared sound.

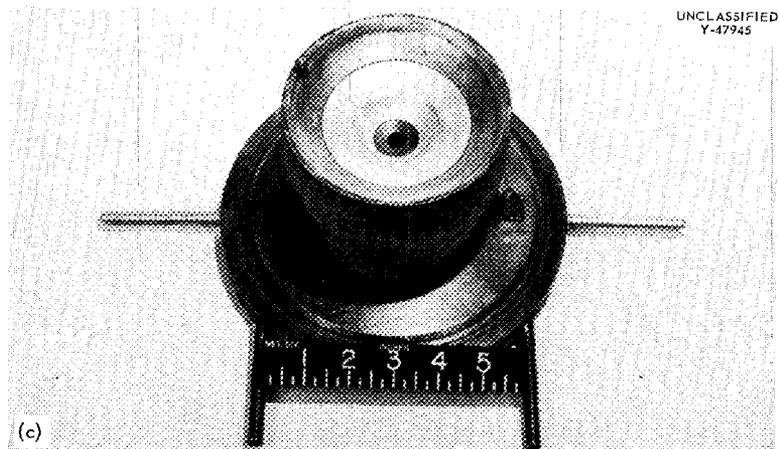
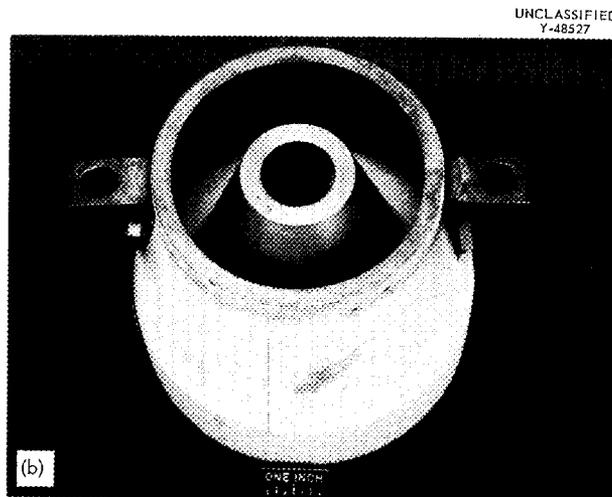
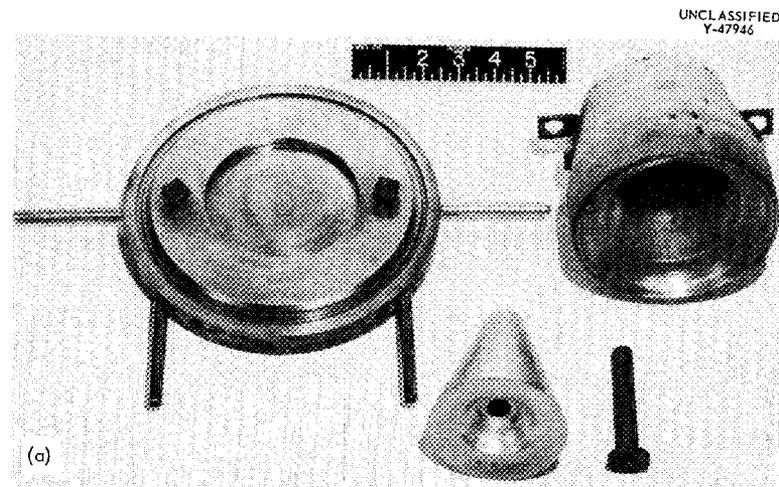


Fig. 2. Copper block mold with copper insert, graphite thimble, and water-cooled base plate for drop-casting uranium monocarbide cylinders; (a) top view, (b) bottom view of block mold showing where metal was machined away to reduce heat loss, and (c) assembled mold.

