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**The Recovery of Mechanically
Entrapped Plutonium From Casting
Skulls and Turnings***

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ABSTRACT

The small critical mass of plutonium which limits charge size, coupled with its value necessitating quantitative recovery from all residues, makes the direct recovery of mechanically entrapped plutonium from casting skulls and oxidized turnings an economic necessity. The newest method used at the Los Alamos Scientific Laboratory for the direct recovery of entrapped plutonium involves mechanical stirring and vibration of the plutonium melt to physically break up the spongy skull and oxide film, thus allowing the heavier molten plutonium to settle out at the bottom of the melt. The recovery equipment and procedures are described.

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INTRODUCTION

Plutonium has a high density, a low melting point, and an unprecedented six allotropic modifications between ambient temperature and its melting point. Molten plutonium is extremely reactive, and even the solid metal is subject to rapid oxidation in moist atmosphere.

Plutonium is a fissile material⁽¹⁾ with a low critical mass. For normal foundry operations the Nuclear Safety Committee of the Los Alamos Scientific Laboratory has established a maximum charge size for plutonium rich materials of 6 kg. (A bare sphere of plutonium is ideally critical at 10 kg. and a specific configuration of water and plutonium is critical at a plutonium mass of 510 g.).⁽²⁾

At Los Alamos, plutonium is vacuum cast by the bottom pour method.⁽³⁾ The high density of plutonium (19.8 g./cc) gives this method a distinct advantage, since the oxides and impurities rise to the top of the melt and remain behind in the casting skull. When the oxides and lighter impurities rise to the top of the melt, some good plutonium metal is carried along and mechanically entrapped in the casting skull.

A method for directly recovering this skull entrapped metal as well as a more efficient means of making casting feed out of oxidized turnings has been developed and is now being used. This report describes the process and equipment used in this recovery operations.

ADVANTAGES OF DIRECT RECOVERY

Depending upon the purity of the plutonium which is recovered from skulls and turnings, the metal can be used as recast feed or as electro-refining feed.⁽⁴⁾ The reduction in the quantity of plutonium skulls and turnings which are sent through the aqueous recovery operation is of prime economic concern. The aqueous recovery operation is limited in this Laboratory to batch sizes of only 360 g. of plutonium, due to the low critical mass of dissolved plutonium.⁽²⁾ This necessarily makes the aqueous recovery of plutonium a costly and time-consuming operation.

Since the advent of plutonium foundry operations at the Los Alamos Scientific Laboratory, several different methods have been used to directly recover the good plutonium metal from turnings and casting skulls without resorting to the costly aqueous recovery operation. The method previously used for recovery of skull entrapped metal was to "milk the skull". This was done by simply reheating the skull to a relatively high temperature in a pour crucible. The temperature used was around 1200°C, whereas plutonium melts at 640°C. This increased the fluidity of the entrapped plutonium and did indeed allow some of it to flow out of the spongy skull. One adverse affect of this operation was that the solubility of impurities, which are already present in the skull, is increased at the higher temperatures and the recovered plutonium is quite impure.

Several years ago the plutonium foundry began recovering plutonium from turnings by simply melting and bottom pouring the compacted turnings from a pour crucible.^(3,5) Recovery yields of 90% were not uncommon when unoxidized turnings were used as feed material. A large reduction in recovery yields was realized, however, when the turnings became oxidized. This

reduction in recovery yields can be attributed to two separate causes. The plutonium oxide reduces the percentage of plutonium available for recovery, and it also entraps good plutonium which rises to the casting skull with the lighter plutonium oxide.

The latest method which has been developed at Los Alamos Scientific Laboratory for the direct recovery of entrapped plutonium from casting skulls and oxidized turnings, involves the use of a mechanical stirrer to physically break up the casting skull and oxide film which forms on the turnings, and a mechanical vibrator connected to the melt crucible to help shake loose any entrapped plutonium remaining in the final casting skull.

MECHANICAL STIRRER AND CRUCIBLE VIBRATOR

The mechanical stirrers which are used at Los Alamos Scientific Laboratory are shown in Figs. 1 & 2⁽⁶⁾ Generally the perforated flat disk-shaped stirrer is used for recovery operations, whereas a contoured dash pot stirrer which also works satisfactorily is used for alloying operations. The stirrers are made out of tantalum, which is one of the few materials not severely attacked by molten plutonium.

Figs. 3 and 4 show the stirrer mounted in a high vacuum casting furnace. The stirrer is connected to an extension rod which is moved vertically through a vacuum seal. The recovery melt is stirred in a solid bottom tantalum melt crucible.

The crucible vibrator consists of a tantalum sleeve welded to the bottom of a tantalum pour crucible. Two gas-operated vibrators are strapped to the tantalum tube as shown in Fig. 5. Operating the vibrators at 60 psi gives a vibrational amplitude of approximately 0.015 in. and a vibrational frequency of 160 cycles/sec., depending upon the amount of charge. The vibrators are equipped with both inlet and outlet gas lines, so that the assembly can be used in vacuum.

RECOVERY PROCESS

The process for recovering plutonium from skulls and turnings consists of two operations. In the first operation the skulls and turnings are packed into a solid-bottom tantalum melt crucible and then they are melted and stirred at 750-800°C. The stirring is accomplished by moving the tantalum stirrer vertically (driven by an externally mounted air cylinder) through the melt at 10-30 strokes/min. During the down stroke, the stirrer breaks up and pushes the light oxide and impurity layers down into the melt. Occasionally during the run some of the light skull collects on top of the stirrer. Spinning the stirrer allows the skull to drop back down below the stirrer where it can be further broken up and pushed back into the melt on the next down stroke. A stirring time of 15 minutes is usually sufficient to obtain a completely mixed solid ingot.

