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THE DOW CHEMICAL COMPANY
ROCKY FLATS DIVISION
GOLDEN, COLORADO

MASTER

REDUCTION OF PLUTONIUM COMPOUNDS
TO PLUTONIUM METAL

by

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ABSTRACT

A survey was made of the literature which was published on the reduction of plutonium compounds to plutonium metal between 1950 and April, 1963. This report is a summary of the information which was gathered during the survey.

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1. Summary

A literature search was conducted to determine what work had been done on the reduction of plutonium compounds to plutonium metal. It was discovered that all of the production scale reduction facilities in the United States use the bomb reduction method described by R. D. Baker³ of the Los Alamos Scientific Laboratory. This method, as well as other methods which were discussed in the literature, is presented.

2. Introduction

The necessary end result of any reduction is a well formed regulus, or button, which is easily separated from the slag. It is also necessary that the reduction yield and the metal purity be as high as possible.

Most of the equipment and procedure used in the United States for reducing plutonium compounds to plutonium metal is very similar in principle to that which is used at the Rocky Flats Plant. The procedure and equipment which are used at Rocky Flats and at other plants is described.

3. Discussion

3.1. The Reduction Reaction

When a plutonium compound, usually a halide, is reacted with an alkaline earth metal, usually calcium, a thermochemical reaction takes place which produces plutonium metal. A necessary property of the reaction of a plutonium compound with a reductant is that the heat of reaction be high enough to keep the slag molten long enough to allow the metal to separate from the slag and coalesce into a well shaped button. Also, the plutonium compound and the reductant should be stable with respect to humid air for ease of handling. Some of the reactions which have been investigated for the production of plutonium metal are listed in Table I.

TABLE I 4, 5

Reactions Investigated for the
Production of Plutonium Metal

		H at 291° K K Cal/Mol Pu	Melting Point of Slag, °C
$\text{PuO}_2 + 2 \text{ Ca} \longrightarrow$	$\text{Pu} + 2 \text{ CaO}_2$	- 52.4	2572
$\text{PuO}_2 + 2 \text{ Mg} \longrightarrow$	$\text{Pu} + 2 \text{ MgO}$	- 40.5	2500-2800
$\text{PuF}_4 + 4 \text{ Li} \longrightarrow$	$\text{Pu} + 4 \text{ LiF}$	-159.2	870
$\text{PuF}_4 + 2 \text{ Ca} \longrightarrow$	$\text{Pu} + 2 \text{ CaF}_2$	-149.5	1418
$\text{PuF}_3 + 3/2 \text{ Ca} \longrightarrow$	$\text{Pu} + 3/2 \text{ CaF}_2$	- 54.8	1418
$\text{PuCl}_3 + 3/2 \text{ Ca} \longrightarrow$	$\text{Pu} + 3/2 \text{ CaCl}_2$	- 56.0	772
$\text{PuBr}_3 + 3/2 \text{ Ca} \longrightarrow$	$\text{Pu} + 3/2 \text{ CaBr}_2$	- 56.9	765
$\text{CaPuF}_6 + 2 \text{ Ca} \longrightarrow$	$\text{Pu} + 3 \text{ CaF}_2$	-----	1418
$\text{PuI}_3 + 3/2 \text{ Ca} \longrightarrow$	$\text{Pu} + 3/2 \text{ CaI}_2$	- 59.9	575

Of the reactions listed in Table I, three have proven satisfactory for producing plutonium metal; the others have not. The reactions which have been found unsatisfactory will be considered first.

3.1.1 Unsatisfactory Reactions

The first two reactions in Table I are unsatisfactory because the high melting point of the slag, coupled with the fairly low heat of reaction, does not give the metal time to separate from the slag before the slag freezes. Attempts have been made to reduce plutonium dioxide with calcium⁵, using a calcium chloride flux; and lithium, using a lithium chloride flux. In both cases the reduction resulted in finely dispersed metal.

3.1.1 Unsatisfactory Reactions (Contd.)

The reduction of calcium plutonium hexafluoride with calcium⁵ gave very poor metal-slag separation because the globules which were formed did not agglomerate. This reaction was also tried using lithium, with the same result.

It is possible to reduce plutonium tetrafluoride with lithium, but the reaction is difficult to handle safely.

