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CASTING OF MULTIPLE ^{233}U METAL
TARGET DISKS

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CASTING OF MULTIPLE ^{233}U METAL TARGET DISKS

William V. Conner and Donald L. Baaso

Abstract. The preparation of four uranium-233 metal target disks is described. The disks were 1.5 inches in diameter with thicknesses of 0.050, 0.100, 0.200, and 0.640 inches. The disks were prepared using casting techniques. The three thinner disks were prepared simultaneously in a single casting. The variation in the thickness of each disk was within 3.5% of the average thickness of the disks. The conversion-to-metal of the U_3O_8 feed provided for the project is also described.

INTRODUCTION

Casting techniques have been developed at Rocky Flats for preparing small actinide metal targets for neutron cross-section and other measurements.¹ These casting techniques were developed to eliminate the large amounts of scrap generated by the rolling procedure formerly used for preparing targets.² Using the proper mold design and casting parameters, it has been possible to cast 1.00 ± 0.010 -inch-diameter plutonium-metal disks varying in thickness from 0.010 to 0.180 inch with minimal scrap losses. Recent casting procedure improvements have made possible the simultaneous casting of several metal disks in one mold. This multiple disk casting procedure was used to prepare uranium-233 metal disks for the Oak Ridge National Laboratory (ORNL).

ORNL requested that four uranium-233 metal disks be cast from 700 g of uranium (99.4% ^{233}U) which Oak Ridge supplied as U_3O_8 . The finished disks were to be 1.50 ± 0.005 inch in diameter and 0.050-, 0.100-, 0.200-, and at least 0.575-inch thick. The final machining of the disk diameters was to be performed at ORNL and the machined disks were to weigh at least 500 g. Rocky Flats was to supply cast disks at least 1.55 inches in diameter to provide sufficient material for machining.

EXPERIMENTAL

The uranium-233 used for this project was isotopically enriched (99.4% ^{233}U) and, therefore, could not be contaminated with other uranium isotopes or with other actinide elements. Before this project was started, the glovebox was cleaned by sweeping up all loose dust and then wiping the inside surfaces of the glovebox with damp paper towels. The hydrofluorination, reduction, and casting

equipment was also cleaned. The equipment in intimate contact with the ^{233}U , such as the hydrofluorination boat, reduction crucible, melting crucibles, and molds, was all new.

EQUIPMENT

Conversion-To-Metal

The conversion of the $^{233}\text{U}_3\text{O}_8$ to UF_4 was performed in a horizontal tube furnace (See Figure 1). The furnace consisted of a heated 24-inch long by 5-inch diameter Inconel tube with HF , H_2 , and N_2 supply lines and a HF off-gas and disposal system. A gold-plated Inconel hydrofluorination boat, 8-inches long by 4-inches wide by 2-inches deep, was used to hold the U_3O_8 .

Figure 1. Hydrofluorination Furnace and Boat Used to Convert $^{233}\text{U}_3\text{O}_8$ to UF_4 .

The reduction vessel, (Figure 2), consisted of a MgO crucible and crucible lid, and a stainless steel pressure vessel and vessel lid. The crucible was placed in the pressure vessel and the annulus between the crucible and pressure vessel was filled with MgO sand. A copper gasket was used to obtain a seal between the pressure vessel and pressure vessel lid.

Figure 2. The 500-g Scale Reduction Vessel.

Casting

The casting furnace, (Figure 3), was a System VII General Purpose Metallurgical Facility manufactured by Vacuum Industries. This furnace consisted of a tilt-pouring, 4-inch diameter by 6-inch high water-cooled induction coil enclosed in a water-cooled vacuum chamber. The coil contained a quartz insulator sleeve and a ¼-inch-thick sleeve of WDF graphite felt insulation manufactured by

Figure 3. Interior of Casting Furnace.

Union Carbide. The power for the coil was supplied by an Inductotherm 15-kW motor-generator unit. The 3-kW ceramic mold heater was a resistance-type unit fabricated by the Rocky Flats Research and Development Ceramics Laboratory. Temperature measurements were obtained with either chromel-alumel or platinum-rhodium thermocouples, or an optical pyrometer, depending upon the temperature range to be measured. The vacuum system consisted of a 4-inch diffusion pump with a mechanical roughing pump.

The melting crucibles and molds used for this project were fabricated from stackpole graphite. The mold and crucible surfaces in contact with the uranium were coated with a zirconium oxide mold coating preparation before being used.

