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FABRICATION OF PLUTONIUM INGOTS FROM PLUTONIUM TURNINGS

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ABSTRACT

Kilogram quantities of delta-stabilized and pure plutonium turnings can be cast directly into ingots of normal quality with high yields. This report describes the equipment and process used.

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INTRODUCTION

Metal fabrication of plutonium shapes by machining results in the production of metal turnings which must be collected and processed into reusable metal. This recycle can be accomplished by one of four methods.

1. Reduction: Clean surface turnings of short length are added to the reduction charge in the metal production operation. A yield of 99 percent is achieved; however, there is a limit to the quantity of turnings which can be processed depending on the metal production rate, since there is a fixed maximum allowable charging ratio of turnings to plutonium tetrafluoride.

2. Hydrofluorination and Reduction: Clean surface turnings of short length are hydrofluorinated to form plutonium tetrafluoride, which is then reduced in the standard metal production manner. A yield of 97 percent results; however, although the hydrofluorination step is usually controllable, it can be hazardous, and furthermore, 3 percent of the plutonium throughput is diverted into slag, crucible and other residues.

3. Dissolution, Purification, Hydrofluorination and Reduction: Turnings are dissolved in a nitric-hydrofluoric acid mixture and the resulting nitrate solution is processed through the metal production operation in the standard manner employing a peroxide (or oxalate) precipitation for purification and production of a product which is hydrofluorinated, and reduced to metal. Metal of excellent quality is produced; however, with an over-all yield of 95 percent, plutonium is diverted into dissolution, purification and reduction residues. The process is time-consuming and relatively expensive.

4. Briquetting: Clean surface turnings are briquetted in a steel die; the briquet along with the other metal is melted in a magnesia pouring crucible, and the melt is drip-cast into the desired mold. A yield of 85 percent and a product of poor quality is obtained.

An investigation of the direct casting of turnings into a usable shape as a more economical method of recovery was undertaken.

Background experiments showed that plutonium turnings could be melted into a fused mass, but one of below average density and of poor physical appearance. (This can be attributed to plutonium oxide inclusion in the mass, due to the large surface area of the turnings and the tendency of plutonium surfaces to oxidize readily.) Subsequent drip-casting of the fused mass produces an ingot of average density and good quality.

A vacuum casting apparatus was, therefore, assembled which permitted controlled melting of turnings and pouring of the melt in the same equipment.

EQUIPMENT

Furnace Assembly

Figures 1, 2 and 3 show the various components of the casting furnace assembly. The assembly consists of a brass can and base plate, a quartz furnace tube, and a brass cover plate. Vacuum seals are obtained with neoprene gaskets and O-rings. The water-cooled vacuum can is 9 in. in diameter, 12 in. high and contains a vacuum outlet, O-ring groove and bolt circle. A base plate with a 3.25 in. diameter center hole and gasket groove for the quartz furnace tube is bolted to the vacuum can. Both ends of the 4 in. diameter, 12 in. high quartz furnace tube are flared slightly out and ground flat. The cover plate is water-cooled and contains a gasket groove for the quartz furnace tube, thermocouple fittings and a 0.25 in. diameter Wilson seal for the stopper-rod adapter.

Vacuum

Vacuum is obtained with a Model UMF-20-04 Consolidated Vacuum Corporation diffusion pump backed by a Megavac mechanical pump. A standard thermocouple gage is used to measure vacuum.

Heating

Power is supplied to a water-cooled induction coil by a 20 K.W. Ajax Spark-Gap Converter. The induction coil is 5 in. in diameter, 5.5 in. high and contains 21 turns of flattened 3/8 in. diameter copper tubing. Temperature measurements are made using a platinum, platinum-rhodium thermocouple with a Brown recorder.

Ceramics

Type V-2 magnesia pouring crucibles fitted with a 0.375 in. diameter pouring hole and countersunk for a hollow stopper rod are used exclusively.