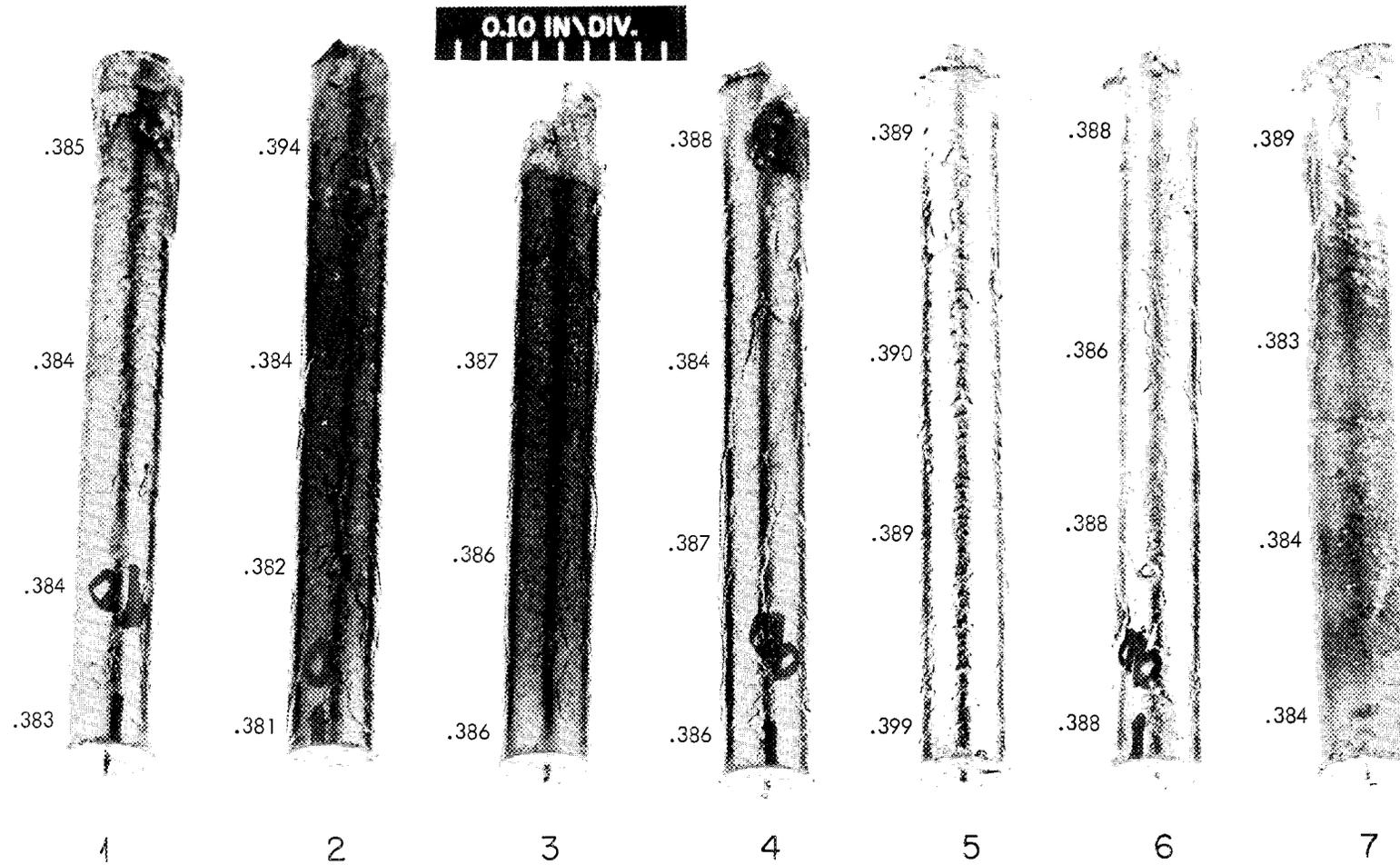
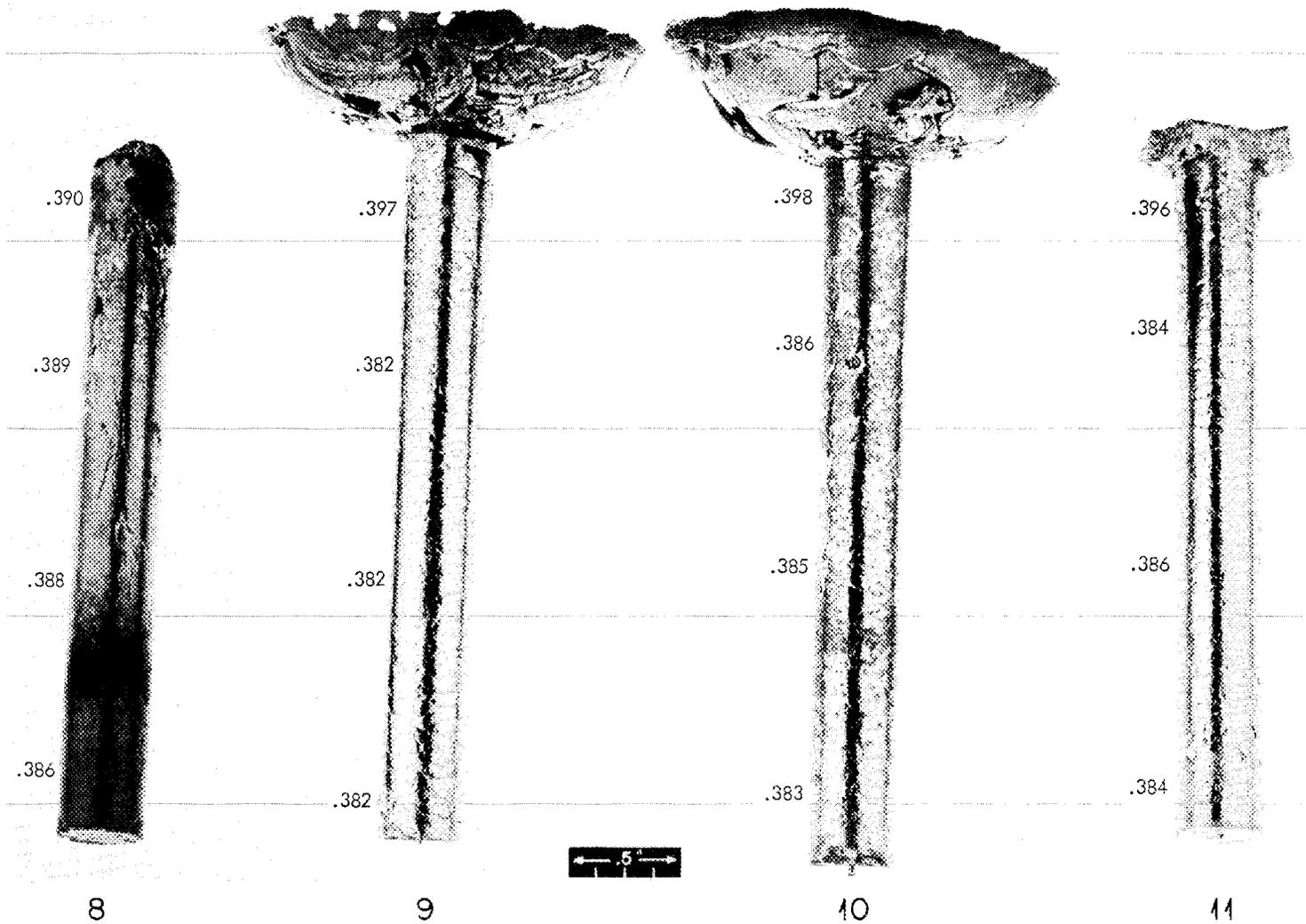


Fig. 3a. Photograph of casting numbers 1 through 7.



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Fig. 3b. Photograph of casting numbers 8 through 11.

3. CUTTING AND POLISHING

Four-gram pellets with a diameter of 0.38 to 0.39 in. and a thickness of 0.17 in. were made by grinding the cast rods (Sec 2) to a maximum diameter of 0.39 in. and then slicing.

The as-cast rods were stored overnight in a vacuum desiccator and transferred daily in a portable desiccator containing 5-A molecular sieve to the ORNL Special Materials machine shop. Rods were removed from the portable desiccator as needed, and the bottom ends were placed immediately in a standard machine chuck on an exhaust-hooded Bridgeport milling machine. A continuous stream of perchloroethylene was directed on the grinding, facing, or slicing operation at all times.

The rod surface was ground to the specified diameter by a 6-in.-diam diamond-impregnated wheel with a 1/2-in. face. The measurements listed below are ± 0.001 in.

<u>Rod No.</u>	<u>Ground to Max. Diam (in.)</u>	<u>Diam Range (in.)</u>
1	Not ground	0.382-0.387
2	0.385	0.381-0.385
3	Not ground	0.386-0.387
4	Not ground	0.381-0.388
5	0.385	----
6	Not ground	0.384-0.389
7	Not ground	0.382-0.389
8	0.384	---
9	0.386	0.379-0.386
10	0.388	0.383-0.388
11	0.387	0.383-0.387

The end of the rod was then squared with a 3-in. cup wheel. Slices were cut by slowly hand-feeding a 6-in.-diam diamond-impregnated wheel with a 40-mil face into the carbide rod. When cutting was almost complete, the slice broke from the rod, leaving a nodule in the center. This nodule was removed and the surface roughly polished by holding the slice against the rotating cup wheel. The finished slices from each rod were placed in

numerical order in a sample bottle with a 0.55 in.-ID glass liner to prevent shifting of the slices, and returned to the desiccator.

4. PREIRRADIATION EXAMINATION

4.1 Summary of Findings

The final pellets were relatively high-purity uranium monocarbide, according to chemical and metallographic examination, with visible, small star-shaped cracks. Small quantities of α -uranium and uranium dicarbide were detected metallographically. Hydrolysis studies indicated a composition of about 92% UC, 3.5% UC₂, 4% U metal, and 0.3% WC. The procedures used for the chemical analyses and the hydrolysis studies were reported earlier³.

4.2 Results of Visual Examination

Small, star-shaped cracks radiating from the center of the slices, which were numbered consecutively starting from the skull end, were present in all the rods. This effect was most pronounced near the top of the rod and in the vicinity of a cavity. Fewer cracks were observed in the second batch (rods 8 to 11) than in the first. Cavities were found in rods 1, 3, 6, 7, 8, and 9. In rod 1, slices 3, 4, 8, 9, 10, and 11 had holes; in rod 3, slice 8 had a pin hole. There were small holes in rod 6, slice 11; rod 7, slice 10; rod 8, slice 3; and rod 9, slice 11. Rod 8 was the most crack-free, followed by rods 9 and 10. Rod 5 was the best in the first batch. Ninety-nine good slices were obtained from rods 2 to 11.