During the second operation of the recovery process, the stirred ingot is placed in the crucible vibrator, melted, and bottom poured. The vibrators are left on during and after the pour. The size of the resultant skull determines the length of time that the vibrators are left on after the pour. If the resultant skull is relatively large, then the vibrators should be left on longer in order to shake loose the larger amount of entrapped plutonium.

Before the advent of the crucible vibrator, the second operation of the recovery process consisted of simply placing the stirred ingot in a pour crucible, melting, and bottom pouring the melt.

RESULTS AND DISCUSSION

The purity and amount of oxidation present in the recovery feed has a great influence upon how much advantage is gained by stirring and vibrating the plutonium melt. As an example, when 3.2 kg. of clean (unoxidized) turnings were recovered by simply melting and bottom pouring the turnings, a recovery yield of 91% was obtained. When 3.2 kg. of identical turnings were stirred, remelted, and bottom poured a recovery yield of 95% was obtained. This is a gain of only 4%. When the same turnings were allowed to become slightly oxidized recovery yields of 82% and 89% were obtained. The advantage gained from stirring had gone up to 7%. In general the more oxidized the turnings become, the more advantageous the stirring becomes. In like manner, the more entrapped metal which remains after the stirring process the more advantage will be gained by vibrating the charge during the pour.

One important aspect of stirring recovery feed is that the melt will take on a homogeneous smooth texture when the stirring is completed. This is true when either or both oxidized turnings or skulls are recovered.

The plutonium compounds which make up most of the skull are broken up and pushed down into the molten plutonium. Thus there is the case of a small piece of a plutonium compound completely surrounded by molten plutonium. The plutonium compounds are assimilated in the surrounding molten plutonium. The recovered plutonium will be less pure due to stirring, however, the plutonium which is present in the skull as either entrapped metal or as plutonium compounds is recovered in a useable metallic form. Tables 1 and 2 give typical chemical analyses of batches of recovered plutonium.

Most of the insoluble plutonium oxide which is present in the recovery melt is held in suspension by the stirring. The suspended plutonium oxide does float to the top of the melt when the stirred ingot is remelted and bottom poured, and thus stays behind in the skull. Most of the plutonium which is entrapped by this rising plutonium oxide is recovered by continued vibration of the skull during the pour.

From the above discussion of the recovery process, it is clear that a recovery batch should not be made up entirely of skulls. In practice if the skulls are held to less than 35% of the recovery feed, good results can be obtained. For skulls and turnings recovery, an increase of 28% yield was realized by vibrating a stirred ingot as opposed to straight pouring of the skulls and turnings. When skulls and turnings are properly recovered by the process described in this report, recovery yields of 90 to 96% can be consistently obtained. Table 3 gives some data on recovery feeds where the feed varies from unoxidized turnings to badly oxidized skulls and turnings.

Table 1

Chemical Analysis of
Plutonium Recovery Lot No. JUO-1267
(Stirred & Vibrated during Pour)

Pu ^a	99.81%	Bi	< 1
Li ^b	< 0.2	Co	<10
Be	0.2	Zn	<10
Na	<10	Ta	<35
Mg	<10	Th	<15
Ca	<10	Mo	3.0
Al	55	W	16.5
La	<10	Zr	<10
Si	80	Fe	320
Pb	5	U	50
Cu	10		
Ni	160		
Cr	50		
B	1		
Mn	15		
Sn	< 1		

a. Pu is reported as weight percent.

b. This and the remaining elements are reported as parts per million (ppm) on a weight basis.

Table 2

Chemical Analysis of
Plutonium Recovery Lot No. JUO-1245
(Stirred & Vibrated During Pour)

Pu ^a	99.89%	Bi	< 1
Li ^b	<0.2	Co	<10
Be	0.5	Zn	<10
Na	110	Ta	<35
Mg	<10	Th	<15
Ca	50	Mo	3.5
Al	250	W	9
La	<10	Zr	12
Si	70	Fe	330
Pb	5	U	60
Cu	25		
Ni	100		
Cr	55		
B	<0.5		
Mn	10		
Sn	1		

a. Pu is reported as weight percent.

b. This and the remaining elements are reported as parts per million (ppm) on a weight basis.

Table 3

Data on Recovery Yields

<u>Lot No.</u>	<u>Charge (g)</u>	<u>Recovered Ingot (g)</u>	<u>Recovery Yield (%)</u>	<u>Condition of Charge</u>	<u>Process</u>
JUO 1104	3215	2932	91%	Unoxidized turnings	Straight pour
JUO 1105	3215	3143	95%	Unoxidized turnings	Stirred and poured
JUO 1127	2901	2374	82%	Slightly oxidized turnings	Straight pour
JUO 1129	2880	2566	89%	Slightly oxidized turnings	Stirred and poured
JOO 1214-1	3323	1204	36%	Badly oxidized skulls and turnings	Straight pour
JOO 1214-2	3304	2110	64%	Recovered ingot and skull from JOO 1214-1	Vibrated during pour
JUO 1291	4725	4470	95%	Slightly oxidized skulls and turnings	Stirred, vibrated during pour
JUO 1299	4771	4409	92%	Slightly oxidized turnings	Stirred, vibrated during pour

Fig. 1
Tantalum Stirrer

0 1 2 3 4 5 6 7 8 9 10
LASL CMB II

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Fig. 2
Mechanical Stirrer Unit

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Fig. 3
Stirrer Mounted In High Vacuum Furnace

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FIG. 4 CROSS SECTION OF STIRRER MOUNTED IN HIGH VACUUM FURNACE

Fig. 5
Crucible Vibrator

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