Plutonium tribromide and plutonium triiodide can be reduced with calcium, but they are very hygroscopic and care must be taken to insure that they are kept free from moisture.

3.1.2 Satisfactory Reactions

The remaining three reactions have all been shown to produce metal which separates easily from the slag and is produced with good yield and high purity.

3.1.2.1 Plutonium Trichloride

Plutonium trichloride was one of the plutonium compounds which was successfully reduced in the early work³ (1943-1946) at Los Alamos. Reductions of 1, 10, and 25 grams were made using 25% excess calcium with an iodine-calcium booster. Yields of 98% were reported.

In a later study¹⁸, 20-gram scale reductions were made with good results. Using 33% excess calcium and an iodine-calcium booster, yields of 97% were obtained. The reduction of plutonium trichloride without the use of a booster was also tried in this study, with fair results.

The reduction of plutonium trichloride has also been done on the 1000-gram scale¹⁵. In this study the reductions were made using an iodine-calcium booster and yields of 97-99% were reported, with total impurities in the metal of less than 0.2%.

3.1.2.1 Plutonium Trichloride (Contd.)

This report also stated that good mixing of the reduction charge was not always necessary. In fact, the report stated that the charge could be unmixed if the plutonium trichloride was on top.

One of the problems associated with the use of plutonium trichloride is its hygroscopicity. For this reason the plutonium trichloride must be stored in sealed containers. If moisture is present the plutonium trichloride will turn to the hexahydrate and, after a few days, to the oxide. The presence of moisture in the reduction could lead to hazardous bomb pressures.

3.1.2.2 Plutonium Trifluoride

Plutonium trifluoride was reduced on the 20-gram scale by F. Anselin². He reported that although the heat yield in the reduction of plutonium trifluoride was less than the heat yield in the reduction of plutonium tetrafluoride by a factor of 2.5, this did not seem to affect the metal yield with the heating method used.* The yields reported were generally good with the exception of one or two which were quite low; but in the reductions using plutonium trifluoride the reactions were more violent than when plutonium tetrafluoride was used.

The use of plutonium trifluoride was discontinued due to trouble with partial pyrohydrolysis of the trifluoride by traces of moisture to plutonium dioxide, which was reported to cause extensive spattering during reduction.

In another study¹⁶ plutonium trifluoride was reduced on the 20-gram scale with no apparent problems. Metal yields of 95-97% were reported using an iodine-calcium booster and 30% excess calcium.

* See Page 12

3.1.2.2 Plutonium Trifluoride (Contd.)

Plutonium trifluoride was used for several years at the Savannah River Plant to produce plutonium metal on a production scale.¹³ The reductions were made on a 2-kilogram scale using an iodine-calcium booster, 0.6-0.8 moles of iodine per mole of plutonium. Metal yields of 99% were reported for this method under normal operating conditions.

The problems arising from this process were not caused by the plutonium trifluoride, but by the iodine. The iodine caused considerable corrosion in the process equipment and interfered with metal recovery from slags and crucibles.

Due to these corrosion problems, a process was developed to oxidize the plutonium trifluoride to a mixture of plutonium tetrafluoride and plutonium dioxide. This mixture can be reduced without the use of an iodine booster. However, the yield obtained when this mixture is reduced, 95-97%, is lower than that obtained when the trifluoride is reduced with an iodine booster.

3.1.2.3 Plutonium Tetrafluoride

All of the production scale reduction facilities in the United States, and as near as can be determined, England and France, produce plutonium metal by reducing plutonium tetrafluoride. Therefore, the rest of this report will deal almost entirely with the preparation of plutonium metal from plutonium tetrafluoride.*

The reaction of plutonium tetrafluoride with calcium to produce plutonium metal is driven to completion by the addition of excess calcium. The minimum amount of excess calcium required seems to be between 20 and 25%. Many runs have been reported using anywhere from 25-50% excess calcium, all with good results. Reductions which were made using less than 20% excess calcium gave very poor yields.

