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PREPARATION OF ACTINIDE METAL TARGETS USING SPECIAL CASTING TECHNIQUES*

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INTRODUCTION

The fabrication of actinide metal targets for cross-section and other measurements usually begins with the oxide of the metal of interest. The oxide is converted to metal which is fabricated into the desired shape. For several years the Rocky Flats Plant has been involved in the conversion of actinide oxides to metal for target fabrication. The chemical conversion process produces a metal button which is typically 0.250-0.500 in. thick. The fabrication of targets from metal buttons usually involves the rolling of the buttons to the desired thickness, and cutting, punching, or machining the resulting rolled sheet or foil into the desired shape. However, the rolling and forming operations will ordinarily generate large amounts of scrap.

The target materials are usually highly purified both isotopically and chemically. These materials are expensive to prepare and are only available in limited quantities. Therefore, a technique was desired for fabricating targets which would generate less scrap. One method for accomplishing this would be to cast the metal buttons into the desired shape and eliminate the rolling and forming operations.

Various casting techniques and mold designs have been evaluated at Rocky Flats for preparing actinide metal targets. With the proper mold design, a tilt-pour casting technique can be used to cast targets >0.040 in. thick. The tilt-pour technique is not very effective for casting targets <0.040 in. thick. An injection casting technique has been developed for the thinner castings. The casting techniques described in this paper have been used to cast plutonium, neptunium, and uranium metal ingots and disks ranging in thickness from 0.005-0.600 in.

EXPERIMENTAL

Equipment

The casting furnace used to prepare the targets is a System VII General Purpose Metallurgical Facility manufactured by Vacuum Industries (Fig. 6.1). The furnace consists of a tilt-pouring, 4-in.-diam by 6-in.-high water-cooled

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Fig. 6.1. Vacuum Furnace Used for Target Casting.

induction coil enclosed in a water-cooled vacuum chamber. The coil contains a quartz insulator sleeve and a 1/4-in.-thick sleeve of WDF graphite felt insulation manufactured by Union Carbide. The power for the coil is supplied by an Inductotherm 15-kW motor-generator unit. A 3-kW ceramic mold heater can be added to the vacuum chamber if a separate mold heater is required. The mold heater is 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 involved. The vacuum system consists of a 4-in. diffusion pump with a mechanical roughing pump.

The basic mold design used for the target casting work is shown in Fig. 6.2. This is a split mold with a cylindrical melting section which opens into an ingot cavity; the mold is vented from the top of the ingot cavity. This design can be varied depending upon the size and shape of the item to be cast. Molds are fabricated from Stackpole graphite and, for plutonium and neptunium castings, these are coated with a calcium fluoride (CaF_2) mold coating preparation. Zirconium oxide (ZrO_2) mold coating is used for uranium castings. Molds are fabricated with a thermocouple well which extends to within $1/8$ in. of the ingot cavity.

Additional equipment required is dependent upon the thickness of the metal being cast. For castings where the desired thickness is <0.040 in., the only additional equipment required is a tantalum metal rod. For items >0.040 in., a separate melting crucible is used. A tantalum melting crucible is used for plutonium and neptunium castings. The tantalum crucible is heated to 500°C in air for 20 to 30 min to form a protective oxide coating on the surface of the tantalum before the crucible is used. A graphite melting crucible is used for uranium castings. The surfaces of the crucible which contact the uranium metal are coated with ZrO_2 before the crucible is used.

Fig. 6.2. Cross-Section Showing Basic Design of Split Mold Used for Target Casting.

Procedure

The procedure used for the target casting is also dependent upon the thickness of the item to be cast. The procedure for items <0.040 -in. thick is quite different from the procedure used to cast items >0.040 -in. thick. An injection

casting technique is used to cast items from 0.005-0.040 in. thick and a tilt-pour technique is used for thicker castings. The procedure used for the thicker castings will be described first.

Tilt-Pour Casting Procedure

The melting crucible is placed in the induction coil and the mold is placed in the mold heater. A graphite susceptor is used with the tantalum melting crucible for plutonium and neptunium castings. For uranium castings the melting crucible and susceptor are fabricated as a single unit. Metal to be cast is placed in the respective melting crucible and the vacuum chamber is sealed and evacuated to 10^{-4} to 10^{-5} torr. The metal is heated to 200-300°C above its melting temperature, and the mold is heated to the melting temperature of the metal being cast. The molten metal is then poured into the heated mold and allowed to cool under vacuum.

