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THIRD QUARTERLY REPORT

THE FABRICATION OF CLAD MASSIVE UO_2 FUEL ELEMENTS BY COEXTRUSION

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Nuclear Metals, Inc.
Concord, Massachusetts

Contract No. AT(30-1)-1565

A. R. Kaufmann
Technical Director

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I. INTRODUCTION

This report, the ninth of the series, describes the experimental work performed during the months of November and December 1959, and January 1960, on "The Fabrication of Clad Massive UO_2 Fuel Elements by Extrusion". This work is sponsored by the AEC Fuel Cycle Development Program on Contract No. AT(30-1)-1565.

The primary objective of the program includes the development of the multi-temperature coextrusion technique as a procedure for the economic production of stainless or Zircaloy-clad uranium-oxide fuel elements. Arbitrarily, the dimensions and some specifications of the elements to be used for the Yankee Reactor are assumed to be a prototype. The elements contain 90 inches of oxide, 0.294 inch in diameter, clad with 21-mil stainless steel.

The experimental work of this report period has five objectives: (1) extrusion of large cores, (2) higher billet reductions, (3) elimination of graphite as a coextruded sheath, (4) improvement of the core-clad interface by the incorporation of a porous, thin-walled sheath of UO_2, which, during extrusion, would insulate the core, and (5) approach to the actual dimensions of the Yankee prototype. In addition, correlations between billet design and diameter of extruded core and clad, and the interface between them, were sought by analysis of longitudinal and transverse sections.

II. SUMMARY

Six rods of uranium oxide were extruded at high temperature to produce, in general, high density oxide of desirable stoichiometry. Four extrusions were produced from 1.5-inch billet cores, two at reductions of 12X, and two at reductions about 20X. Two other extrusions were performed directly in stainless steel from 0.875-inch billet cores at reductions of 12X without an inner sheath of graphite or tantalum.

III. EXPERIMENTAL PROCEDURE

A. Pre-extrusion Preparations

1. Billet Cores

All the extrusion cores for the experiments of this quarter were prepared in graphite molds by hot pressing "sinterable material", or ADU, which is supplied by the Davison Chemical Company. In general, a temperature of 1750°C and pressures up to 5000 psi were employed to produce compacts 2.0 inches long with diameters of 1.0 and 1.50 inches. The smaller compacts were machined to a diameter of 0.875 inch with a 120° conical point. The larger compacts were machined to have a 120°, 0.5-inch chamfer.

2. Billet Shells and Inserts

Two billet designs, formerly used for 0.875-inch cores, were modified to produce coextrusions with 1.50-inch cores. One type, in which the core is heated and then coextruded in tantalum, has been referred to as Design IV, and is shown in Fig. 1. In the other (Fig. 2), the core is heated and coextruded in graphite (Design VII). A third and new design (Design VIII) was machined and used to extrude UO_2 directly in 304 stainless steel without a coextruded inner sheath, and is shown in Fig. 3. For a variation of this design, a uranium-oxide cup, 3.0 inches deep, inner and outer diameters 0.90 and 0.95 inch, respectively, was prepared by slip casting ADU oxide in an appropriate mold.

3. Heating the Billet Components

The cores were heated in an argon atmosphere by a graphite susceptor. Usually, 15-minute heating cycles were used to reach and soak (for two minutes) at the desired core temperature, which was measured by an optical pyrometer. Containers for heating were either a tantalum or graphite can, or graphite manipulator. The latter were essentially two concentric tubes with tapers arranged in such a way that sufficient compression could be maintained on the hot core to permit its vertical withdrawal from the furnace without dropping out.

Other components of the billets, the steel shell and stainless inserts, were heated in graphite containers in an air furnace. These parts were soaked at temperature for one hour.

4. Billet Assembly

After the oxide core, heating in the graphite susceptor, had been maintained at its extrusion temperature for two minutes, a die and cone, and the metal components of the billet were withdrawn from their respective furnaces and charged into the "liner" or pressure vessel. In some cases, a room-temperature stainless insert (either with or without an oxide insulator) was inserted into the billet at this time. Immediately after, the hot core was assembled in the billet, either by insertion of the charged heating can (tantalum or graphite) and snapping a constriction in the handle next to the back of the can, or ejection from the graphite collet or manipulator by a central push rod. Another steel component called the cutoff, which is designed to seal the back of the billet during extrusion, precedes insertion of the dummy block, a hardened steel cylinder that prevents the backward extrusion of the billet. The time for assembly, which ideally would be instantaneous, visually requires about 20 to 30 seconds. However, complications arising from the expansion of UO_2 and the vertical furnace design prevented quick ejection of the hot core. In these cases, about one minute was required before the billet was upset (received pressure from the ram) and was extruded.

B. Extrusion Conditions

The fastest ram speed obtainable with the press is 135 in./min, and graphite and oil lubrication were employed for all extrusions. The dies (900 and 600-mils ID) and the liner (2.80-inch ID) were maintained at 900^{o}F. Steel components of the billet, the shell and cutoffs, were heated and extruded at the temperatures (1200 to 1500^{o}F), which, for the given billet designs and reductions, were anticipated to require the highest pressures of which the extrusion tools (ram and liner) were capable.

The oxide cores were heated to the temperatures which had formerly been found to produce the best results for a particular billet design: 3200°F in graphite and 3600°F in tantalum. The stainless parts were either heated with the steel shells to the same temperature as the steel components or were unheated, being installed in the billet at room temperature. The details of the extrusion conditions are listed in Table I in relation to the experimental objective of the extrusion.

C. Post-Extrusion Preparations and Determinations

Transverse cuts produced sections which were prepared for dimensional inspection or measurement and core density and stoichiometry determinations. Three types of specimens were prepared for dimensional analysis: a 4-inch longitudinal section for gross over-all examination, and a transverse and short longitudinal for metallographic study.

