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# TEMPERATURE MEASUREMENT AND CONTROL IN THE BOMB REDUCTION OF PLUTONIUM TETRAFLUORIDE


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## CONTENTS

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# TEMPERATURE MEASUREMENT AND CONTROL IN THE BOMB REDUCTION OF PLUTONIUM TETRAFLUORIDE

*Jack L. Long, James C. Brown, and Robert G. Auge*

**Abstract.** A technique was developed for estimating the minimum temperatures attained in small and large scale bomb reductions of plutonium tetrafluoride with calcium metal. The temperatures in both the small (50 gram) scale and large (1.8 kg) scale exceeded 1900°C. The addition of calcium fluoride slag from previous reductions lowered the temperature to less than 1536°C.

Slag addition to 2.5 kg reductions resulted in practically no splattering of the molten reaction products and in increased reduction yields.

## INTRODUCTION

The standard method for the production of plutonium metal at Rocky Flats is by the reduction of plutonium tetrafluoride with calcium metal. The reaction is carried out in a magnesium oxide crucible which is sealed inside a pressure vessel. The equipment is shown in Figure 1. The pressure vessel is evacuated and back-filled with argon. The vessel and contents are heated until the reduction reaction (equation 1) is initiated:

$$PuF_4 + 2Ca \rightarrow Pu^\circ + 2CaF_2 \qquad (1)$$

Figure 1. Pressure Vessel, Copper Gasket, and Crucible.

The reaction is exothermic, 149.5 kilocalories per mole of plutonium,[1] and the reaction products are rapidly heated to the molten state. Calcium fluoride melts at 1414°C,[1] and the temperature within the reaction mass was generally believed to be in the range of 1600°C to 1700°C. More precise knowledge of the maximum temperature attained during the reduction is necessary to have a better understanding and control of the process.

## EXPERIMENTAL

The technique used for temperature measurement was to suspend within each reduction vessel a maximum of three tantalum tubes of 0.125 in. i.d., 0.010-in.-wall thickness, and 6- to 9-in. long, containing pure metals of different melting points. The amount of pure metal in each tube was less than 1 gram. The tubes were positioned so that the bottom of the tube was always above the level of the plutonium metal product which collected in the bottom of the crucible after reduction. Figure 2 is a radiograph of a small scale crucible after reduction and shows typical tube positioning. The plutonium metal, slag layer, and tantalum tubes are discernible. Similar positioning was used for temperature measurements in large scale reductions.

Temperature control was accomplished by adding calcium fluoride slag from previous reductions to the reduction charges. The constituents of any given reduction charge were pre-mixed in a closed polyethylene container which was rotated and inverted many times.

## RESULTS

The crucible charges and results of five, small-scale reductions are presented in Table 1. The percent yield was calculated as the weight of the plutonium metal button divided by the weight of plutonium in the compound(s) multiplied by 100. Comparison of experiments 71-1 and 71-2 shows that the inclusion of 25 wt% slag reduced the temperature from higher than 1900°C to lower than 1536°C with no difference in the yield.

The results of experiments 71-3 and 71-4, where a mixture of plutonium tri- and tetrafluoride was reduced, are