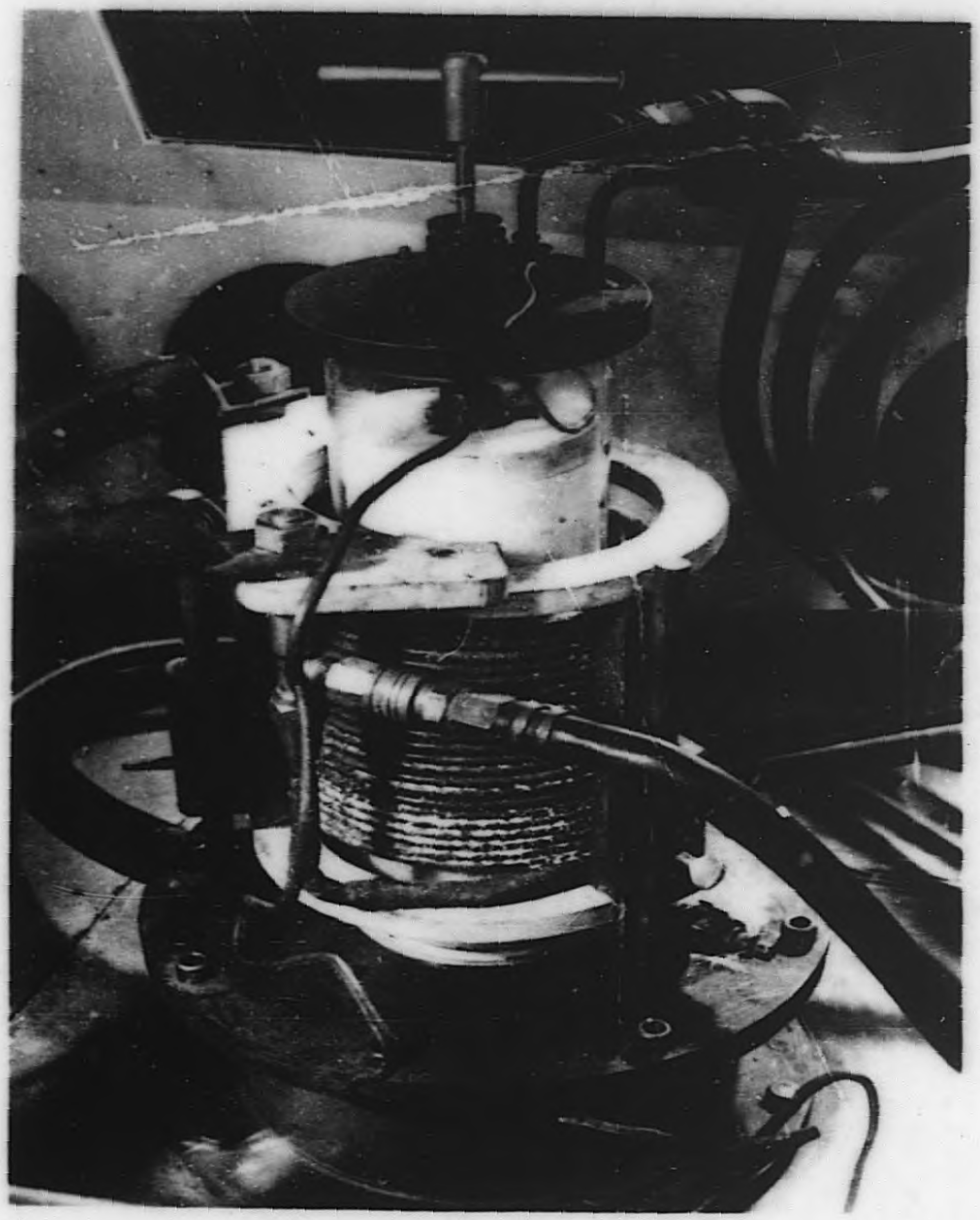

Fig. 1. Assembled Casting Unit.