* It should be noted that, to a great extent, the same procedures and equipment could be used for plutonium trifluoride and *plutonium trichloride*

3.1.2.3 Plutonium Tetrafluoride (Contd.)

Even if the reaction goes to completion, good metal-slag separation is necessary if good yields are to be obtained. In order to achieve good separation of the metal from the slag, it is necessary that the reaction products be in the molten stage long enough and the viscosity of the slag be sufficiently low to allow the metal to collect into a well formed regulus. Depending upon the size of the charge and the configuration of the reduction equipment, the heat of reaction may not always be sufficiently high to accomplish this. Various methods may be used to overcome this difficulty; such as supplying additional heat by post heating the reduction vessel, adding to the reduction charge a material which lowers the melting point of the slag, or having a highly exothermic "booster" reaction take place within the reaction vessel.

3.1.2.3.1 The Booster Reaction

Taking the latter methods first, the reaction between calcium and iodine:

supplies large quantities of heat, it lowers the starting temperature of the reduction, and the CaI_2 lowers the melting point of the CaF_2 slag¹¹.

The amount of iodine needed for the booster reaction depends upon the size of the charge. With small charges the ratio of moles of iodine to moles of plutonium should be large and with large charges the ratio of iodine to plutonium should be small. As the size of the charge increases, the heat provided by the booster reaction becomes less significant. However, even on the 1 to 2 kilogram scale, the addition of small amounts of iodine to the charge increases the yield and improves metal-slag separation. The optimum ratios of iodine to plutonium as reported by investigators at Hanford are shown in Table II⁴.

3.1.2.3.1 The Booster Reaction (Contd.)TABLE II

Optimum Ratio of Iodine to Plutonium Tetrafluoride in Various Nominal Charge Sizes for Reduction by Calcium

<u>Scale</u> <u>(Grams of Pu)</u>	<u>Booster Ratio</u> <u>(Moles I₂/Mole PuF₄)</u>
5	0.50
50	0.35
500	0.30
1000	0.15
1500	0.10

One other booster reaction that has been tried⁹ is the reaction between sulfur and calcium. This reaction did not prove satisfactory for the following reasons:

1. Lower Yields - average yield of 96.8% with sulfur as compared with average yield of 97.5% using iodine in the same equipment.
2. The slag was wetted by the metal and up to 10 hours pickling in nitric acid solution was required to remove the slag from the metal.
3. Small amounts of metal were found in the slag and trapped between the slag and the crucible wall.

Even though the iodine-calcium booster reaction is used at other AEC installations, it is not, at present, used at Rocky Flats. At Rocky Flats additional heat is supplied by continuing to heat the reaction vessel for two minutes after the reduction has taken place. Although it has been shown that the use of the booster reaction is helpful, it is not felt that the difficulties encountered in handling iodine and calcium iodine containing slag are justified by the increase in yield obtainable with the booster reaction.

3.1.2.3.1 The Booster Reaction (Contd.)

The French² experiments on the reduction of plutonium tetrafluoride were also run without the use of a booster reaction. They heated the reaction products for from 1 to 1.5 minutes after reduction had occurred and obtained yields of 95-97% on the 20 gram scale and 98-98.5% on the 50 gram scale.

3.1.2.3.2 The Bomb Atmosphere

The use of an inert atmosphere, such as argon, in the reduction vessel is necessary to obtain good yields. Air or nitrogen within the vessel will result in much of the slag freezing on the walls and the button will be poorly formed and difficult to clean³. An argon atmosphere will also prevent the formation of calcium oxide and nitride.

3.2 Reduction Equipment

The reduction equipment which is used at Rocky Flats is very similar to the equipment used at Los Alamos^{1,2}, but it has been scaled up to increase production capacity. The reduction vessel consists of a magnesium oxide crucible which is placed within a stainless steel pressure vessel. The annulus between the crucible and pressure vessel is filled with magnesium oxide sand. A crucible lid is placed on the crucible and a copper gasket is placed in a groove around the top of the pressure vessel.

Another type of pressure vessel which has been used consists of a steel bomb with a flanged lid which contains a screw for introducing inert gas and which is bolted onto the bomb. This vessel also utilizes a crucible lid and a copper gasket between the bomb and bomb lid.

The reduction vessel described in the French report² is quite different from those used in the United States. Their reduction vessel consisted of a sintered calcium fluoride crucible which was placed within a guard crucible of calcium fluoride. They reported that the inner crucible was strongly attacked during

3.2 Reduction Equipment (Contd.)

reduction and that the guard crucible was sometimes cracked. For this reason they have gone to a little different vessel for their production scale equipment⁸. They now place the calcium fluoride crucible in a silica guard crucible and fill the space between the crucibles with fluoride powder.