Densities of cores were determined by first removing the cold-rolled steel exterior from a one-inch section of the extrusion. Then density of the stainless clad with core was determined, followed by the density of the clad only. (The oxide was consumed by HNO_3.) The density of the UO_2 was calculated by difference. As a check, a quantitative analysis of the weight of uranium dissolved in the acid was performed. In this way the measured densities included porosities arising from spacings in cracks and other voids.

The stoichiometry was determined by oxidizing a known weight of UO_{2+x} to stoichiometric U_3O_8, reweighing, and calculating the original excess oxygen of the sample.

IV. RESULTS

Dimensions - The reductions in area between the billet and extrusions' cross sections differed according to the billet design and die size. The larger cores extruded through the larger dies suffered about the same reduction as the smaller cores extruded through the smaller dies. In all the extrusions except one (148), the core reduced more than any other component in the billet. This might imply that the clad maintains a transverse compressive force on the core as it passes through the die. In the extrusion whose core experienced a reduction higher than the clad, a large amount of necco-wafering or segmenting occurred, possibly implying the presence of tensional forces exerted by the clad. A list of sizes and reductions of each extrusion appears in Table II. The variation of core diameter and clad thickness is large as seen in Table III. These pronounced dimensional changes occur as interlocking transverse rings of core and clad. The exterior surface of the clad reflects to some extent its changes of thickness. Where a transverse crease appears on the outside, a larger crease occurs directly beneath on the inside. Dimensional variations are revealed in Fig. 4, which shows the transverse sections, and Fig. 5, which shows longitudinal sections of the extrusions performed during this report period.

Stoichiometry - The oxygen-to-uranium ratio of the starting material was found to be greater than the fabricated or extruded material. The starting powders were determined to have oxygen-to-uranium ratios of 2.134 (Table IV), while the extruded material varied from 2.00 to 2.088.

Densities - The theoretical density of the extruded oxide cores was determined to be high. The values vary from 87.6 to nearly 100 percent theoretical density (Table IV).

Cracks - Coextruded UO_2 cores always show a number of cracks. Since these are revealed only when the clad rods are opened for inspection by machining, it is difficult to determine the extent of this cracking prior to cutting. Longitudinal cracks in radial and circumferential directions are observed as well as cracks perpendicular to the rod axis. Figure 6 shows an idealized representation of a typical cracking pattern of hot-extruded UO_2 with stainless clad. The occurrence may or may not affect bulk density, depending on the spacing between the cracks.

Metallography - Metallographic studies of coextruded UO_2 show that the grains are deformed plastically (Fig. 7) and that some recrystallization has taken place. The figure shows a comparison between the condition of the grains in a plane parallel and perpendicular to the extrusion direction. The extrusion conditions have caused the grains to elongate in the extrusion direction so that their length-to-diameter ratio is increased by a factor of about 10.

V. DISCUSSION OF RESULTS

A. Evaluation

Ratings of each extrusion may be made relative to the several experimental objectives, as indicated in Table I. Not all extrusions were aimed to achieve the dimensions of the imaginary Yankee prototype, which is conceived to be a monolithic 100 percent dense oxide cylinder evenly clad with a 23-mil stainless sheath. Larger cores (1.50 inches compared to 0.875 inch) were extruded for the first time, and a higher reduction of oxide was achieved in a stainless-graphite and stainless-tantalum coextrusion in a cold-rolled shell. The two extrusions that contained no inner sheaths of graphite or tantalum represent the closest approach to the Yankee prototype thus far achieved. Their oxide is nearly monolithic, almost to theoretical density, but their clad is too thick and the core-clad interface is uneven. Of the two, the extrusion whose core was insulated by a slip-cast sleeve gave better results. Figure 8 shows the effect of the presence of the insulator.

B. Operation of Manipulator

An essential device for coextrusion of stainless steel directly over the UO_2 core was a graphite manipulator permitting the heating and handling of bare UO_2. The high coefficient of thermal expansion of uranium oxide caused some difficulty in the ejection of the hot core from the manipulator because insufficient clearance was provided for the core's expansion on heating. In addition, it was necessary to lift the specimen out of the furnace vertically, which complicated the design and construction of the ejector. However, despite its inadequacies, the design provided the first graphite-free extrusions of high density uranium oxide.

C. Selected Comparisons

If the three extrusions that contained coextruded graphite inner sheaths are compared, it is apparent that they have similar appearance despite the 22X reduction of one and the 10X reduction of the other two. However, a comparison of the pair of high reduction extrusions reveals a marked difference in the appearance of the oxide sheathed in tantalum instead of graphite.

As mentioned, if a comparison is made between the extrusions in which the oxide bears directly against the stainless steel, it is evident that the presence of a slip-cast uranium oxide crucible improves the interface between the stainless steel and the hot core. The high density of the oxide (99.7% theoretical density) in this extrusion reveals that the slip casting, even though it was originally at room temperature and of high porosity before upset, achieved a high theoretical density during extrusion.

VI. CONCLUSIONS

The central problem of the program is no longer the effect of temperature on the plasticity of UO_2, but the roughness of the interface between the core and the clad. The smoother interface produced by the coextruded tantalum implies that stiffer cladding components in the billet produce a more even core-clad interface. The UO_2 insulator, which reduces the heat transfer to the stainless cladding component, maintains the oxide nearer the optimum extrusion temperature and reduces overheating of the stainless. Desirable results probably will be obtained if the stainless component is kept as stiff as in the tantalum extrusion and the core as hot as in the insulated extrusion.

UO_2 can undergo reductions of over 20X without apparent sacrifice of extrusion quality.

VII. FUTURE WORK

The most promising extrusion from the standpoint of the imaginary prototype thus far produced is the one that contained the uranium-oxide insulated core. Future work will include exploitation of this insulation concept and attempts will be made to reduce the time of contact between the cladding component and the hot core to the minimum: the instant of extrusion. In addition, the hot press, the core-heating furnace, and the core manipulator will be redesigned to permit horizontal operation. Other objectives are: cores larger in length and diameter, elimination of the cold-rolled component of the billet, lower shell temperatures.