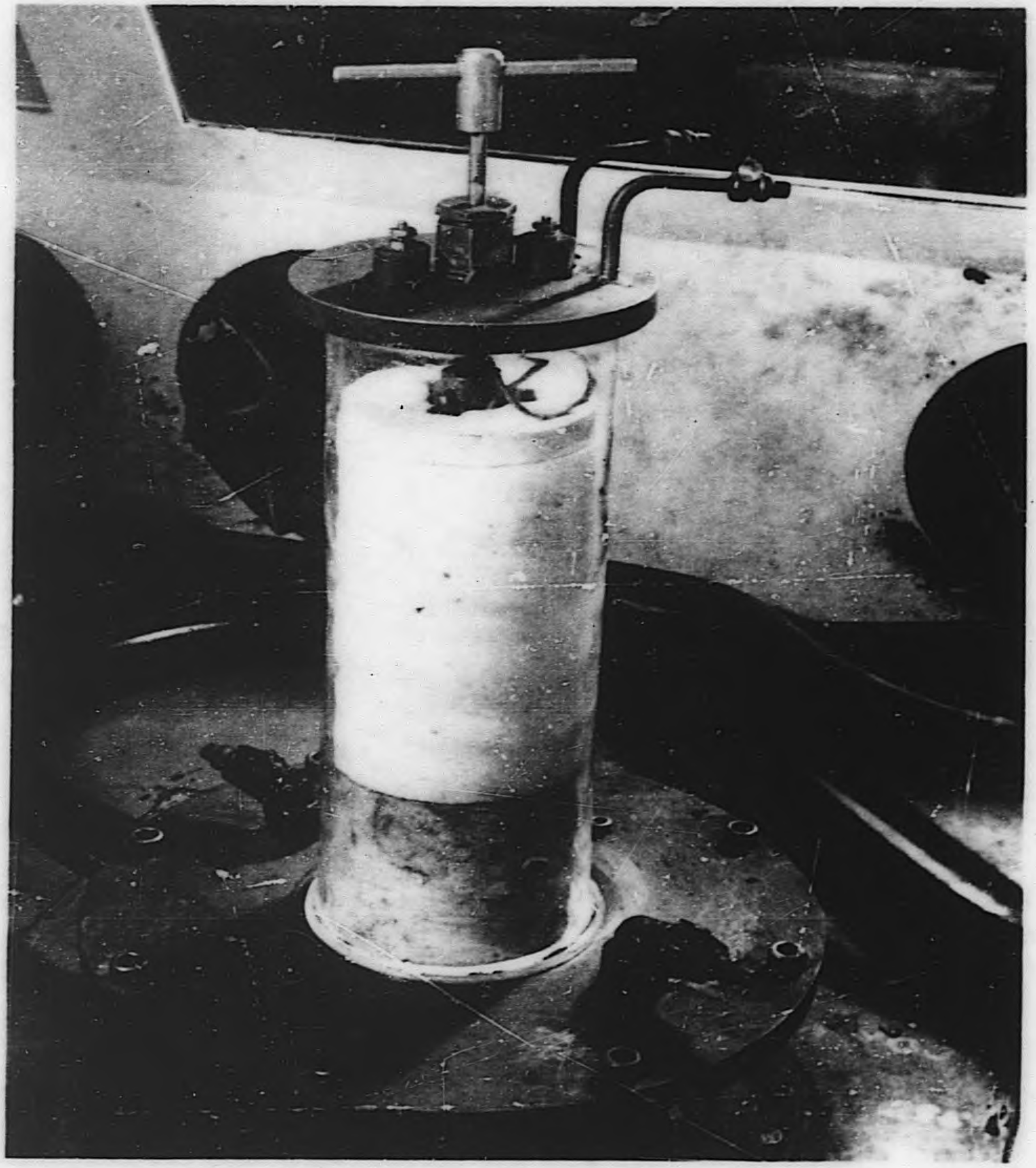

Fig. 2. Vacuum Assembly. The base of the induction coil was placed even with the base of the pouring crucible.