PROCEDURE

Conversion To Metal

The $^{233}\text{U}_3\text{O}_8$ was calcined at 600°C for 6 hours to remove any volatiles and to ensure a reactive oxide. The calcined U_3O_8 was placed in the Inconel boat and weighed. The loaded boat was placed in the furnace which was sealed and checked for leaks. The furnace was heated to 650°C and H_2 gas was passed through the furnace at 5 liters per minute for 2 hours to reduce the U_3O_8 to UO_2 . This was followed by a mixture of $4\frac{1}{2}$ liters per minute of HF and one liter per minute of H_2 for $9\frac{1}{2}$ hours to convert the UO_2 to UF_4 . The furnace was allowed to cool and the boat was reweighed to determine the weight gained during the hydro-fluorination.

The UF_4 was reduced in two batches containing 350 g of uranium each. The reduction charges were prepared using a 30 mole% excess of Ca^* and 0.5 moles of I_2 per mole of uranium. Calcium chloride (0.2 moles CaCl_2 /mole U) was added to the charges to improve the metal yield. The charges were loaded as follows:

A small portion of the Ca and I_2 (0.04 moles I_2 /mole U) was placed in the bottom of the crucible. The UF_4 was then mixed with the bulk of the Ca and I_2 (0.4 moles of I_2 per mole of U) and this mixture was added to the crucible. The CaCl_2 was then placed on top of the main charge and the remaining Ca and I_2 (0.1 mole I_2 /mole U)

*The excess of calcium is the amount of calcium in excess of that required stoichiometrically to react with all of the UF_4 and I_2 in the charge. This excess of calcium was distributed such that each charge layer containing I_2 or UF_4 was provided with a 30% excess of calcium.

was placed on top of the CaCl_2 layer. The lid was bolted on the vessel and the assembled vessel was checked for leaks and purged with argon. The prepared reaction vessel was placed in a vertical crucible furnace which had been preheated to 950°C . After the charge reacted, as determined by a decrease in the neutron flux, the furnace was turned off and the vessel allowed to cool. The metal buttons were pickled in distilled water and buffed to remove any adhering Ca and slag.

Casting

The three thinner disks (0.050, 0.100, and 0.200 inch) were cast first using all of one button plus a portion of the second button. The uranium metal, 484 g, was placed in the melting crucible and the loaded crucible was placed in the induction coil. The mold was placed in the mold heater and the chamber was sealed and evacuated to 10^{-4} torr. The metal was heated to 1400°C and the mold was heated to 930°C . The molten metal was then poured into the heated mold and allowed to cool under vacuum.

The thick disk (0.575 inch) was cast using the excess of metal from the first casting plus the remaining portion of the second metal button. This metal, 429 g, was heated to 1350°C before being poured into the mold which had been heated to 1000°C . The metal was allowed to cool to 100°C under vacuum before being removed from the furnace.

RESULTS

The conversion of the $^{233}\text{U}_3\text{O}_8$ to metal produced a 98% metal yield. The hydrofluorination step produced a 95.6% conversion to uranium tetrafluoride, while the reduction step produced metal yields of 98.2 and 97.9%. The hydrofluorination and reduction data are given in Table 1.

The cast disks satisfied all of the specifications. The disk diameters were all greater than 1.55 inches and the maximum variation in the thickness of the four disks was 3.4%. The disk produced by the second casting was 0.640-inch thick, which was over 0.050-inch thicker than the required minimum of 0.575 inch. The total weight of the four disks was 590 grams, which was more than sufficient to provide the required 500 g of uranium in the finished product after machining at ORNL. The weights and dimensions of the four disks are given in Table 2.

Samples of the excess of metal from the casting operation were submitted for isotopic and alpha pulse height analysis. The isotopic analysis on the U_3O_8 supplied by ORNL and the analysis obtained from the metal sample are given in Table 3. As the data in Table 3 show, the analysis obtained from the metal sample was the same as the analysis obtained from the U_3O_8 , within precision limits of the analytical technique used. The results of the alpha pulse height analysis obtained on the metal and U_3O_8 are given in Table 4. These analyses show that there was actually a

Table 1. Hydrofluorination and Reduction Data.

HYDROFLUORINATION							
<u>U₃O₈ Wt (g)</u>	<u>UF₄ Wt (g)</u>	<u>Conversion to UF₄ (%)</u>	<u>Temp. (°C)</u>	<u>H₂ Flow Rate (liter/min)</u>	<u>HF Flow Rate (liter/min)</u>	<u>Length of Hydrofluorination (hours)</u>	
834	923	95.6	650	1	4½	9½	
REDUCTION							
<u>Batch No.</u>	<u>UF₄ Wt (g)</u>	<u>U Wt (g)</u>	<u>Ca Excess (%)</u>	<u>mol I₂ mol U</u>	<u>CaCl₂ Wt (g)</u>	<u>Button Wt (g)</u>	<u>Metal Yield (%)</u>
1	461.5	350	30	0.5	33.3	343.6	98.17
2	461.5	350	30	0.5	33.3	342.7	97.91

Table 2. Weights and Dimensions of ^{233}U Disks.