Table I

Extrusion Conditions

Experimental Objective	Extrusion No.	Design No.	Core			Heating Can		Shell Insert(s)			CR Shell		Die	Core	
			Dia. (in.)	Wt. (g)	Temp. (°F)	Mat'l.	Wall (in.)	Mat'l.	Wall (in.)	Temp. (°F)	Wall (in.)	Temp. (°F)		P (tsi)	R (X)
1	23145	VII	1.500	440	3200	Graphite	0.220	SS	0.188	1200	0.156	1200	0.900	NR	12
1	23147	VII	1.500	440	3200	Graphite	0.220	SS	0.188	1100	0.156	1100	0.900	72	12
1-2	23148	VII	1.500	445	3200	Graphite	0.220	SS	0.188	1500	0.156	1500	0.600	68	18
1-2	23230	IV	1.500	446	3630	Tantalum	0.060	SS	0.188	1500	0.156	1500	0.600	83	26
3-5	23227	VIII	0.875	188	3200	None	None	SS SS	0.045 0.030	1200 RT	0.750	1200	0.900	NR	12
3-4-5	23225	VIII	0.875	182	3200	None	None	SS SS UO_2	0.045 0.030 0.050	1200 RT RT	0.750	1200	0.900	62	12

Notes: 2.80-inch ID liner; liner and dies at 900°F; ram speed of 130 in./min; cold-rolled cutoffs; graphite and oil lubrication; core length of 2.00 inches.

Objective No. 1 = Larger cores
Objective No. 2 = Higher billet reduction
Objective No. 3 = Elimination of graphite
Objective No. 4 = Insulation of core
Objective No. 5 = Make Yankee prototype

Legend:
CR = Cold-rolled steel
SS = Stainless steel
RT = Room temperature
NR = Not recorded
P = Running pressure
R = Reduction

Table II

Tabulation of Billet Dimensions, Extrusion Dimensions, and Reductions*

Extrusion No.	Billet Dimensions (mils)				Extrusion Dimensions (mils)				Die Dia. (mils)	Reductions			
	Core Dia.	Can Wall	Clad Wall	Shell Wall	Core Dia.	Can Wall	Clad Wall	Shell Wall		Core	Can	Clad	Shell
145	1500	220	188	156	483	63	97	69	900	12	15	8	7
147	1500	220	188	156	453	63	97	123	900	12	15	8	7
148	1500	220	188	156	343	71	32	15	600	20	26	24	21
230	1500	60	188	156	297	26	90	66	600	26	5	3	16
227	875	None	75	750	250	None	63	44	900	12	None	6	8
225	875	None	75	750	281	None	272	37	900	12	None	7	8

* Approximate reductions are tabulated, since the change in cross-sectional areas of the components of the billet which occurs at upset are not known.

Table III

Measurements of Transverse Sections

Extrusion No.	Core (mils)		Inner Sheath (mils)			Outer Sheath (mils)		
	Diameters		Mat'l.	Wall		Mat'l.	Wall	
	Min	Max		Min	Max		Min	Max
145	406	460	Gr	50	110	SS	62	120
147	420	450	Gr	22	50	SS	86	92
148	340	370	Gr	42	76	SS	36	50
225	258	290	UO_2	0	44	SS	12	42
227	260	290	None	None	None	SS	29	60
230	262	290	Ta	10	22	SS	90	172

Gr = Graphite

Table IV

Stoichiometry and Densities of Extrusions

Extrusion No.	Stoichiometry		Density	
	Raw Material O/U	Extruded Material O/U	Extruded Oxide	
			g/cm^3	% TD
145	2.134	NR	NR	NR
147	2.134	NR	NR	NR
148	2.134	NR	NR	NR
230	2.134	NR	NR	NR
227	2.134	1.999	9.61	87.6
225	2.134	2.088	10.92	99.7