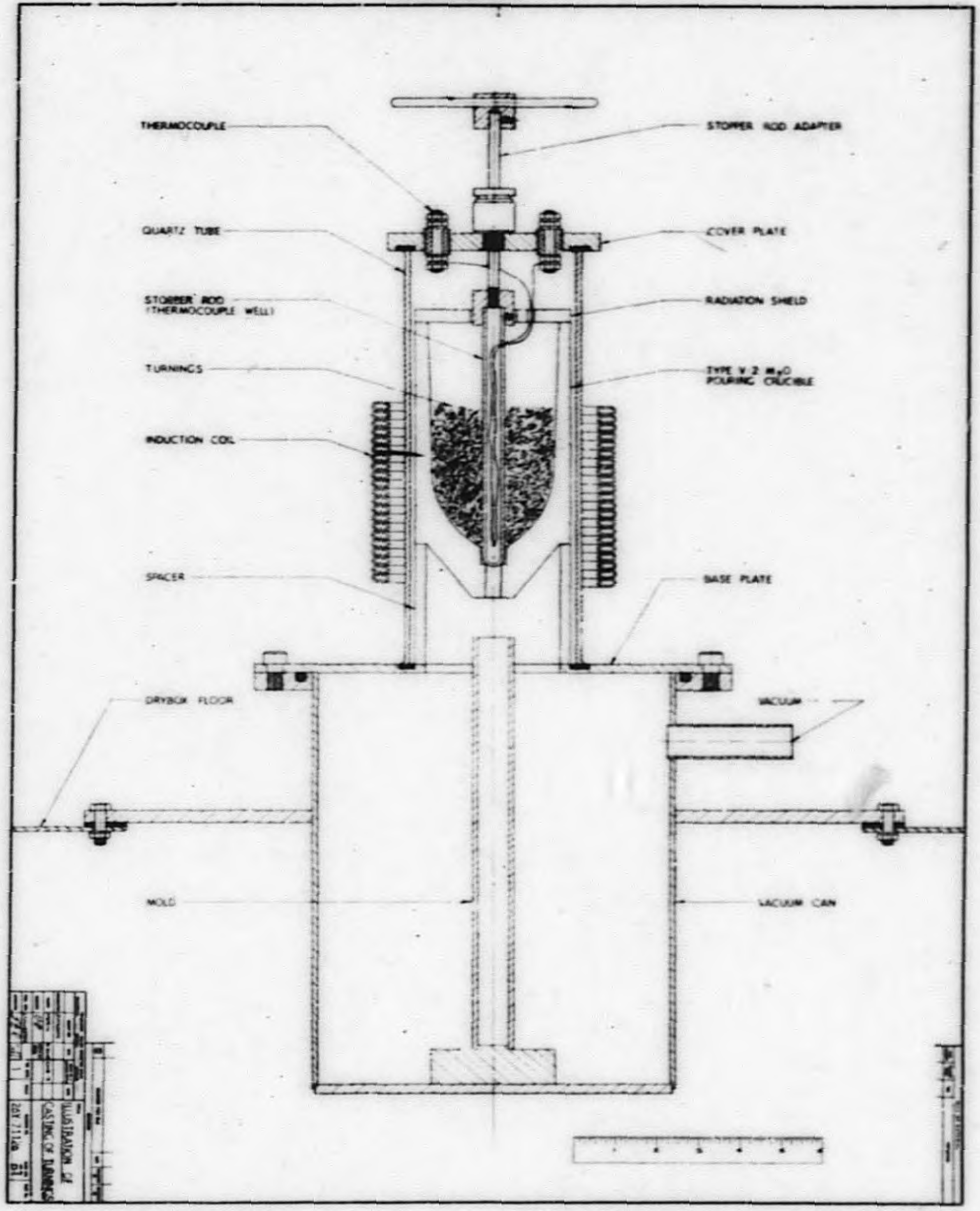

Fig. 3. Cross Section of Casting Unit.

The magnesia stopper rod is 0.625 in. in diameter, 7.5 in. high and also serves as a thermocouple well.

Molds

Pure plutonium metal is chill-cast into a mold constructed of thick-walled copper tubing silver-soldered to a copper base. The copper tubing is 1 in. in diameter, has a wall thickness of 0.125 in. and is 12 in. high. The copper base is 3 in. in diameter and 1 in. thick. Delta-stabilized plutonium is chill-cast into type A-263 magnesia crucibles 3 in. in diameter or into a split-copper mold which is 1 in. in diameter and 13.5 in. high with a wall thickness of 0.75 in. (Fig. 4).

CASTING PROCEDURE

The casting procedure used in this investigation is as follows:

The stopper rod is placed in the countersunk hole of the pouring crucible and held in place while turnings are added. ("As-machined" turnings were used throughout the investigation, and no effort was made to remove any oxide film.) Loosely packed turnings are tamped to facilitate larger charges.

The casting apparatus is assembled as shown in Fig. 1 and the vacuum is reduced to less than 1 micron.

The turnings are heated to a temperature in the vicinity of 1000°C and the stopper rod is pulled, allowing the melt to pour. Heating rates are controlled to maintain a vacuum of better than 50 microns.

Castings are allowed to cool at least four hours before the apparatus is disassembled.

Figure 5 is a time-temperature recording of a typical run.

DISCUSSION

The object of this investigation was to determine the feasibility of the direct casting of turnings into a usable metal ingot. Experiments were designed to determine the conditions and equipment necessary to produce sound castings of standard purity.

Three casting techniques were investigated. The data are summarized in Table 1. In all cases the turnings (and in one method, the turnings with the skull from the preceding run) were melted and the melt was heated to from 200 to 400°C above the melting point. The method of treatment for the melt was as follows:

Fig. 4. Molds. From left to right: Type A-263 MgO crucible, split copper mold, copper tubing mold.