Disk No.	Disk Wt (g)	Diameter (inch)		Thickness (inch)		Variation in Thickness (%)
		Minimum	Maximum	Minimum	Maximum	
1	29.80	1.591	1.599	0.0500	0.0517	3.4
2	61.10	1.578	1.582	0.1009	0.1034	2.5
3	124.38	1.575	1.585	0.2016	0.2034	0.9
4	374.95	1.582	1.592	0.6350	0.6550	3.1

Table 3. Isotopic Analysis of ^{233}U .

Isotope	ORNL Analysis of $^{233}\text{U}_3\text{O}_8$ (Wt%)	Rocky Flats Analysis of ^{233}U Metal (Wt%)
^{232}U	0.8 ppm	—
^{233}U	99.40	99.47
^{234}U	0.17	0.17
^{235}U	0.07	0.06
^{236}U	0.02	0.01
^{238}U	0.34	0.29

Table 4. Alpha Pulse Height Analysis of ^{233}U .

Isotope	Alpha Energy (MeV)	ORNL Analysis of $^{233}\text{U}_3\text{O}_8$ (%) of total alpha	Rocky Flats Analysis of ^{233}U Metal (%) of total alpha
^{233}U	4.82	91.8	94.1
^{239}Pu and ^{240}Pu	5.15	4.7	3.98
^{238}Pu and ^{241}Am	5.50	2.6	1.76
Other	Other	0.9	0.16

decrease in the levels of non-uranium actinide impurities. This decrease in the level of alpha emitting impurities can probably be explained by volatilization and slagging in the casting operation. The above analysis shows that the integrity of ^{233}U was maintained throughout this project and no cross contamination with other actinide elements or uranium isotopes occurred.

DISCUSSION

The conservation of material was a prime consideration during the planning and performance of this project. Only 700 g of uranium were available, and the finished disks, after machining at ORNL, were to weigh at least 500 g. The cast disks, therefore, had to weigh between 550 and 600 g to provide sufficient material for machining.

Several precautions were taken during the conversion-to-metal operations to maximize the metal yield. The U_3O_8 was calcined prior to hydrofluorination to remove any volatiles and to ensure a reactive oxide. The reduction charges were prepared with 0.2 moles CaCl_2 /mole U to improve the metal yield. The addition of CaCl_2 to reduction charges has been found to have a beneficial effect on metal yields.³

Even with the excellent metal yields, it was doubtful that the required disk weights could be provided if each disk was cast separately. Therefore, it was decided to attempt to

cast the three thinner disks in one mold. The mold used for this casting is shown in Figure 4. This mold was designed so the molten metal could be poured into the holding basin at the top of the mold and then run through the 200-mil disk cavity into the 50 and 100-mil disk cavities.

Figure 4. Multiple-Disk Mold Used for Casting 0.050-, 0.100-, and 0.200-inch-thick ^{233}U Disks.

The casting is shown in Figure 5 as it appeared after removal from the mold but before the disks were separated from one another. As the figure shows, the casting produced three well-formed disks.

Figure 5. Multiple-Disk Casting After Removal From Mold.

The fourth disk was cast using the melting crucible and mold shown in Figure 6. The melting crucible shown is identical to the one used to cast the three thinner disks.

The cylindrical mold was designed with a removable lid, and the metal was poured through the hole in the lid. The

Figure 6. Melting Crucible and Mold Used for Casting 0.575-inch-thick ^{233}U Disk.

mold was designed with no restriction on the height of the disk so that the disk could be cast as thick as the available metal allowed.

There was no excess of metal remaining following the last casting. The majority of the excess of uranium (95 g) was in the form of casting skull which was burned to oxide. The remainder of the uranium (15 g) was contained in the

reduction residues. The cast disks contained 85% of the uranium which was furnished for this project.

Another important consideration was maintaining the integrity of the isotopically enriched ^{233}U during the project. The hydrofluorination furnace used for this project had in the past been used for ^{236}U , ^{235}U , ^{238}U , and various isotopic levels of ^{233}U . The glovebox used for the reduction and casting work had been used for processing various plutonium isotopes and americium-241. By using careful cleaning procedures, it was possible to use these facilities without contaminating the ^{233}U . There were no rigid dimensional tolerances required for this project, but ORNL indicated that the flat surfaces of the disks would not require any machining if the variation in thickness of each disk could be held to $\pm 5\%$ of the average thickness of the disk, or a total variation of 10%. All the disks were well within this specification, with the maximum variation in thickness being 3.4% for the 0.050-inch-thick disk. The variation in the thickness of the 0.200-inch-thick disk was 0.9%. This shows that it is possible to cast to fairly tight dimensional tolerances.

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