TD = Theoretical density
NR = Not recorded

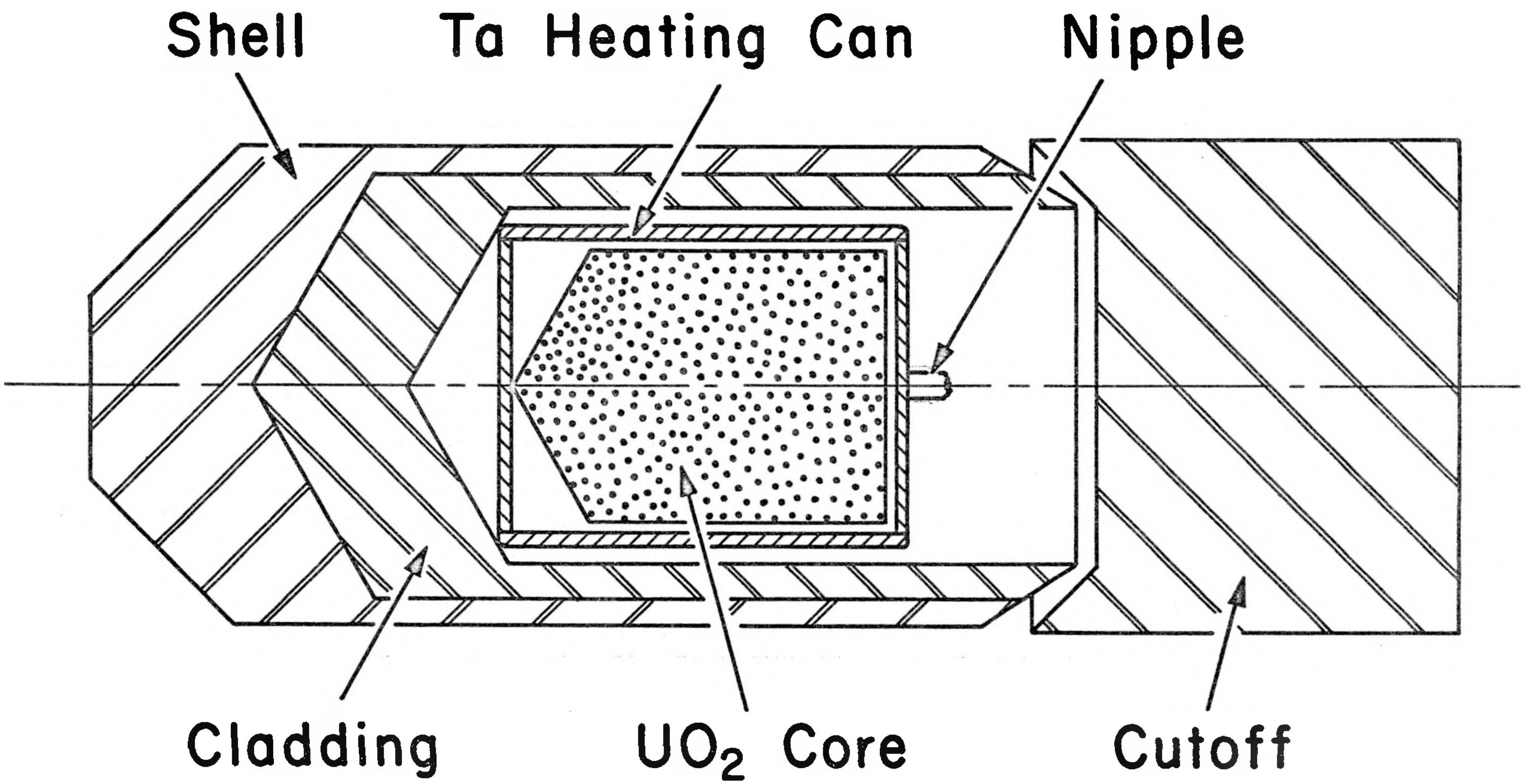


Fig. 1

Billet Design IV (modified) for Coextruded Tantalum Inner Sheath

Formerly used for 0.875-inch cores, this design was modified to produce extrusion 230 from a 1.50-inch core. Drawing No. RA-1505