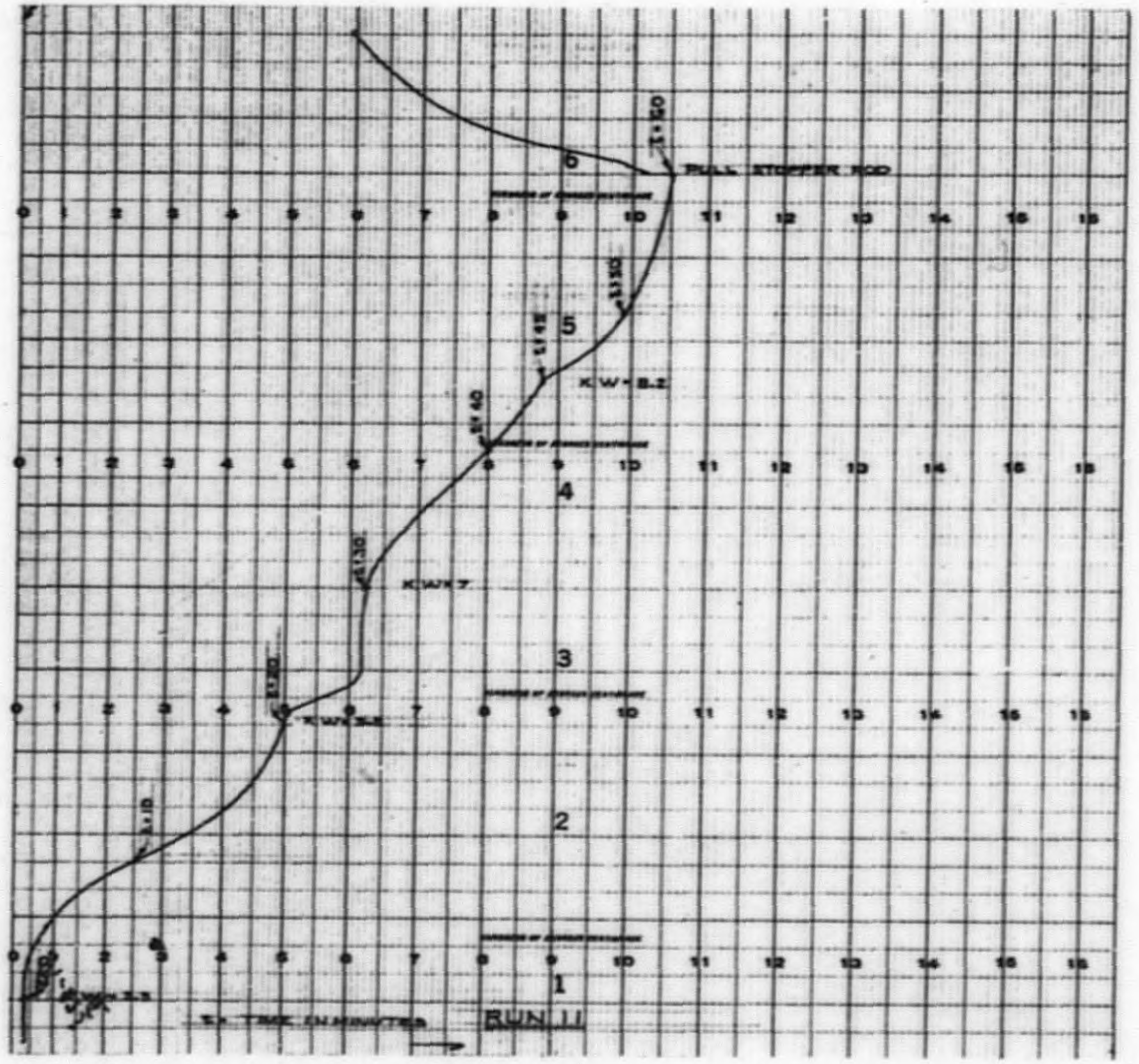

Fig. 5. Time-Temperature Recording.

TABLE 1

SUMMARY OF CASTING DATA

Run No.	Mold	Composition	Max. Temp. (°C)	Pouring Temp. (°C)	Heating Time (min.)	Total Charge (g.)	Clean Turnings (g.)	Product Wt. (g.)	Recovery Yield (%)	Adjusted Yield (%)	Density (g./cc.)
1	Copper	Pure	1000	c	44	1288	1288	1265	---	---	19.252
2	Copper	Pure	900[b]	900	24	1265[d]	1265	1038	82.05	80.58[e]	19.383
3	MgO	1%[a]	1005	850	45	937	937	611	65.19	---	15.937
4	MgO	1%	1000	1000	60	1002	1002	779	77.77	---	15.779
5	MgO	1%	960	860	70	996	996	842	84.46	---	15.782
6	MgO	Pure	1020	1015	90	1559	1559	1154	74.04	---	18.441
7	MgO	1%	890	c	75	956	956	952	---	---	15.845
8	MgO	1%	900[b]	900	480	952[d]	952	848	89.06	88.72[d]	15.810
9	Copper	1%	1015	1015	50	1449	1449	1084	74.81	---	15.819
10	Copper	1%	1045	1045	65	1423	1058	992	69.68	93.72[e]	15.815
11	Copper	1%	1050	1050	60	1304	873	831	63.72	95.23[f,g]	15.850

a. Plutonium was stabilized in the delta phase as a 1 wt. % gallium alloy.