3.2.1 Crucibles

The crucibles which are used in the reduction vessels can be of many shapes and sizes and they can be made from several different materials. The crucible size is dependent upon the size of button which is desired and the size of the button is limited by criticality considerations. Most of the buttons now being produced on a production scale are between 1 and 2 kilograms.

Although magnesium oxide crucibles are used at Rocky Flats, crucibles have been fabricated from calcium fluoride, calcium oxide, and a mixture of calcium oxide with from 2 to 10% by weight of calcium fluoride¹. Also, a process¹⁷ has been developed where a magnesia crucible is placed inside a pressure vessel and the space between the crucible and pressure vessel filled with magnesia sand. Calcium chloride, or some other suitable salt or mixture of salts, is then melted in the vessel and allowed to impregnate the crucible and sand, forming a unitary reaction vessel.

3.2.2 Induction Furnace

The reduction charge is usually brought up to reaction temperature by placing the reaction vessel in an induction coil and inductively heating the pressure vessel. The French², however, do not heat the pressure vessel, because they do not use one, but couple directly to the calcium in the reduction charge. This method heats the charge directly without first heating the calcium fluoride crucible. This results in a much shorter heating time before the charge fires.

3.2.2 Induction Furnace (Contd.)

The power is supplied to the induction coil by a high frequency converter, many different types of which are available.

3.3 Procedure

The procedure for reducing plutonium tetrafluoride to plutonium metal which is used at Rocky Flats is similar to the procedure which is used at the other plutonium producing facilities in the United States. All of the operations described below are carried out in glove boxes which are maintained at a pressure from 0.4 to 0.8 inches of water below the pressure in the room. These precautions are necessary because of the extreme toxicity of plutonium.

Plutonium tetrafluoride, which is produced by hydrofluorinating plutonium dioxide, is mixed with calcium and this mixture is placed in a prepared reaction vessel. A crucible lid is placed on the crucible, a copper gasket is placed in a groove on top of the pressure vessel, and the vessel is transferred to the induction furnace.

Of the several methods which have been tried for mixing and loading the charge, no one method seems to have any great advantage over the other methods. The mixing has been done in a "Y" blender and it has been done by hand. The charges have been prepared by loading with alternate layers of calcium and plutonium tetrafluoride or by mixing the calcium and plutonium tetrafluoride and placing a layer of calcium, or calcium and iodine if a booster is used, beneath and above the mixed charge. The method presently being used at Rocky Flats is to hand mix the plutonium tetrafluoride and calcium.

The French, however, use a different procedure³. They mix plutonium tetrafluoride with 50% excess calcium and compact this mixture into cylinders 30 mm in diameter and 22 mm high, under a pressure of 1 ton/cm². Each cylinder is equivalent to 15 grams of plutonium. The following advantages were listed for this method:

1. The volume of the charge is reduced by 50%.

3.3 Procedure (Contd.)

2. There is better contact between the plutonium tetrafluoride and calcium.
3. The amount of gas retained in the mix is minimized
4. The cohesion of the cylinder is good and only a few particles of crushed calcium or fluoride remain on the walls of the die or the face of the punches.

When the charged reaction vessel arrives at the reduction station at the Rocky Flats Plant, it is placed on a hydraulic ram which lifts it into the induction coil. The gasket, which is placed in the gasket groove around the top of the pressure vessel, mates with a groove in a block valve which is bolted to the top of the glove box. The force of the ram seals the reaction vessel against the block valve. The vessel is then evacuated three times and filled with argon to assure that all the air in the vessel is replaced with argon.

After the vessel has been purged, it is inductively heated until the reaction temperature is reached. The reaction itself proceeds in a matter of seconds, during which temperatures up to 1600°C may be reached. The power to the coil is left on for two minutes after the reaction starts.