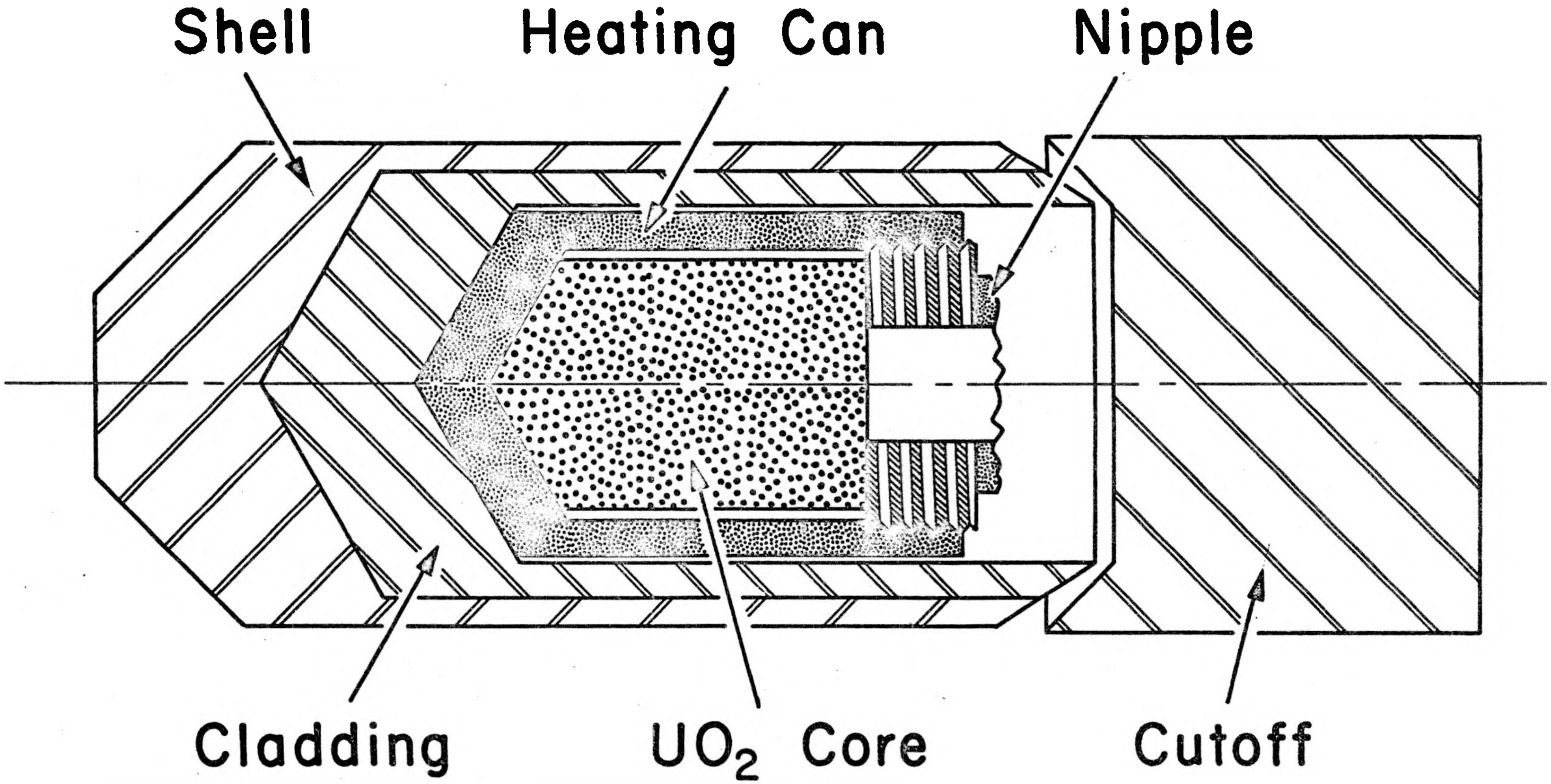
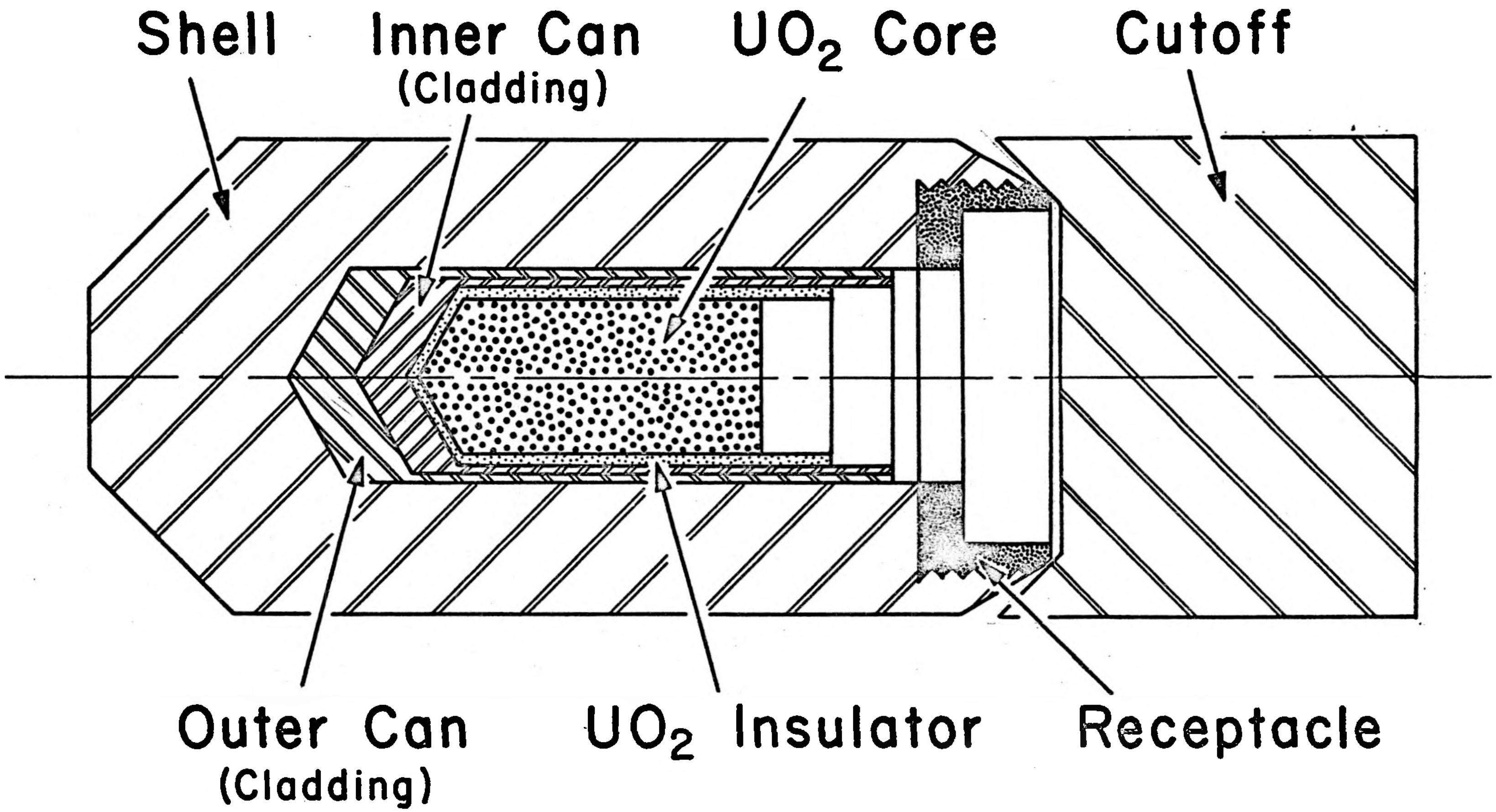


Fig. 2

Billet Design VII for Coextruded Graphite Inner Sheath

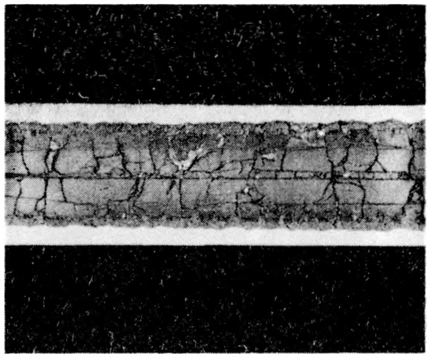
Formerly used for 0.875-inch cores, this design was modified to produce extrusions 145, 147, and 148 from 1.50-inch cores. Drawing No. RA-1506



<u>Fig. 3</u>

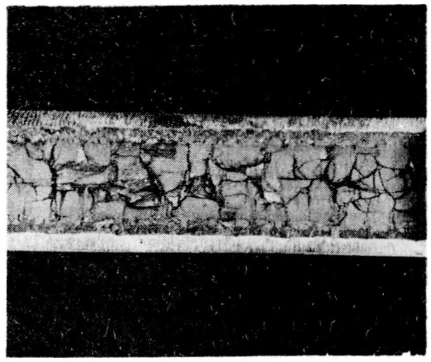
<u>Billet Design VIII for Coextruded Stainless Without Inner Sheath</u>

This design facilitated coextruded uranium oxide and stainless steel without graphite or tantalum inner sheath from a 0.875-inch core (extrusion 227). An unsintered UO_2 cup was inserted into the billet during the assembly to insulate the stainless insert (extrusion 225). Drawing No. RA-1507



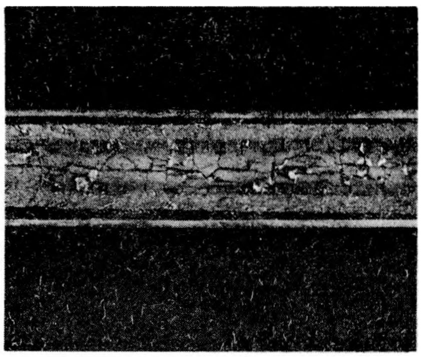
(a) Extrusion 145, RF 7429

Obtained from billet design VII reduced about 10X. The graphite inner sheath is distinguishable between the stainless and the oxide.