b. Metal was drip-cast in a resistance furnace.

c. Product was not poured.

d. Charge was composed of the product from the previous casting.

e. Yield was based on the initial charge of the previous casting.

f. Yield was based on the clean turnings used in the charge.

g. The over-all yield for Runs 9, 10 and 11 was 86.00% recovery.

1. Direct: Chill-casting (the turnings alone) into a mold (Run Nos. 3, 4, 5, 6 and 9).

2. Successive-Direct: Chill-casting (the turnings plus the preceding casting skull) into a mold (Run Nos. 10 and 11).

3. Fused-Mass: Cooling (the turnings alone) into a fused mass, transferring the mass into a pouring crucible, heating to 200 to 400°C above the melting point and drip-casting into a mold (Run Nos. 1-2, 7-8).

Charges varied in size from 937 to 1559 grams, depending only upon the amount of feed material available at the time. A correlation between the charge size and the yield was not evident over the range investigated.

The charge, which was loaded into the magnesia melting crucible, consisted either of loosely packed or pre-cut turnings. The addition of pre-cut turnings was a handling convenience. Loosely packed turnings were tamped into the crucible to increase the volumetric density. No differences in melting properties were apparent between the two loading methods. Time-temperature recordings revealed a close similarity between the melting characteristics of these charges and those of button charges.

The charge was heated by means of an induction coil in all runs except Run No. 8, which employed resistance heating to remelt and drip-cast the fused mass created by Run No. 7.

Since the rate of heating was determined by the amount of out-gassing of the turnings, oxidized turnings required much lower heating rates. A tantalum sheath, which served as a secondary heater, was secured to the lower portion of the stopper rod in Run Nos. 9, 10 and 11. This allowed elevated temperatures to be reached more quickly and reduced the temperature gradient within the crucible; however, the net effect of this method on yield appeared to be nil.

The most pronounced heating effect on yield was the length of time the charge was held molten before it was poured. Runs 3, 4 and 5, each of which was heated to a melt at the same rate, indicate that the yield increased as the molten-heating time was lengthened.

Previous experience indicated that the production of a sound casting is best achieved through the use of chill-cast molds. Since the melt was poured at 200 to 400°C above its melting point, it was not necessary to pre-heat the molds. Copper and magnesia molds were used in these experiments.

The Fused-Mass Method (Run Nos. 1-2 and 7-8), in which the turnings were melted into a fused mass and then poured by a second melting step, gave the best yield for a single casting (89.06 percent). However, this method is time-consuming and the yields produced are not high enough to warrant consideration.

The Direct Method (Run Nos. 3, 4, 5, 6 and 9), in which turnings are melted and chill-cast in the same operation, also has the limitation of poor yield. The highest yield obtained was 84.46 percent in Run No. 5.

Successive-Direct Method (Run Nos. 10 and 11), in which turnings and the pouring skull from the preceding run are melted and chill-cast together, gave yields of 93.72 and 95.23 percent, based on the weight of turnings added in the charge. A consideration of the over-all process of successive castings indicates that nearly all of the turnings added after the initial casting are converted into solid metal. As a consequence, the over-all yield gradually increases with each successive run.

The only limitation of such a process would be the gradual build-up of impurities in the ingot product. The analyses of ingots produced by the Successive-Direct Method are given in Table 2. Many additional castings may be made before impurities would materially affect the product. The analysis of the ingot from Run No. 9 is also shown in Table 2. Analyses of other castings are comparable to those tabulated.

Density determinations, shown in Table 1, and radiographic examination indicated that the castings were sound without exception. The castings made were machined to desired shapes (Figs. 6 and 7), and served as satisfactory feed in the metal-fabrication operation.