4.3 Results of Elemental Analyses

The pellet composition was close to theoretical: 95.2% U and 4.80% C. Rod 1 was sampled at the top, middle, and bottom. The sum of the uranium and tungsten varied from 95.26 to 95.33%, while the total carbon varied from 4.72 to 4.74% (Table 1). The free carbon was 0.03%, and oxygen 0.02 to 0.05%. Rods 2 to 10 were sampled about 1/4 the distance from the top or bottom on an alternate basis. The sum of the uranium and tungsten varied from 95.10 to 95.23%, while the total carbon varied from 4.69 to 4.78%. Free carbon was less than 0.02%, and oxygen less

than 0.08%. Semiquantitative analyses for tungsten varied from 0.2 to 0.4%.

Table 1. Composition of UC Specimens
Theoretical for UC: 95.2% U; 4.80% C

Sample	Amount (wt %)		
	U + W ^a	Total C ^b	O
R-1-2 ^c	95.26	4.74	0.03
R-1-8	95.28	4.74	0.05
R-1-13	95.33	4.72	0.02
R-2-4	95.23	4.78	0.08
R-3-8	95.22	4.76	0.05
R-4-3	95.16	4.76	0.03
R-5-10	95.22	4.69	0.01
R-6-4	95.15	4.76	0.02
R-7-10	95.11	4.78	0.03
R-8-3	95.13	4.77	0.02
R-9-11	95.14	4.76	0.08
R-10-3	95.10	4.74	0.02
R-11-7	95.10	4.78	0.01

^a Semiquantitative analyses for W: 0.2 to 0.4%.

^b Free C: 0.02% except for rod 1, which was 0.03%.

^c R-1-2 = rod 1, 2nd slice from top.

4.4 Results of Metallographic Examination

The uranium monocarbide pellets were prepared for metallography by the techniques described by Gray *et al.*⁶ Examination showed relatively high-purity uranium monocarbide containing small amounts of α -uranium, predominantly in grain boundaries, and uranium dicarbide precipitated in a Widmanstätten pattern to a depth of approximately 10 mils from the surface that had been in contact with the graphite mold. Some areas containing fine needles of uranium dicarbide were found at greater depths from the surface.

Typical microstructures are shown in Figs. 4 to 16. Each slice examined had areas of high-purity uranium carbide (Figs. 5a and 10), regions where α -uranium was readily detected, particularly at higher magnifications (Figs. 4 to 6, 8, 12, and 14), and regions containing fine needles of uranium dicarbide (Figs. 7, 9, 11, 13, and 15).

4.5 Results of Hydrolysis Studies

Hydrolysis of the UC pellets in water at 80°C by the procedure described previously³ yielded a gelatinous, greenish-colored, tetravalent uranium compound and 90 ml (STP) of gas per gram of sample. The gas consisted of 88% methane, 9% hydrogen, and 3% C₂- to C₈- hydrocarbons (Table 2). Small amounts of ethylene, butene-1, and cis- and trans-butene-2 were present. After dissolution of the nonvolatile residue in 6 N HCl, 95 to 99% of the uranium was in the tetravalent state.

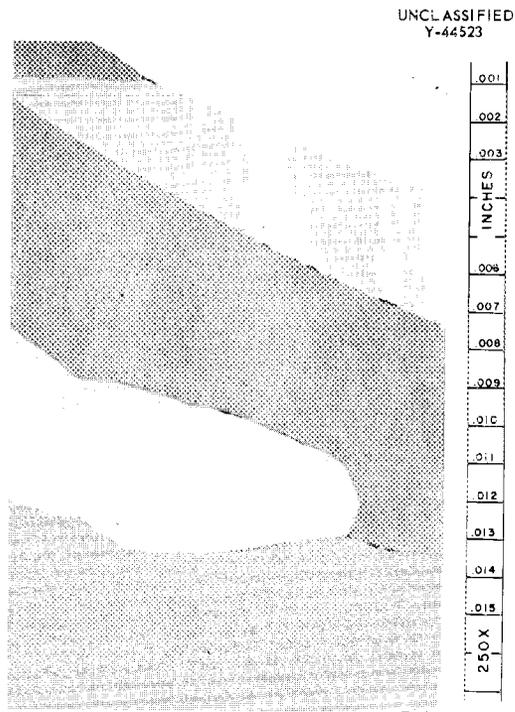
Of the 3.96 mg-atoms of carbon originally present in each gram of this carbide, hydrolysis at 80°C yielded an average of 3.56 mg-atoms as methane and 0.34 as volatile C₂- to C₈- hydrocarbons. The inert tungsten carbide accounted for 0.01 to 0.02 mg-atoms of carbon. The "missing" or unaccounted for carbon, about 0.06 mg-atom, probably resulted from the UC_{1.86} impurity, which is known to yield nongaseous products upon hydrolysis.

The average composition of the carbide based on the hydrolysis experiments was estimated as 92% UC, 3.5% UC₂, 4% U metal, and 0.2 to 0.4% WC, by comparison with the hydrolysis products from as-cast buttons containing 2 to 10 wt % C.⁷

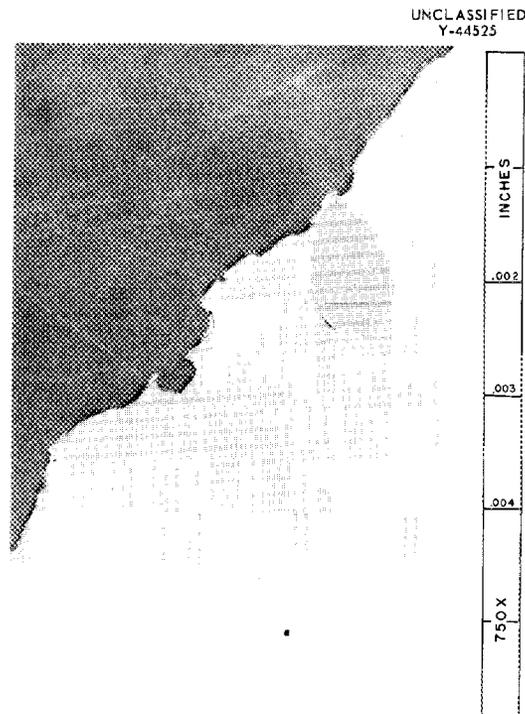
Except for the WC, the composition is similar to that of the arc-melted UC buttons which were not subjected to the drop-casting procedure³.