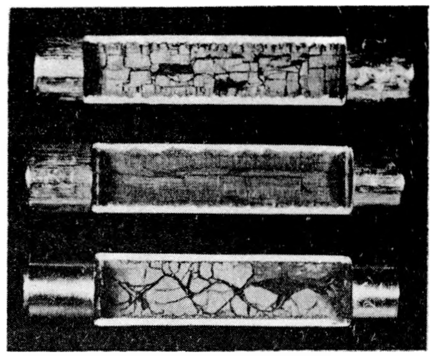
(b) Extrusion 147, RF 7428

Repeat of 145 with cooler outer shell (1100 instead of 1200°F). The graphite inner sheath is distinguishable between the stainless and the oxide.



(c) Extrusion 148, RF 7430

Obtained from billet design VII reduced about 20X. The graphite inner sheath is distinguishable between the stainless and the oxide.

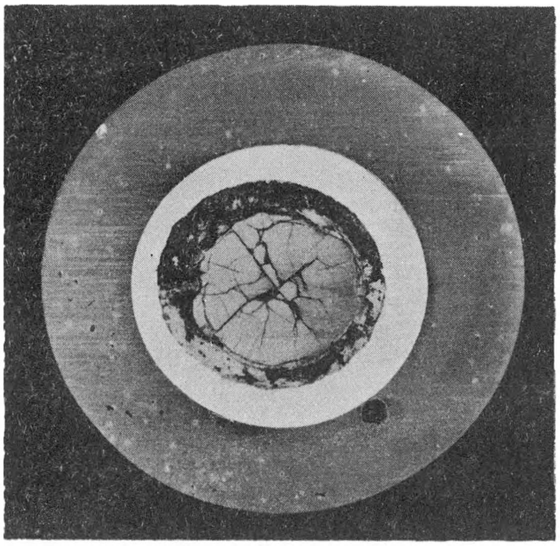


(d) Extrusions 227, 225, 230 (from top to bottom), RF-7426

The upper two extrusions were produced from billet design VIII at reductions of about 10X. The middle extrusion (225) contained the UO_2 insulator. For the lower extrusion (230), billet design IV (modified) was used and reduced about 20X.

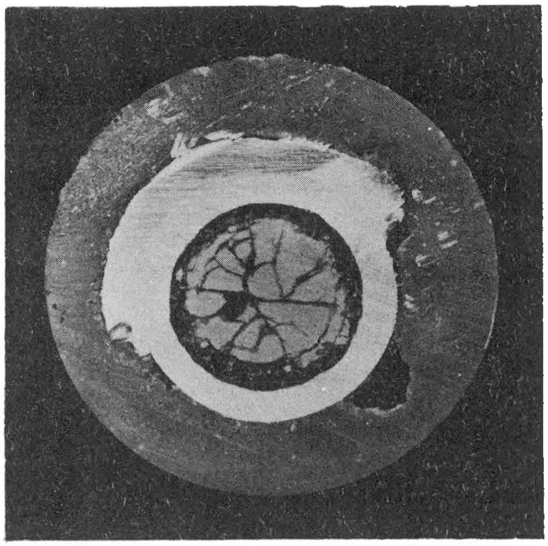
Fig. 4

Longitudinal sections of UO_2 rods. As-polished. Actual size.



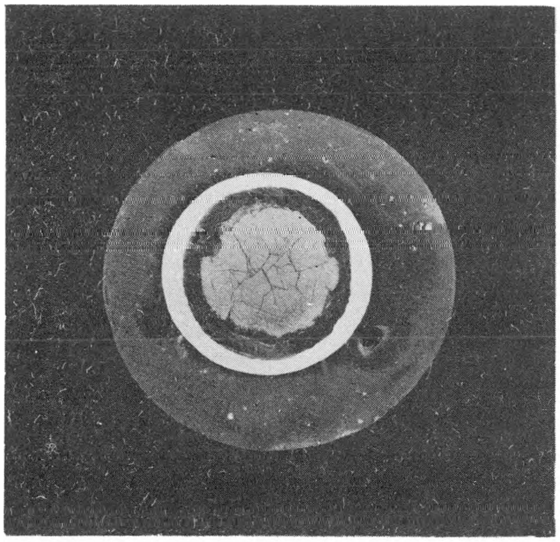
2X RF 7432

(a) Extrusion 145 from billet design VII.



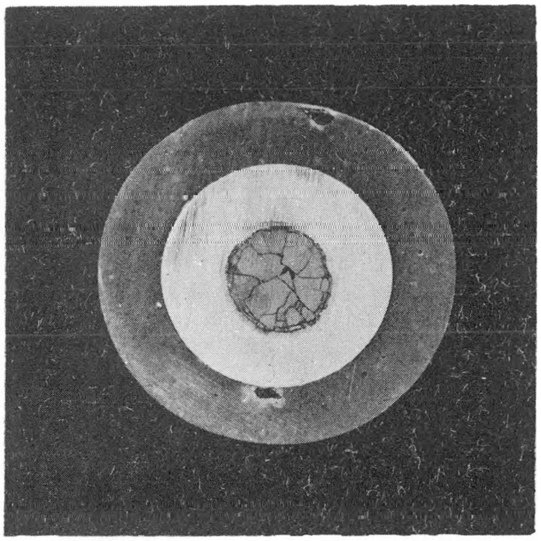
2X RF 7435

(b) Extrusion 147 from billet design VII.



2X RF 7431

(c) Extrusion 148 from billet design VII.