It has been reported³ that too rapid a heating rate will cause the reaction to occur on the outer surfaces of the charge while the rest of the charge remains at too low a temperature. The heat of reaction from the outer surfaces is dissipated in heating the rest of the charge and the products will solidify too rapidly. It has also been reported that when a $\text{Ca} + \text{I}_2 \rightarrow \text{CaI}_2$ booster is used and the heating rate is too slow, the calcium is coated with a CaI_2 layer which retards the rate of the reaction. The heat of reaction is lost to the liner and vessel before the reaction is complete and poor metal collection results.

The actual heating time varies with the size of the charge and the plant at which the reductions are being made. At the Rocky Flats Plant, the charge is heated for 10 minutes at 10 kw and then the power is increased to 13 kw until the charge fires. It usually takes between 25 and 30 minutes for the charge to fire.

3.3 Procedure (Contd.)

After the reduction is completed, the vessel is allowed to cool. Then, the slag and button are removed from the crucible and the button is separated from the slag with a hammer. The button is then pickled in a nitric acid solution to remove the slag which has adhered to the button.

3.4 Reduction Yield

It is, of course, important to obtain as high a yield as possible when reducing plutonium compounds to plutonium metal. Some of the factors which affect the metal yield, such as, the amount of excess calcium, the heating rate, use of an inert atmosphere, and the booster reaction have already been discussed. There are other factors which affect reduction yields and they are discussed below.

In any production facility the quality of the product is bound to vary. This is true of the plutonium tetrafluoride which is used in normal production scale reductions at Rocky Flats. Studies^{7, 14} have been made which show that variations in the quality of the plutonium tetrafluoride have a definite effect on reduction yields. One of these studies¹⁴ showed that if the chemical composition of the plutonium tetrafluoride cake is held constant, the reduction yields obtainable with this plutonium tetrafluoride cake are constant.

Table III shows the data which were obtained in this study. The data were obtained by making several, usually three, reductions from each batch of plutonium tetrafluoride cake. The data in Table III also indicate that much better yields can be obtained if the amount of impurities, percent unaccounted, in the plutonium tetrafluoride cake is held as low as possible.

In another study at Rocky Flats the plutonium tetrafluoride cakes for 100 reductions were sampled and analyzed for the following physical and chemical properties:

1. Physical Properties

- a. Bulk Density
- b. Particle Size

TABLE III
Reproducibility of Reduction Yields

<u>Fluoride Batch No.</u>	<u>% Pu</u>	<u>% F</u>	<u>% H₂O</u>	<u>Unaccounted</u>	<u>Button No.</u>	<u>Yield</u>	<u>Average</u>	<u>Range</u>
1	76.46	16.08	0.01	7.45	1-1	84.00	84.00	0.96
					1-2	84.53		
					1-3	83.47		
2	75.25	18.50	0.01	6.24	2-1	87.50	87.9	1.20
					2-2	88.30		
3	77.74	18.25	0.01	4.00	3-1	98.70	98.7	0.60
					3-2	98.40		
					3-3	99.00		
4	75.52	18.00	<0.01	6.47	4-1	91.00	90.3	1.70
					4-2	89.33		
					4-3	90.67		
5	77.25	17.22	<0.01	5.52	5-1	95.26	95.5	0.60
					5-2	95.40		
					5-3	95.93		

3.4 Reduction Yield (Contd.)

2. Chemical Properties

- a. Percent plutonium
- b. Percent fluoride
- c. Percent sulfate

The object of the study was to determine the effect of changes in the physical and chemical properties of the plutonium tetrafluoride cake on reduction yield.

The data were sent to the Computations Laboratory⁷ for statistical analysis. They reported that no correlation could be found between reduction yield and the physical properties which were studied. There was, however, a correlation between reduction yield and chemical composition. The following regression equation was obtained from the data:

$$y = 259.73 - 2.058x_1 - 0.129 x_2 - 2.267x_3$$

y = the expected yield
x₁ = percent plutonium
x₂ = percent fluoride
x₃ = percent sulfate

In this equation the percent fluoride, x₂, would have to vary over seven (7) percent before the reduction yield would be affected in the units place. This would indicate that the percent fluoride in the plutonium tetrafluoride cake does not affect the reduction yield nearly as much as the percent plutonium and percent sulfate.

At this point it should be noted that the results of this study can be considered as valid only in the following range:

Percent plutonium between 75.5 and 77.5
Percent fluoride between 18 and 23
Percent sulfate between 0 and 4

It is possible that the results of this study are valid over a wider range, but no conclusions can be drawn from this study which pertain to data outside the above range.