5. ENCAPSULATION OF THE URANIUM CARBIDE

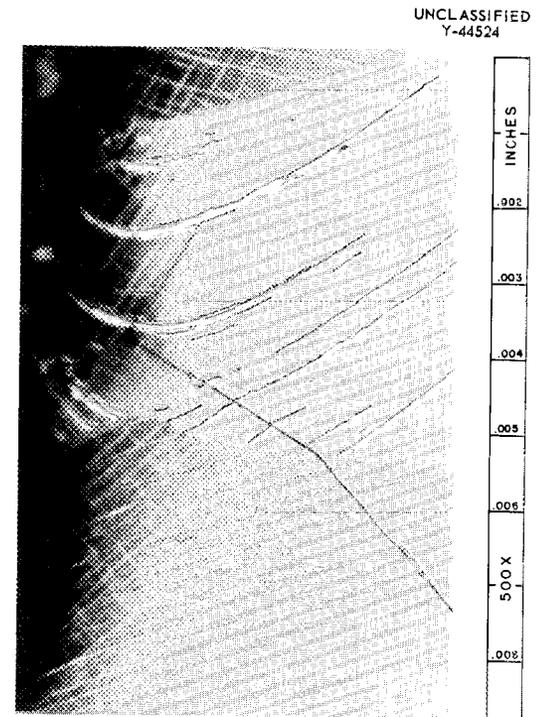
The irradiation capsule is shown in Fig. 17. Twelve UC slices are contained in a niobium tube to which are welded niobium end plugs. The niobium unit is shrunk-fit inside a type 304L stainless steel sleeve, which in turn is welded shut.



(a)



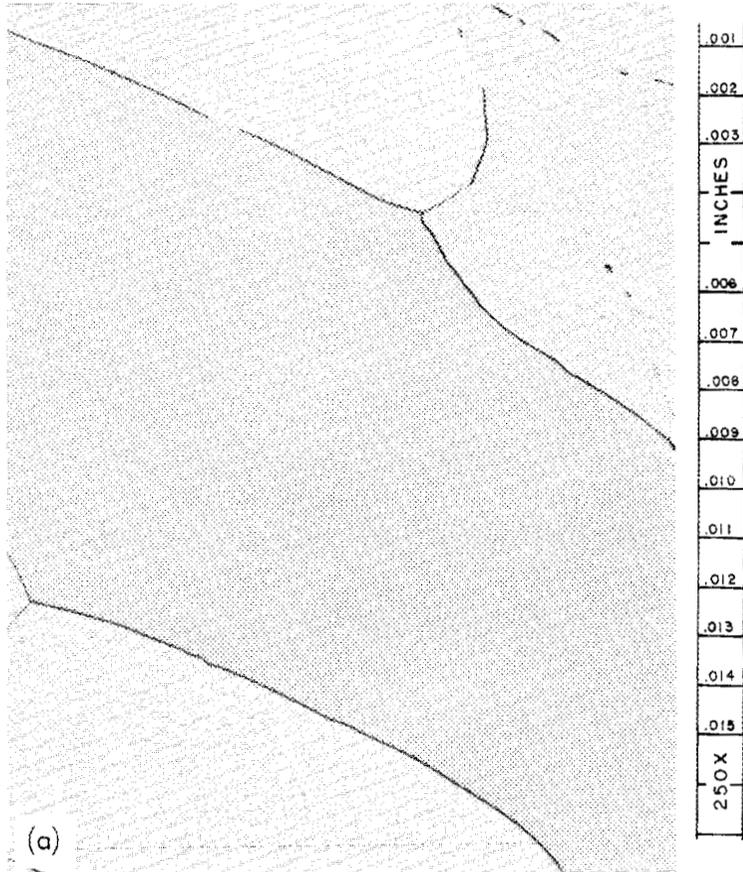
(b)



(c)

Fig. 4. Microstructure of rod 1, slice 2; near the center at a magnification of (a) 250 and (b) 750 diameters showing α -uranium at the grain boundaries, and (c) at the surface showing the UC₂ precipitate where the casting contacted the graphite mold. Reduced 22.5%.

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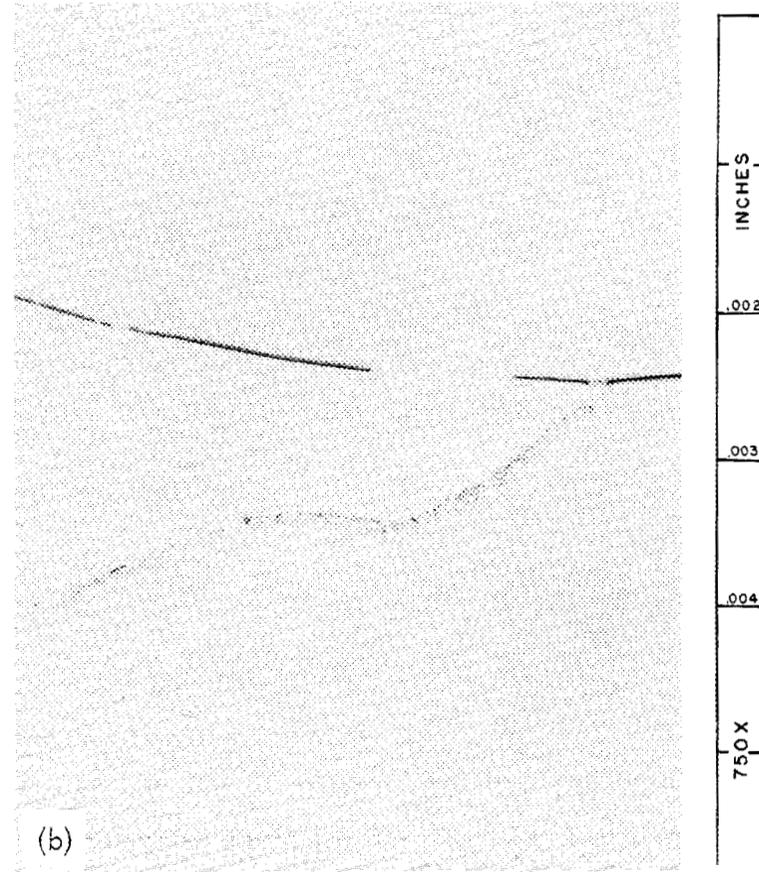
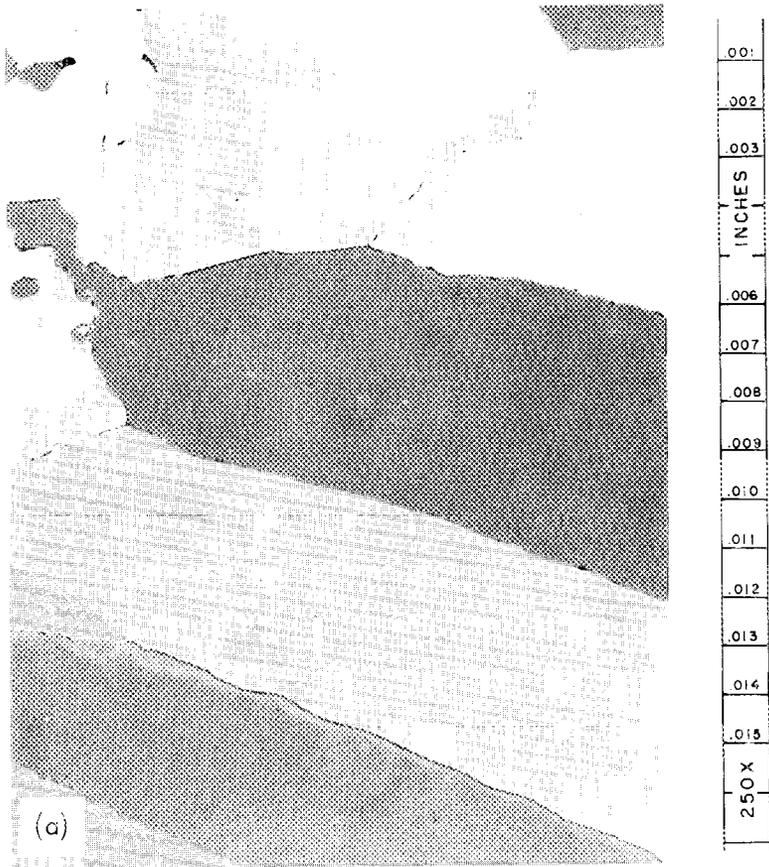


Fig. 5. Microstructure of the center of rod 1, slice 8 at a magnification of (a) 250 and (b) 750 diameters. View (b) shows α -uranium at the grain boundary.