SUMMARY AND CONCLUSIONS

Both pure and delta-stabilized plutonium-metal turnings have been cast into ingots. The quality of the ingots is comparable to that of production castings. Yields for a single casting varied from 65.19 to 84.46 percent recovery, depending primarily upon the length of time the metal was maintained in a molten state. By a process of successive castings in which the pouring skull from the preceding casting is added to a charge of turnings, the over-all recovery yield increases with each casting. After the initial casting, 93 to 95 percent of the plutonium turnings added are converted into an ingot. The conversion of plutonium-metal turnings to solid metal by a process of successive castings or some modification thereof is shown to be both possible and practical.

TABLE 2

CHEMICAL ANALYSES

Element	Run	Run 10	Run 11
Li[a]	<0.6	<0.6	<0.6
Be	<0.2	<0.2	<0.2
Na	<10	<10	<10
Mg	25	15	10
Ca	<5	<5	<5
Al	50	80	40
La	<10	<10	<10
Si[b]	70	90	90
Pb	1	1	1
Cu	100	50	20
Cr[b]	25	25	15
C	145	125	85
O_2	<500	<500	<500
B	<0.5	<0.5	<0.5
Fe	85	180	100
F	<1	<1	<1
Ni[b]	50	<20	170
Ga (%)	1.05	1.05	1.05
Pu (%)	98.9	99.0	98.9

[a] Figures are expressed in ppm except as noted.

[b] Photometric analysis.

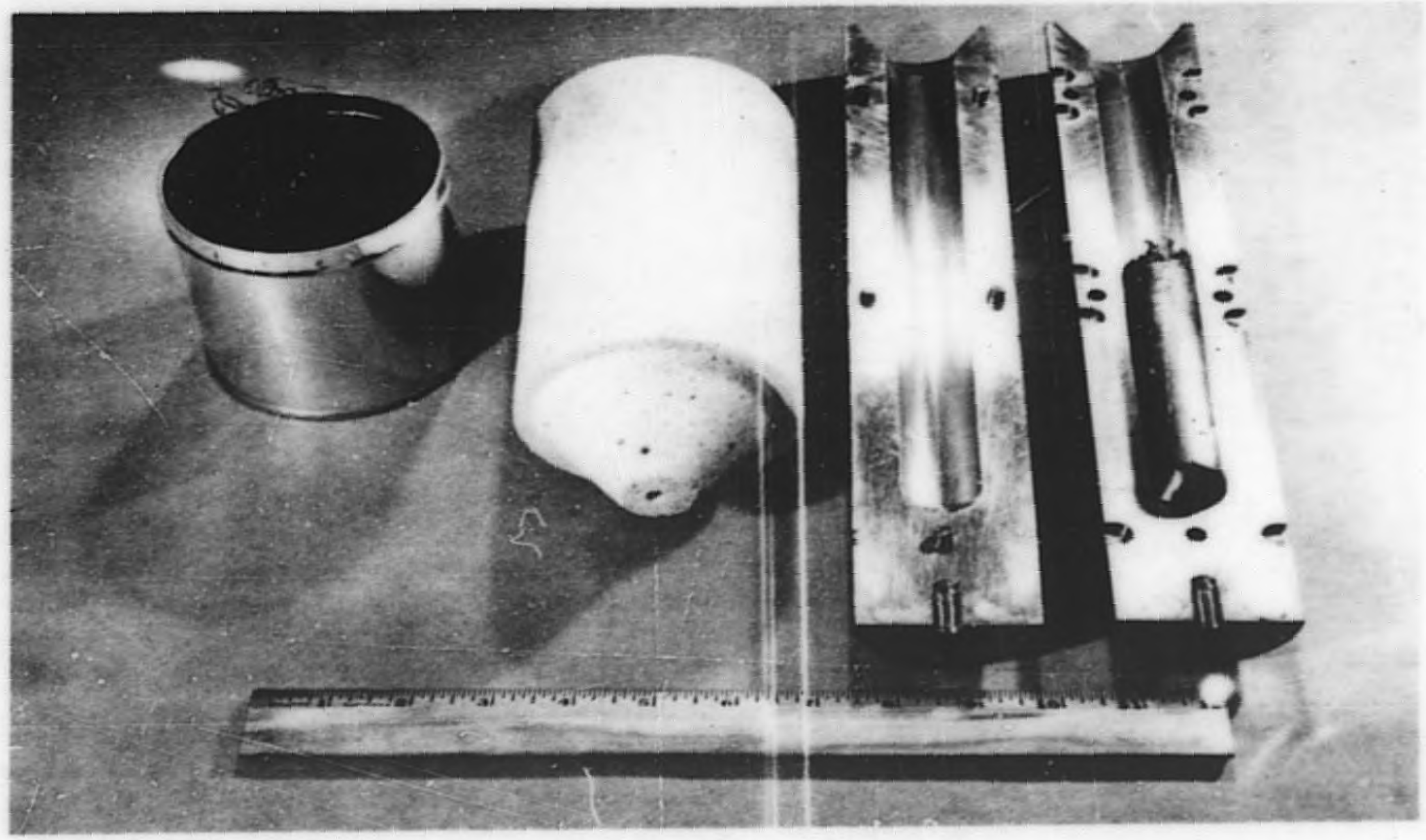

Fig. 6. Turnings, Mold and Product.