Injection Casting Procedure

A separated melting crucible is not used for those castings having <0.040-in. thickness. Rather the mold is placed in the induction coil and the metal to be cast is placed in the melting section of the mold (see Fig. 6.2). The induction coil is tilted at a 45° angle, and a tantalum metal rod is placed in the upper portion of the melting section of the mold. When the vacuum chamber is sealed and evacuated, the mold is heated to 200°C above the melting point of the metal being cast. The induction coil and mold are then moved to a vertical position, allowing the tantalum metal rod to drop onto the molten metal and force it into the ingot cavity of the mold. The mold is then allowed to cool under vacuum.

RESULTS

Casting procedures have been used to cast plutonium and neptunium ingots and plutonium and uranium disks ranging in thickness from 0.005-0.600 in. Most of the development work has been concentrated on the injection casting technique because thinner targets are more difficult to fabricate. Results obtained from some representative injection castings are given in Table 6.1. Ingots have been formed 1-1.25 in. wide by 2-2-1/2 in. long and range in thickness from 0.006-0.045 in. Visual inspection of the cast ingots has shown no evidence of cracks or voids. Verification of these visual observations has been obtained for several 1-in.-wide by 2-in.-long plutonium ingots by density, hardness, and metallographic examination.

Density and hardness data are given in Table 6.2. The first three ingots shown in Table 6.2 were cast from unalloyed, electrefined plutonium metal. Densities of these ingots varied from 19.54 to 19.62 g/cm³, which is 1.2 to 1.6% below the theoretical density of 19.86 g/cm³ (ref. 1) for alpha plutonium. However, the density of cast plutonium metal is usually 1 to 2% lower than theoretical. This is attributed to retention of small amounts of various high temperature phases, or in the case of high purity metal, to the formation of microvoids during the beta to alpha phase transformation.¹ The diamond pyramid hardness of the ingots varied from 252 to 260 which is normal for alpha plutonium. The metallographic examination of the ingots did not reveal any cracks or voids other than the microvoids associated with cast, high-purity plutonium metal.

The last two ingots shown in Table 6.2 were cast from plutonium metal alloyed with 1.2 wt % (10.7 at. %) aluminum to stabilize the plutonium in the delta phase. The densities of these ingots were 14.91 and 15.09 g/cm³, respectively. This compares to a density of 15.01 ± 0.03 g/cm³ for fully homogenized delta-phase plutonium stabilized with 10 at. % aluminum.² The diamond pyramid

hardness (DPH) of the ingots varied from 61.0 to 61.8, which compares to a DPH of 45 for fully homogenized delta plutonium stabilized with 1 wt % gallium.

Table 6.1. Results Obtained Using Injection Casting Technique

Nominal Thickness of Ingot Cavity (in.)	Type of Feed	Ingot Weight (g)	Thickness of Ingot (in.)	Ingot Size (in.)
0.045	Np	39.0	0.0430 ± 0.001	$1\text{-}1/4 \times 2\text{-}1/2$
0.035	Delta Pu	30.2	0.0383 ± 0.0005	$1\text{-}1/4 \times 2\text{-}1/2$
0.035	Alpha Pu	32.5	0.0366 ± 0.0009	$1\text{-}1/4 \times 2\text{-}1/2$
0.033	Np	19.4	0.0390 ± 0.0010	1×2
0.022	Delta Pu	9.7	0.0230 ± 0.0005	1×2
0.022	Alpha Pu	11.3	0.0224 ± 0.0003	1×2
0.006	Delta Pu	3.4	0.0069 ± 0.0007	$1\text{-}1/4 \times 2\text{-}1/2$
0.006	Alpha Pu	2.6	0.0056 ± 0.0004	1×2

Table 6.2. Hardness and Density Data

Sample Number	Type of Sample	Density (g/cm ³)	Diamond Pyramid Hardness
1	0.035-in. alpha Pu ingot	19.62	259.7
2	0.006-in. alpha Pu ingot	19.54	252.5
3	0.006-in. alpha Pu ingot	19.62	257.8
4	0.022-in. delta Pu ingot (1.2 wt % Al)	14.91	61.0
5	0.022-in. delta Pu ingot 1.2 wt % Al) ^a	15.09	61.8

^aEquivalent to 10.7 at. % aluminum.