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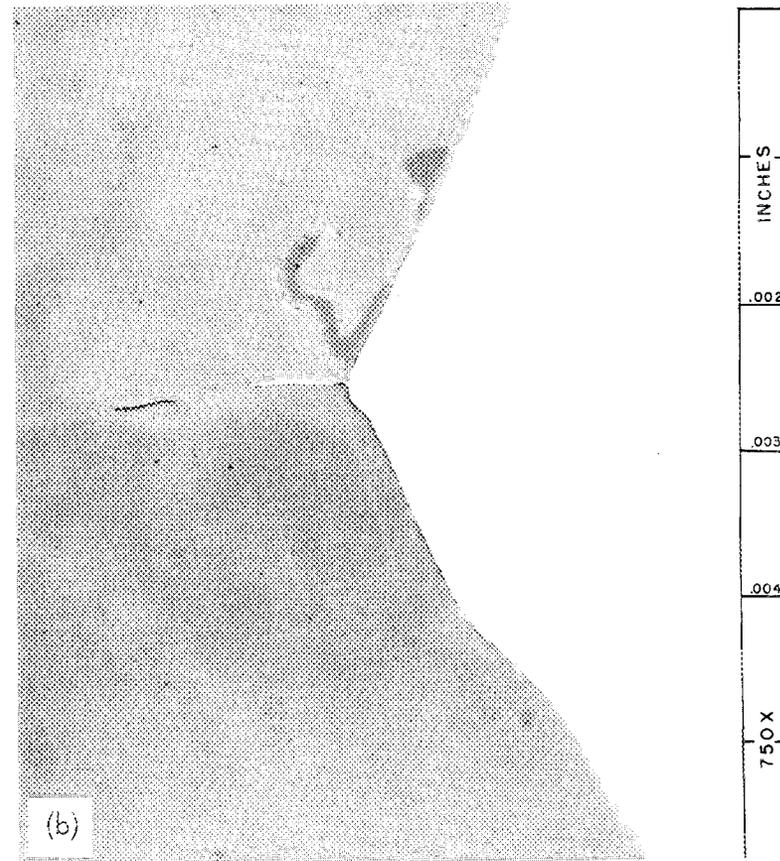


Fig. 6. Microstructure of the center of rod 1, slice 13, showing traces of α -uranium at the grain boundaries at a magnification of (a) 250 and (b) 750 diameters.

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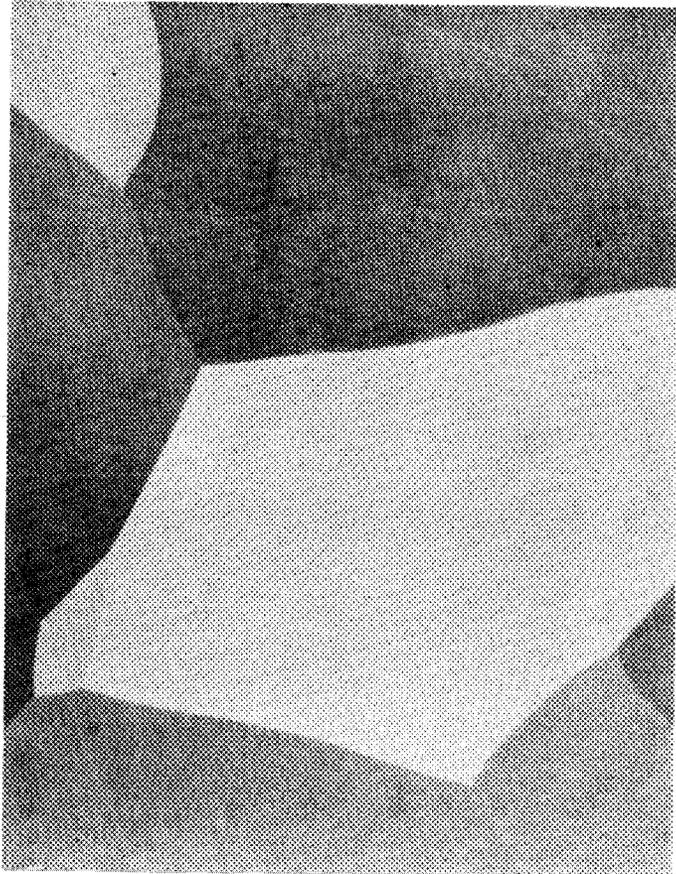


Fig. 7. Microstructure of rod 2, slice 4, showing traces of UC₂ within the grains.

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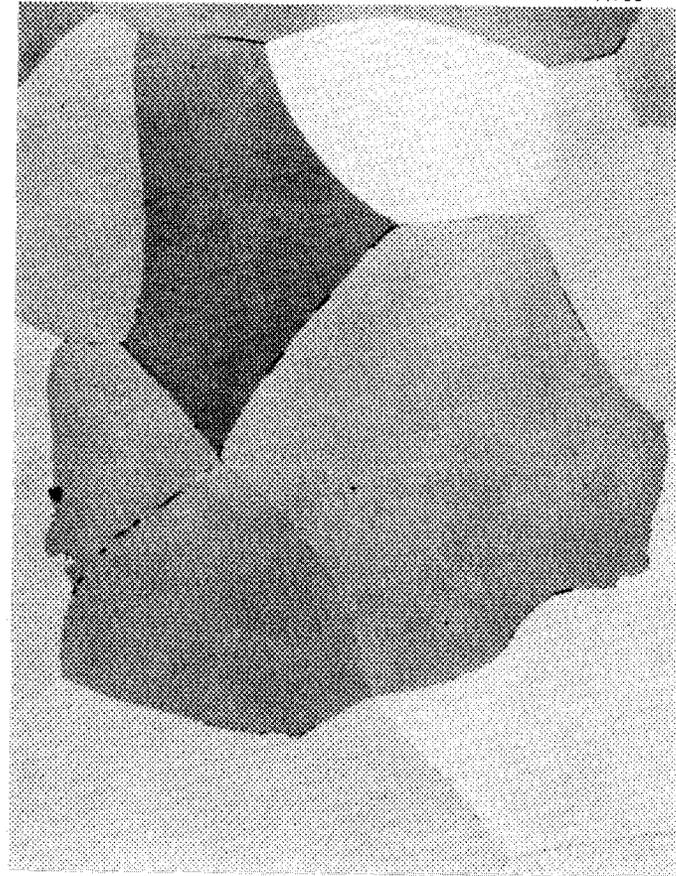
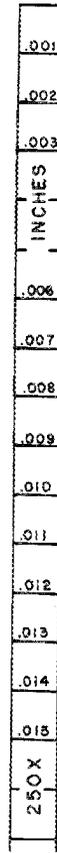


Fig. 8. Microstructure of rod 3, slice 8, showing traces of α -uranium at the grain boundaries.