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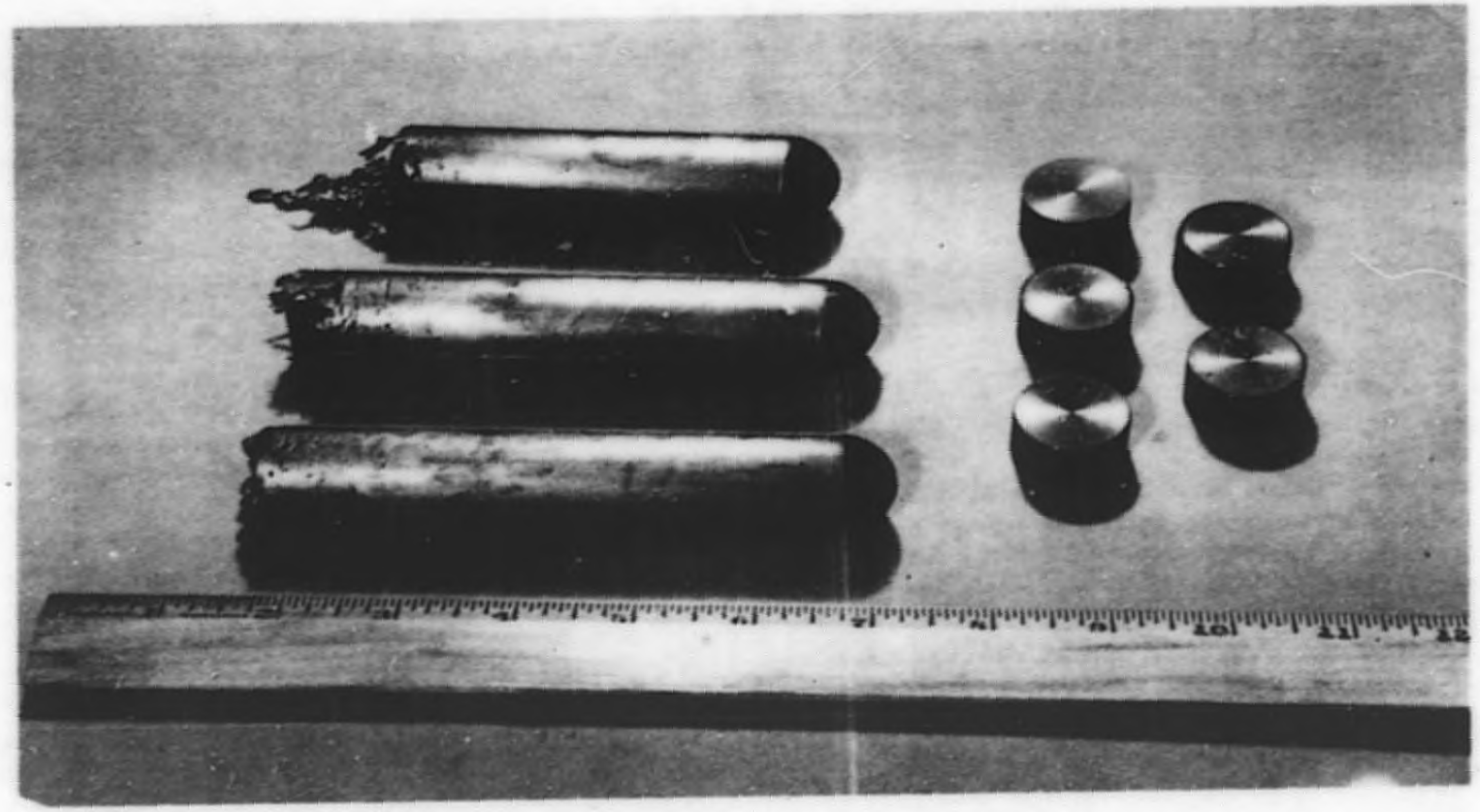

Fig. 7. Castings and Machined Parts.

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