One concern with the injection casting technique was possible contamination of the metal with CaF₂ mold coating and graphite scraped from the walls of the mold by the tantalum metal rod. A sample of the feed metal used to cast the first ingot shown in Table 6.2 and a sample of the ingot were analyzed by spark source mass spectrography for impurities. The results of these analyses are shown in Table 6.3. There was very little pickup of calcium and fluorine from the CaF₂ mold coating and the carbon content was lower in the ingot than in the feed metal. The feed contained 195 ppm total of detectable impurities compared to the ingot which contained a total of 187 ppm of detectable impurities.

Table 6.3. Spark Source Mass Spectrographic Impurity Analysis

Element ^a	Concentration in Feed (ppm)	Concentration in Ingot (ppm)
Al	2.00	4.00
B	0.03	0.06
Be	0.02	0.07
C	56.00	36.00
Ca	0.20	3.00
Cl	3.00	4.00
Co	0.30	0.40
Cr	0.70	1.00
Cu	0.60	1.00
F	3.00	12.00
Fe	7.00	12.00
Ga	0.60	2.00
K	2.00	2.00
Mg	1.00	4.00
Mn	0.20	0.80
N	1.00	1.00
Na	1.00	4.00
Ni	0.70	5.00
Np	4.00	12.00
P	0.20	0.20
S	0.30	0.50
Sc	0.10	0.20
Si	3.00	7.00
Ta	17.00	17.00
Ti	0.10	0.10
V	5.00	14.00
W	76.00	31.00
Zn	6.00	10.00
Total	195.00	187.00

^aOnly detectable impurities are listed.

DISCUSSION

Tilt-Pour Casting

The tilt-pouring technique described for >0.040-in.-thick castings is a standard casting procedure. However, while the procedure is not unique, the limited amounts of material available for most target preparation work necessitates the development of specialized molds for each casting. The need for specialized mold development is illustrated by the target fabrication project described below.

The Oak Ridge National Laboratory required four ²³³U metal disks having a diameter of 1.50±0.005 in. Thicknesses of the disks were 0.050, 0.100, 0.200, and ≥0.575 in. Cast disks were to be at least 1.55 in. in diameter, to provide sufficient excess material for final machining at ORNL.

Only 700 g of uranium (99.4% ²³³U) as U₃O₈ was available for this project, and the finished disks were to weigh at least 500 g. The three thinner disks were cast first, and all the remaining metal was cast into the thick disk. The

limited amount of material available for this project made it doubtful that sufficient metal would be available for casting the thick disk if the three thinner disks were each 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 Fig. 6.3.

Fig. 6.3. Multiple-Disk Mold Used for Casting the 0.050-, 0.100-, and 0.200-in.-Thick ^{233}U Disks.

The U_3O_8 was converted to metal by hydrofluorination of the oxide to UF_4 which was reduced to metal. The conversion process yielded 686 g of metal for a 98% metal yield. A portion of this metal, 484 g, was heated to 1400°C and poured into the heated mold. The casting produced three well-formed disks as shown in Fig. 6.4. This figure shows the casting after removal from the mold but before the disks were separated from each other.

The disks were separated and all the remaining metal was cast into the thick disk using the melting crucible and mold shown in Fig. 6.5. This disk was 0.640 in. thick, which was over 0.050 in. thicker than the required minimum of 0.575 in. Weights and dimensions of the four disks are given in Table 6.4. The total weight of the four disks was 590 g, which was more than sufficient to provide the required 500 g of uranium in the finished product after machining. Maximum variation in the thickness of the four disks was 3.4%.

Fig. 6.4. Multiple-Disk Casting After Removal from Mold.

Fig. 6.5. Melting Crucible and Mold Used for Casting the 0.575-in.-Thick ^{233}U Disk.