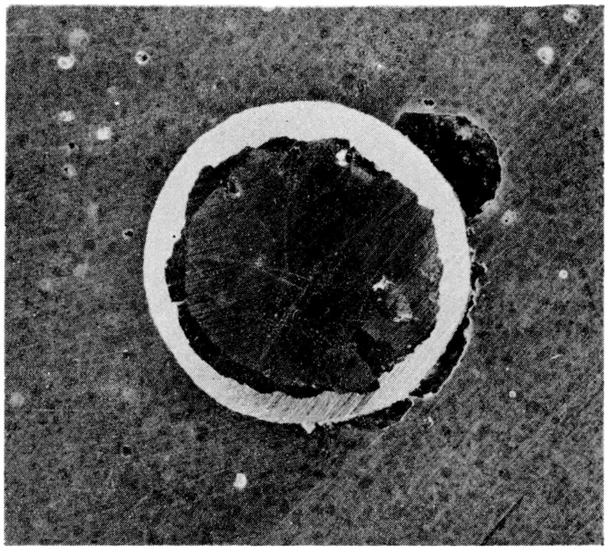


2X RF 7433

(d) Extrusion 230 from billet design IV.

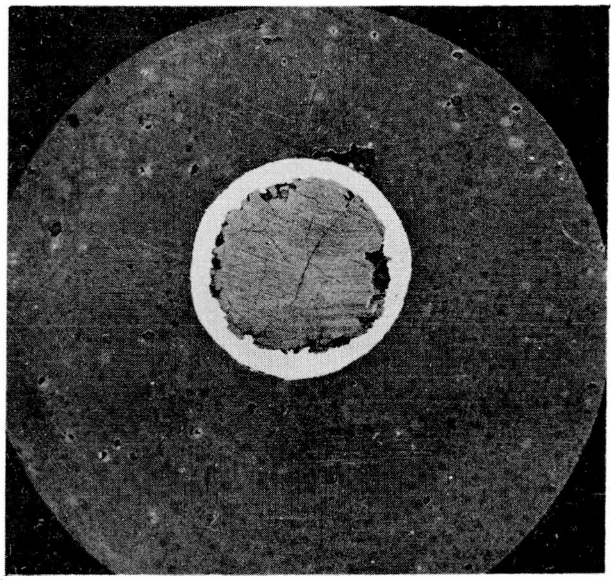
Fig. 5

Transverse sections of UO_2 rods. As-polished.



4X RF 7444

(e) Extrusion 225 which contains the unsintered UO_2 insulator. Portions of the unsintered material are distinguishable at the left portion of the core-clad interface.



4X RF 7445

(f) Extrusion 227

<u>Fig. 5</u> (Cont'd.)

Transverse sections of UO_2 rods. As-polished.

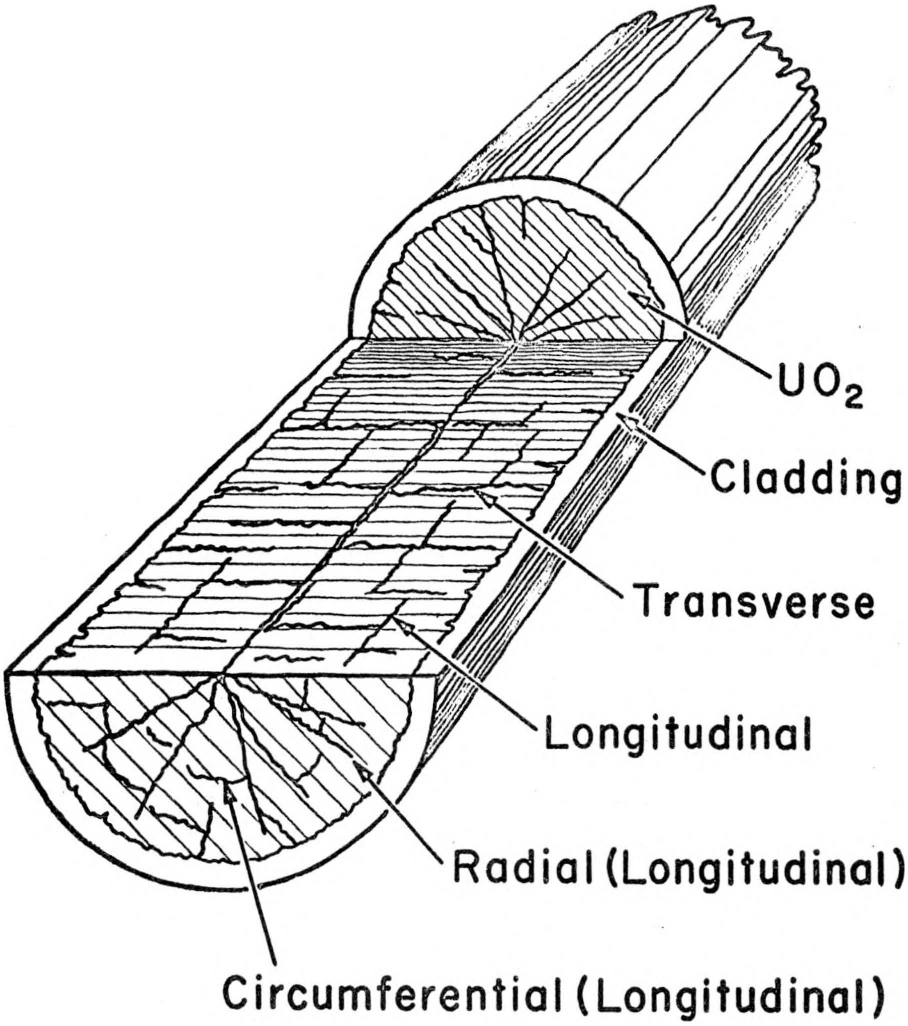


Fig. 6

Idealized Crack Pattern of a Typical Extruded Stainless-Clad UO_2 Rod

Coextruded UO_2 cores always show a number of cracks. Since these are revealed only when the clad rods are opened for inspection by machining, it is difficult to determine the extent of this cracking present to cutting. Longitudinal cracks in radial and circumferential direction are observed, as well as cracks perpendicular to the rod axis. Density measurements have shown that the cracks occupy a negligible volume. Because the occurrence of these cracks does not affect bulk density, they have been of little concern, since it is known that UO_2 always cracks on irradiation.