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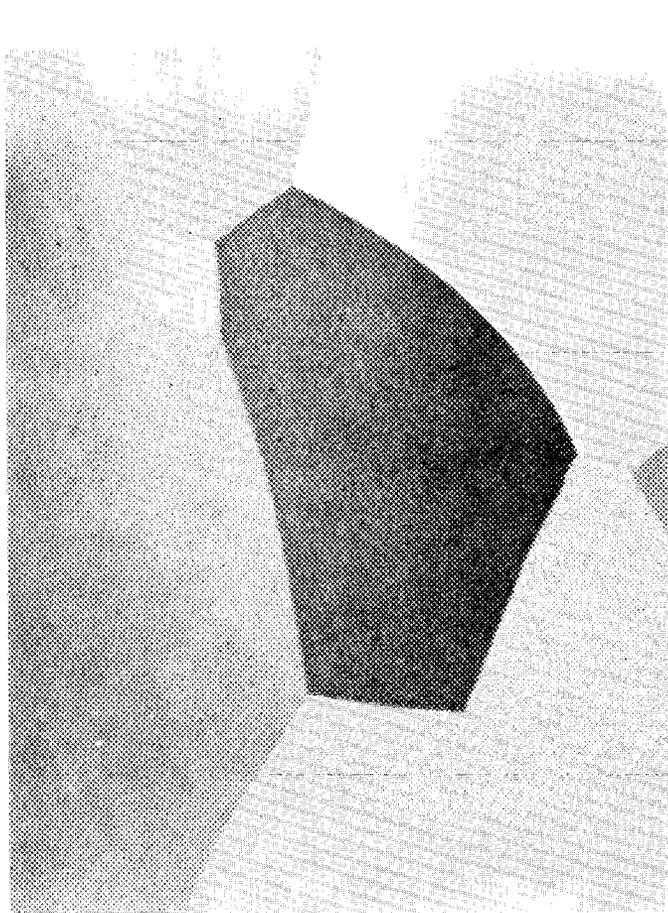


Fig. 9. Microstructure of rod 4, slice 3, showing UC_2 impurity.

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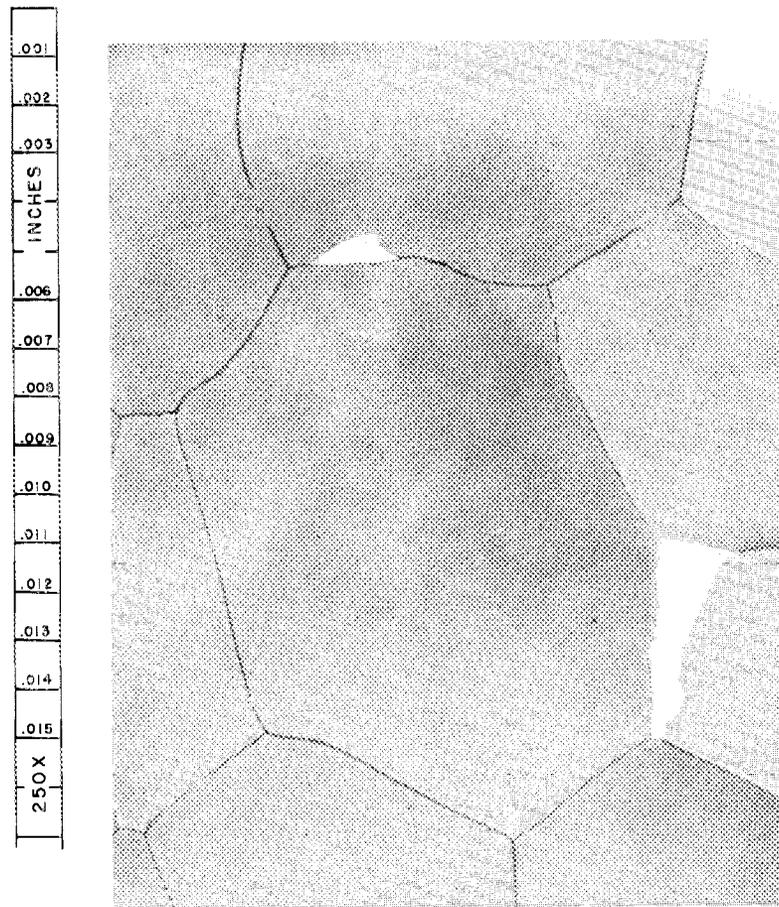


Fig. 10. Microstructure of rod 5, slice 10, showing regions of high-purity UC.

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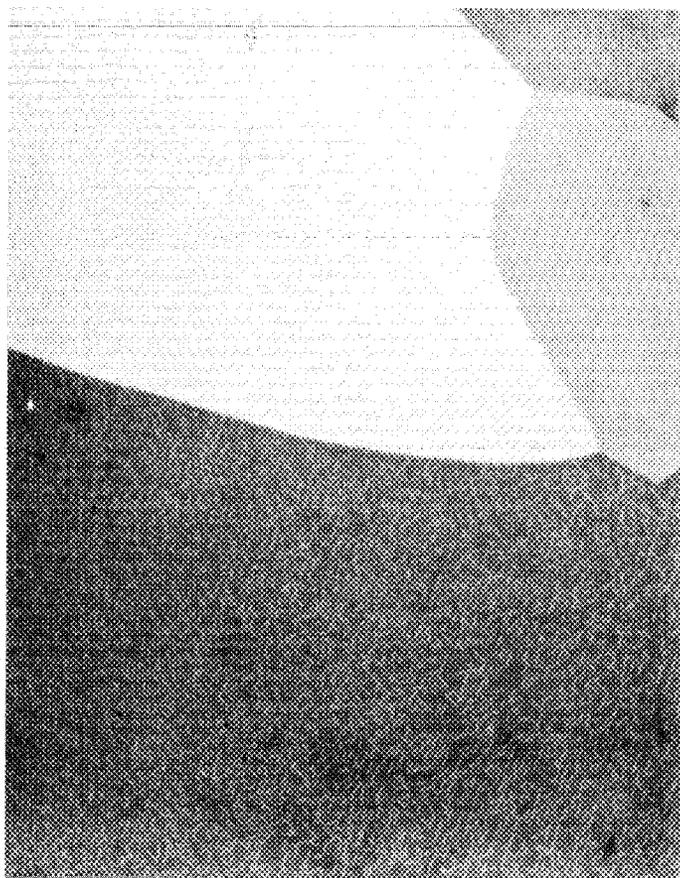


Fig. 11. Microstructure of rod 6, slice 4, showing UC₂ impurity.