3.4 Reduction Yield (Contd.)

When the first regression equation indicated that, within the range of this study, the percent fluoride in the plutonium tetrafluoride cake has very little effect on reduction yield, the following regression equation was obtained using only the sulfate and plutonium data:

$$y = 245.17 - 1.906x_1 - 2.164x_2$$

y = the expected yield

x₁ = percent plutonium

x₂ = percent sulfate

This equation shows that, within the range of this study, the reduction yield increases as the plutonium and sulfate content of the plutonium tetrafluoride cake decrease.

The results of this study are summarized in Figures I, II, and III. The data shown in Figures I, II, and III were calculated using the second regression equation. Figure I shows the probability of obtaining specified yields as the plutonium content of the plutonium tetrafluoride cake varies. Figure II shows the probability of obtaining specified yields as the sulfate content of the plutonium tetrafluoride cake varies. Figure III shows the yields predicted by the second regression equation for various plutonium tetrafluoride cake compositions.

3.5 Metal Purity

The purity of the plutonium metal obtained from the bomb type reductions described in this report seems to depend almost entirely on the purity of the reactants which are used in the reduction. It has been reported that there is very little, if any, purification in the reduction step. One report¹⁹ states that no purification occurs during the reduction step, while another report² states that the only purification is in driving off the very volatile metal impurities. However, other reports^{6, 15} included data which indicated that some purification does occur during the reduction step. Whether there is actually any purification in the reduction step or not, it seems very clear, from the reports which have been studied, that if the plutonium tetrafluoride, calcium, and crucible used in a reduction are free from impurities, then the resulting metal will be free from impurities.

PROBABILITY OF OBTAINING A SPECIFIED YIELD

1.00
0.90
0.80
0.70
0.60
0.50
0.40
0.30
0.20

1/4

75

76

77

78

79

P₀

PROBABILITY OF OBTAINING A SPECIFIED YIELD
YIELD (LBS./AC.)

YIELD (LBS./AC.)

EXPECTED YIELD, PERCENT

100/10 = 10

3.5 Metal Purity

Plutonium metal with as little as 94 ppm^c of total impurities has been produced using the bomb reduction method. Other investigators have reported purities of 99.8% by weight^s (2000 ppm) and 99.96% by weight² (400 ppm).

3. Recommendations

- 3.1. Compaction of reduction charge - Some preliminary tests have been made at Rocky Flats on compacting the plutonium tetrafluoride and calcium reduction charge. It is recommended that these tests be continued and that several test reductions be made using compacted reduction charges.
- 3.2. Reduction equipment - When the new reduction equipment for 71 Building is designed, it is recommended that the type of pressure vessel which is now being used in 81 Building; that is, a pressure vessel with a lid which can be bolted onto the vessel; be considered for this new equipment.
- 3.3. Safety considerations - Because of the health hazards involved in handling large batches of plutonium tetrafluoride, it is recommended that serious consideration be given to the use of remotely operated equipment for producing large buttons.
- 3.4. Reduction process variables - It is recommended that a series of test reductions be made using carefully prepared plutonium tetrafluoride to study the following:
 - a. Determine the concentration of sulfate above which the reduction yield is affected
 - b. Determine the concentration of plutonium dioxide above which the reduction yield is affected
 - c. Determine the concentration of fluoride below which the reduction yield is affected
 - d. Study the effect of adding CaI_2 , or some other salt, to the reduction charge to determine if the improved slag properties would justify its use

Also, determine if the slag and crucible dissolution process used in 81 Building to remove the CaI_2 from the slag could be adapted for use in 71 Building.

3.4 (Contd.)

- e. Study the hydration rate of plutonium tetrafluoride in atmospheres of controlled humidity and determine the effect of water of hydration on reduction yield
- f. Determine if, with the present handling procedures at Rocky Flats, enough of the calcium in a reduction charge could be oxidized by room air to calcium oxide to affect the reduction yield.

5. Remarks

The literature search which this report covers was part of a study which is being made to determine the feasibility of producing buttons in the 3 to 4.5 kg range. Also, if it is determined that buttons this large can be produced, this study is to provide design criteria for reduction equipment which will be capable of producing these buttons.