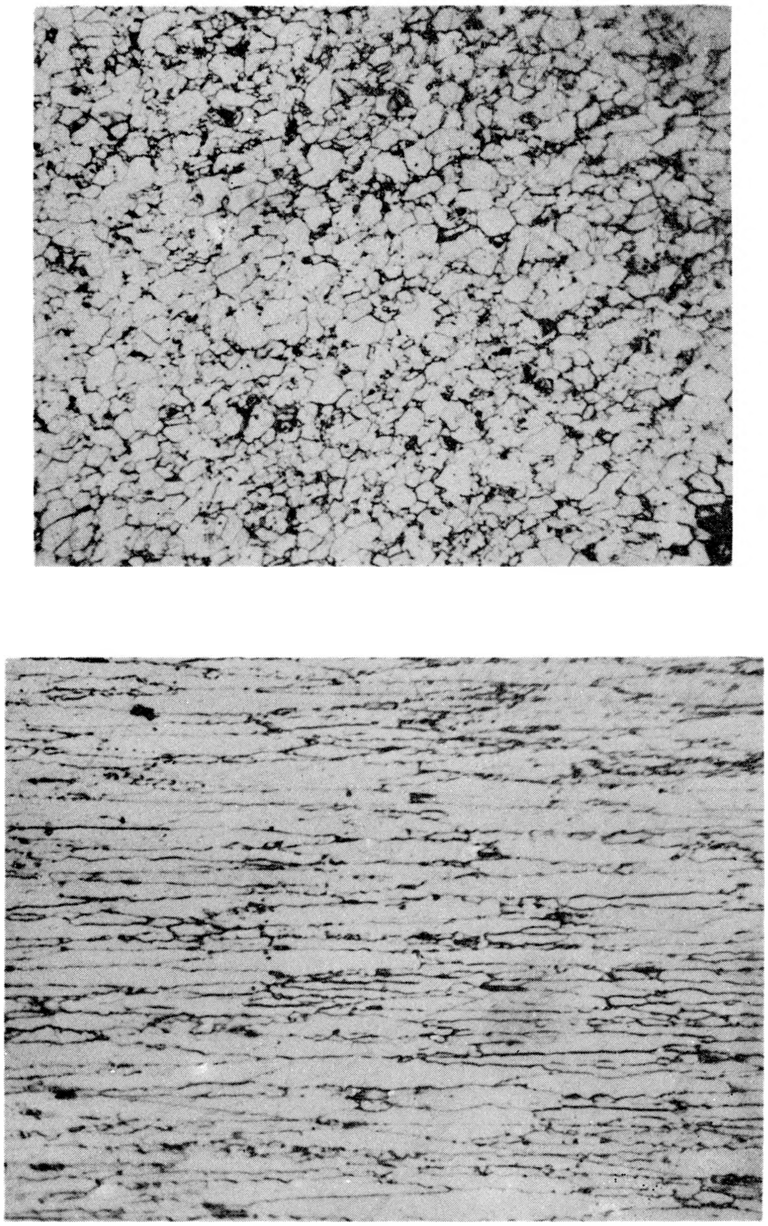


Fig. 7

Microstructure of Extruded UO_2

Metallographic studies of coextruded UO_2 have shown that the grains are deformed plastically. The photograph at the top shows the appearance of the grains in a plane perpendicular to the extrusion direction; the photograph at the bottom shows their appearance parallel to the extrusion direction. Magnification, 500X in each case.

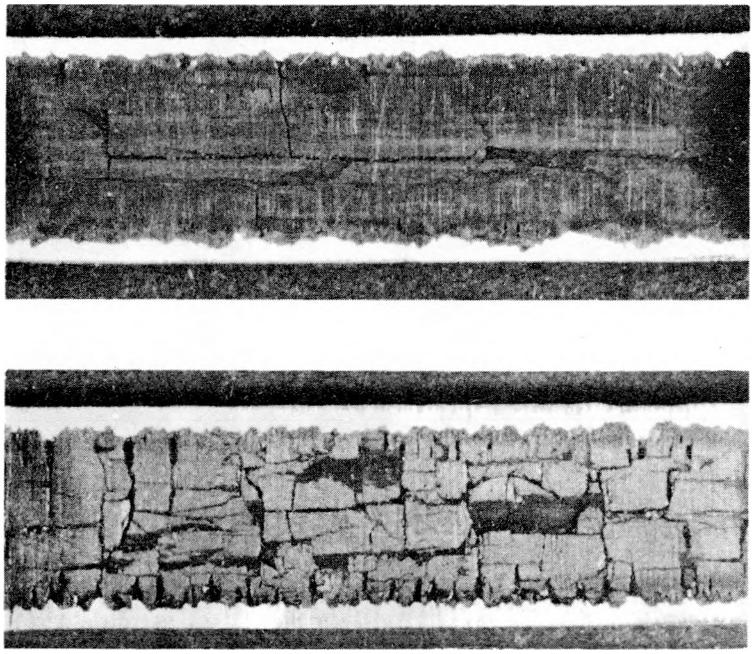


Fig. 8

Effect of the Room Temperature UO_2 Insulator

In the extrusion at the top, a thin layer of unsintered UO_2 was placed on the inside of the cladding material. This was then coextruded at room temperature with the core at a high temperature and served as an insulator to keep the cladding relatively cool. The extrusion on the bottom was performed without the insulator, but conditions were otherwise the same. Extrusion reductions were about 10X. Magnification, 3.5X.