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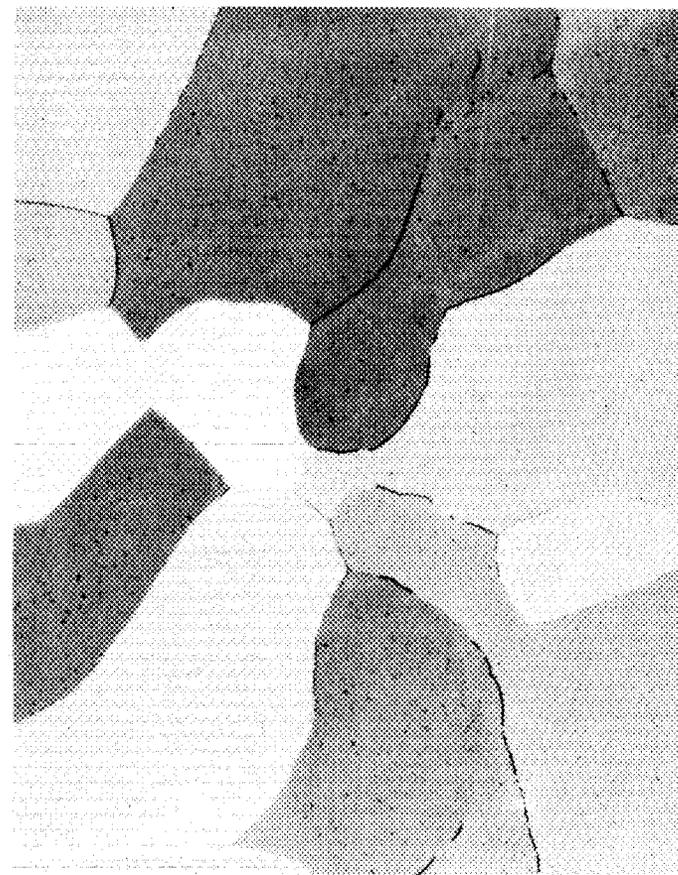
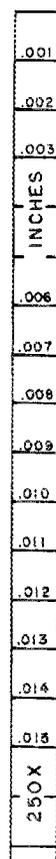


Fig. 12. Microstructure of rod 7, slice 10, showing α -uranium at the grain boundaries and as a spheroidized precipitate within the grains.



Fig. 13. Microstructure of rod 8, slice 3, showing UC₂ impurity.

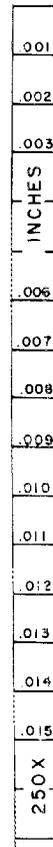


Fig. 14. Microstructure of rod 9, slice 11, showing traces of α -uranium as a spheroidized precipitate within the grains and at the grain boundaries.

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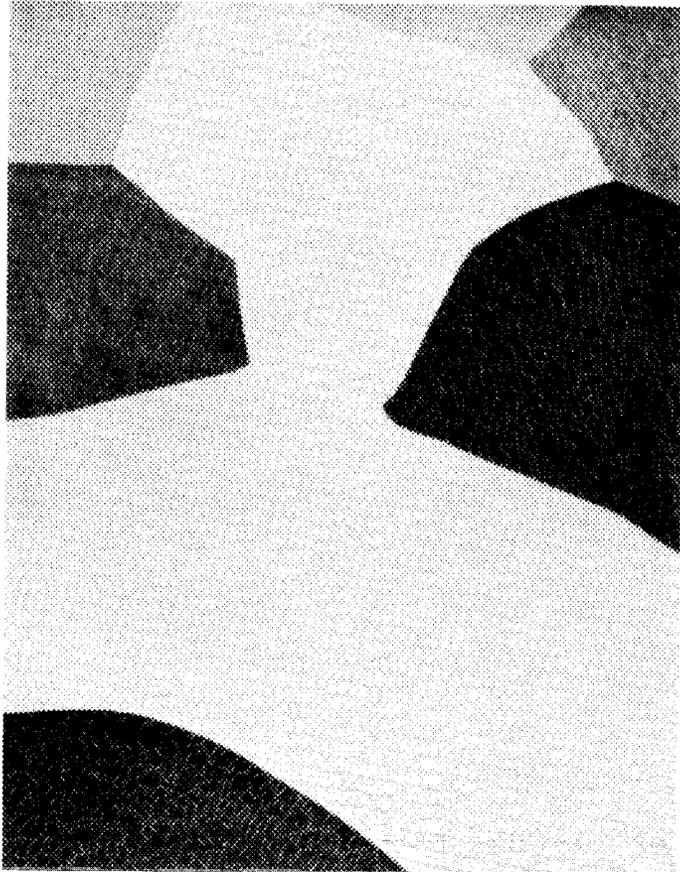


Fig. 15. Microstructure of rod 10, slice 3, showing UC₂ impurity.

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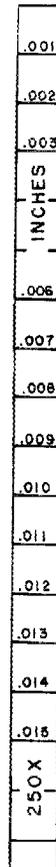


Fig. 16. Microstructure of rod 11, slice 7.