5.1 Compaction of Reduction Charge

The main factor which limits the button size is criticality considerations. For this reason the Nuclear Safety Group was requested to determine the maximum size button which could be produced safely. Their recommendations are listed in Table IV.

TABLE IV

Criticality Recommendations for Large Buttons

<u>Mass, kg</u>	<u>Thickness, Inches</u>	<u>Diameter, Inches</u>
4.5	0.4	6.67
4	0.6	5.24
3	0.7	4.17

The button diameters listed in Table IV were obtained for a cylindrically shaped button of uniform thickness, using a value of 19 g/cm³ for the density of plutonium metal. If 3 to 4.5 kg buttons are to be produced with the diameters shown in Table IV, 16 to 18 inch high crucibles will have to be used to hold the plutonium tetrafluoride and calcium charge. This would mean that the pressure vessel would have to be about 20 inches high. Since it is

5.1 Compaction of Reduction Charge (Contd.)

desirable to keep the pressure vessel as small as possible, for ease of handling, tests will be made in compacting the charge. As was stated on Page 13 of this report, the French reported that, besides other advantages, the volume of the reduction charge could be reduced by 50% when the charge was compacted. If this can be done, then the pressure vessel for a 3 to 4.5 kg button would only have to be about 10 inches high. In fact, if the volume of the charge can be reduced by 50%, 2.5 to 3 kg buttons can be made in the present reduction equipment by using a flat bottom crucible.

5.2 Reduction Equipment

The reduction equipment which is now being used in 71 Building has several drawbacks. One of the main drawbacks is that the induction furnaces are being used as reduction furnaces, purging stations, and cooling stations. Purging and, after the charge has fired, partial cooling of the reaction vessel while it is in the induction furnace reduces the number of reductions which can be made in a furnace per day. Also, if the reaction is violent, molten calcium can escape from the reaction vessel and coat the block valve. When this happens, the furnace must be shut down until the block valve is cleaned and repaired. The type of pressure vessel which has been recommended for the new reduction equipment (Recommendation No. 2) would solve several of these problems. With this type of vessel, purging could be done at a separate purging station and, after reduction, the vessel could be cooled at a separate cooling station. This would decrease the time the furnace was tied up per reduction, and thus, increase the number of reductions which could be made per day.

5.3 Safety Considerations

One of the safety hazards encountered when working with plutonium tetrafluoride is the high neutron flux caused by the alpha-neutron reaction between the plutonium and fluoride. This high neutron flux must be taken into account when designing equipment which will handle plutonium tetrafluoride to make sure that the operators who will work with this equipment will be adequately protected.

The Health Physics Group was contacted concerning the amount of shielding which would be required when handling large batches of plutonium tetrafluoride. Some of their recommendations are listed in Table V.

5.3 Safety Considerations (Contd.)

TABLE V

Shielding Requirements for
Large Batches of Plutonium Tetrafluoride

<u>Button Mass (kg)</u>	<u>Exposure Time (hr/wk)</u>	<u>Operating Distance (feet)</u>	<u>Thickness of Lucite Shielding (inches)</u>
4.5	40	2	8
3.3	40	2	7.5
3.3	20	2	6
3.3	10	2	4.5
3.3	40	4	4.5
3.3	10	4	3

As shown in Table V, the three factors which affect shielding thickness are:

- Button size (which, of course, controls the amount of fluoride needed for a reduction),
- The length of time per week an operator is exposed to the radiation,
- The distance from the fluoride at which the operator works.

It was recommended that consideration be given to the use of remotely operated equipment for reducing large buttons (Recommendation No. 3) because of the excessive amount of shielding needed for manual operation (operating distance of two feet). Remote operation would increase the operating distance, and thus, decrease the shielding thickness. Also, remote operation would increase the safety of the operators.

5.4 Reduction Process Variables

It became apparent during the course of this literature search that more information is needed on the affect of certain process variables on reduction yield. For instance, the study at Rocky Flats, Page 15, indicated that the plutonium and sulfate content of the plutonium tetrafluoride cake have a great effect on the reduction yield, but the fluoride content does not. The present practice at Rocky Flats is to use the fluoride content of the plutonium tetrafluoride cake to determine if it is suitable for reduction or should be refluorinated.

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