Table 6.4. Weights and Dimensions of ^{233}U Disks

Disk Number	Disk Weight (g)	Thickness (in.)		Variation in Thickness (%)
		Minimum	Maximum	
1	29.80	0.0500	0.0517	3.4
2	61.10	0.1009	0.1034	2.5
3	124.38	0.2016	0.2035	0.9
4	374.95	0.6350	0.6550	3.1

Injection Casting

Most of target fabrication work involved preparation of targets of <0.040-in. thickness. The tilt-pour casting technique is not very effective for these thinner castings because the metal will not flow of its own accord into the thin cavities. Also, the tilt-pour casting technique usually requires 40-50 g of excess metal. The Los Alamos Scientific Laboratory has cast 0.010-in.-thick plutonium metal disks by vibrating the mold as the molten metal is being poured.³ The technique developed at Rocky Flats involves injection casting using a tantalum metal rod. The tantalum rod is dropped onto the molten metal, forcing it into the ingot cavity of the mold.

The mold and rod used to cast a 1-1/4- by 2-1/2-in. ingot and the 0.035-in. delta-phase plutonium ingot are shown in Fig. 6.6. The melting section of this mold was 0.875 in. in diameter and the diameter of the tantalum rod was

Fig. 6.6. Ingot Mold Shown with 0.035-in. Delta Phase Plutonium Ingot.

0.867 in. Clearance between the walls of the melting section of the mold and the tantalum rod is very important. If the diameter of the rod is too large, the rod will not slide in the melting section and the necessary force will not be applied to the molten metal; if the rod diameter is too small, metal will be forced up between the rod and the walls of the melting section.

When the development work on the injection casting technique was initiated, one of the objectives was to develop a procedure which required a minimum amount of feed metal. The mold shown in Fig. 6.6 only requires 15-20 g of excess metal for casting 3-40 g ingots. Charge weights required for ingots of various thicknesses using this mold are shown in Table 6.5.

Table 6.5. Charge Weights Required for 1-1/4- by 2-1/2-in. Plutonium Metal Ingots

Nominal Thickness of Ingot Cavity (in.)	Charge Weight Required (g)
0.006	20
0.010	25
0.022	30
0.035	45
0.045	55

Dimensional tolerances obtainable with the injection casting technique can be illustrated by describing the preparation of three ^{242}Pu target disks for the Los Alamos Scientific Laboratory. LASL requested three disks 1.00 ± 0.010 in. in diameter and 0.010, 0.040, and 0.178 in. thick. The actual thickness of the disks was not critical, but the variation in the thickness of each disk was to be within 3% of the average thickness. This gave an allowable variation in thickness of 0.0003 in. for the 0.010-in. disk.

The 100 g of plutonium (98% ^{242}Pu) for this project was supplied as PuO_2 by the Lawrence Livermore Laboratory. The PuO_2 was converted to metal by fluorination of the oxide to form PuF_4 which was reduced to metal. The conversion process produced a 98.8% metal yield. Disks were cast in molds similar to the ingot mold shown in Fig. 6.6 except the rectangular cavity was replaced with a circular cavity. The 0.178-in.-thick disk was cast using the tilt-pour technique and the 0.040 in. and 0.010-in. thick disks were cast using the injection casting technique. Three disks are shown in Fig. 6.7 as they appeared just prior to shipment to LASL. The weights and dimensions of the disks are shown in Table 6.6. The cast disks met all specifications. All disk diameters were within 1.00 ± 0.010 in., and the variation in thickness of the three disks ranged from 0.4 to 2%. The thickness of the 0.010-in.-thick disk varied from 0.0096-0.0098 in., which is a variation of only 0.0002 in.

Table 6.6. Weights and Dimensions of ^{242}Pu Disks

Disk	Disk Weight (g)	Diameter (in.)		Thickness (in.)		Variation in Thickness (%)
		Minimum	Maximum	Minimum	Maximum	
1	34.83	0.9977	1.0038	0.1765	0.1772	0.4
2	7.90	0.9940	1.0037	0.0405	0.0409	1.0
3	1.80	0.9905	1.0005	0.0096	0.0098	2.0

Fig. 6.7. ^{242}Pu Target Disks Prepared for the Los Alamos Scientific Laboratory

REFERENCES

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2. O. C. Wick, p. 43.
3. Delbert Harbur, Los Alamos Scientific Laboratory, private communication.