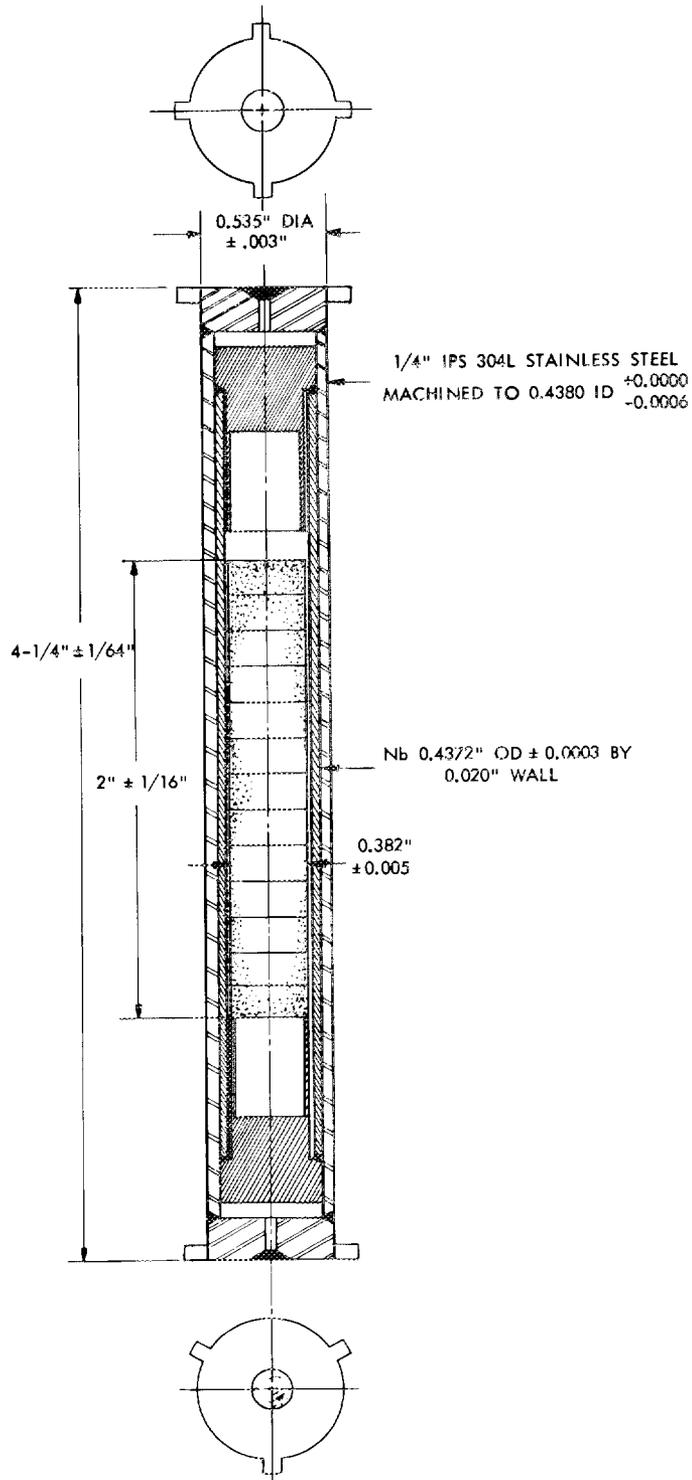


Fig. 17. Uranium monocarbide capsule.

Table 2. Hydrolysis of UC Castings at 80°C

Specimen	Total Vol Gas (ml/g)	Total mg-atoms per g		Composition of Gas Evolved ^a (vol %)								
		C	H	H ₂	CH ₄	C ₂ H ₆	C ₃ H ₈	C ₄ H ₁₀	C ₅ H ₁₂	C ₆ H ₁₄	C ₂ H ₄	C ₄ H ₈
R-1-4	91.0	3.84	15.79	10.6	86.3	2.01	0.42	0.23	0.17	0.06	0.03	0.14
R-4-12	90.7	3.95	16.22	6.8	90.3	1.87	0.41	0.21	0.15	0.03	0.02	0.10
R-9-2	89.7	3.93	15.88	8.0	88.9	1.80	0.49	0.25	0.23	0.04	0.07	0.16
R-11-11	90.0	3.81	15.65	10.1	86.9	1.85	0.42	0.23	0.24	0.04	0.03	0.10
Avg.	90.4	3.88	15.88	8.9	88.1	1.88	0.44	0.23	0.20	0.04	0.04	0.12

^a Traces of C₇ and C₈ compounds were also present.

5.1 Inspection of the Niobium Tubing and Rod

The niobium tubing (nominally 7/16 in. in inner diameter, with a 0.020-in.-thick wall) was checked by ORNL Inspection Engineering for dimensions and by Vidigage, eddy current, and ultrasound methods, and for leak tightness by the dye penetrant method. The outer diameter ranged from 0.4390 to 0.4395 in., and the wall thickness from 0.020 to 0.022 in. A few portions of the tubing, in which defects were indicated, were discarded. Ductility was determined by flattening a 2.5 in. length without cracking. Spectrographic analysis showed about 1% tantalum, 0.01 to 0.1% iron and molybdenum, and less than 0.01% of any other impurities. A niobium bar was swaged to provide the starting stock for plug machining. Spectrographic analyses showed about 0.03% tantalum and 0.01 to 0.1% iron; all other impurities were below 0.01%.

5.2 Fabrication and Uranium Monocarbide Loading of the Niobium Capsules

All welds were performed by the Tungsten Inert Gas process in the Welding and Brazing Shop of the ORNL Fabrication Department. The first end plug was welded to the tube in a dry box that had been evacuated and then backfilled with argon. The weld was dye checked and x-rayed.

The UC slices were raised into the capsule by placing its open end into the glass-lined sample bottle described in Sec 3. The distance from the top of the stack to the top of the tube ranged from 0.753 to 0.775 in. The second niobium weld was made in the dry box after evacuation and helium backfilling. The capsule was helium-leak-checked, dye-checked, and x-rayed. After the outer diameter was machined to tolerance, the capsules were again x-rayed and helium-leak tested. The orientation of the UC slices was established by numbering only the top niobium end plug. Two capsules contain specially polished slices for metallography, which are located at the top of the stack. The complete loading diagram is listed below. Slices are numbered from the top of the capsule, with the original rod number followed by the slice number from the rod.

<u>1-Cycle Irradiation</u> <u>(~300 Mwd/ton U)</u>		<u>~15,000 Mwd/ton U</u> <u>Target Irradiation</u>	
<u>Capsule 1</u>	<u>Capsule 6</u>	<u>Capsule 3</u>	<u>Capsule 4</u>
11-2	7-4 (polished)	11-4	7-3 (polished)
11-3	4-10	11-5	1-6
9-3	4-11	11-6	4-8
9-4	6-7	8-4	4-9
9-5	6-8	8-5	6-3
9-6	6-9	8-6	6-5
9-7	6-10	8-7	6-6
9-8	3-9	8-8	3-6
9-9	3-10	8-9	3-7
9-10	5-11	8-10	5-7
9-12	5-12	8-11	5-8
9-13	5-13	8-12	5-9

<u>~40,000 Mwd/ton U</u> <u>Target Irradiation</u>		<u>Furnace Testing</u> <u>(5 slices/capsule)</u>	
<u>Capsule 5</u>	<u>Capsule 9</u>	<u>Capsule 7</u>	<u>Capsule 8</u>
11-8	1-5	4-4	4-7
11-9	4-5	1-7	1-12
11-10	4-6	7-6	7-8
10-5	6-12	7-7	7-9
10-6	6-13	7-12	7-13
10-7	3-4		
10-8	3-5		
10-9	3-6		
10-10	5-4		
10-11	5-5		
10-12	5-6		
10-13	2-12		

5.3 Stainless Steel Canning

The stainless steel tubular shell was placed on a hot plate and the niobium capsule inserted. The stainless steel plugs were welded to the

tube in an argon atmosphere, leaving vent holes in both end plugs unsealed. The orientation of the UC slices was identified by having the top end plug bear two numbers and the bottom only one.

The end vents were sealed in the dry box after evacuation and helium back filling. The units passed the helium-leak test, but x-ray examination showed insufficient depth of penetration on a few of the vent closures. Rejected welds were machined out. After repair, the units were again x-rayed and helium-leak tested. The units were machined to the required diameter and again x-rayed and helium-leak tested.

Four spacer nubs were welded to the top plug and three to the bottom plug to ensure orientation identification, and the units were x-rayed again. The bottom slice of capsule 6, i.e., slice 13 of rod 5, was cracked.

6. REQUESTED IRRADIATION

The irradiation requested is: Capsules 1 and 6 for one cycle at a thermal neutron flux of 6×10^{13} ; capsules 3, 4, 5, and 9 to a time-integrated flux of 2.3×10^{21} (at 6×10^{13}) that to be followed by irradiation at a thermal neutron flux of 9×10^{13} to a cumulative time-integrated flux of 5.2×10^{21} . For capsules 5 and 9, an additional irradiation at a flux of 1.2×10^{14} for a time-integrated flux of 1.2×10^{22} was requested. The capsules are to be oriented with the top up in the reactor to avoid pressure on the polished slices.

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