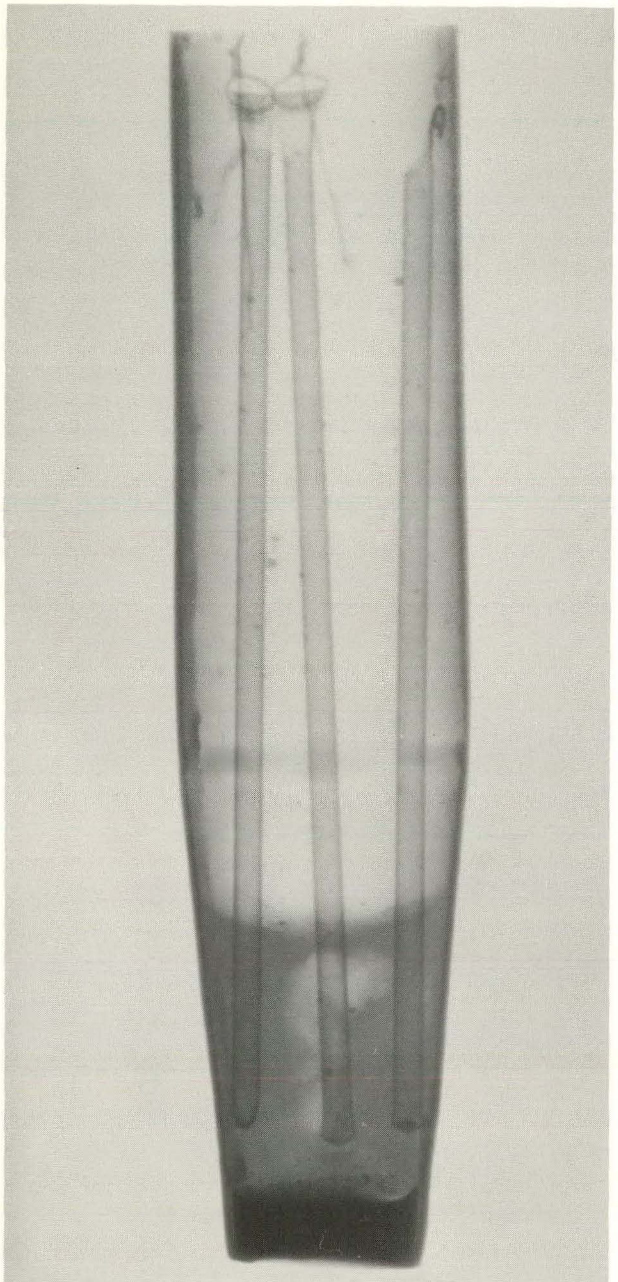

Figure 2. Radiograph of Tantalum Crucible Showing Suspended Tantalum Tubes.

essentially the same as for experiments 71-1 and 71-2. The slightly lower reduction yield from 71-4 is not considered to be significant. Experiment 71-5 illustrates the effect of substituting plutonium dioxide for plutonium tetrafluoride. A temperature reduction is evident but an extremely low reduction yield resulted.

The temperature profile of a 1.8-kg reduction charge, without slag added, was investigated by positioning three tantalum tubes containing pure metals across the diameter of a 4 ½ in.-diameter MgO crucible containing the charge. One tube, containing niobium, was located near the center of the crucible. A second tube, containing vanadium, was located approximately ¼ in. from the crucible wall. The third tube, also containing vanadium, was located approximately equidistant between the other two tubes. The vanadium in the tube adjacent to the crucible wall, and the niobium in the tube near the center of the crucible did not melt, but the vanadium in the third tube did melt.

A series of five 2.5-kg reductions was made with slag added. The amount added was in the same ratio as was used for the small scale work; that is, 25 percent by weight of the $PuF_4$ weight. For comparison purposes, these reductions were made concurrently with eighteen 2.0-kg routine-production runs without slag added. The calcium metal and plutonium tetrafluoride used were from production stock. The results of the two sets of runs are shown in Table 2. Note the increase in yield when slag was added. Less splattering was observed when slag was added, as evidenced by the lack of metal or solidified slag on the crucible cover. In contrast, solidified materials were frequently found on the crucible cover when no slag was added.

## DISCUSSION

The maximum temperature of 1900°C to 2415°C suggested by the metal-melting experiments, for both small and large scale reductions, could be low because of heat losses through the tantalum tubes. Similarly, the maximum temperature suggested for the small scale reductions, with slag additions, could be low. However, the addition of slag to the small scale reductions apparently lowered the temperature by at least 364°C (m.p. vanadium – m.p. of iron). The metal button yield was not affected by slag addition to the iodine-boosted, small scale reductions. Therefore, the decreased temperature was not detrimental.

The increased yields obtained in the large scale reductions, with slag added, may have been due, in part, to slower cooling and solidification of the slag, thereby allowing more time for the metal to coalesce. A factor, which was probably more important, was reducing the maximum temperature and causing a decrease in the reaction between the molten plutonium metal and the magnesium oxide crucible. One of the reaction products would be magnesium metal which has a high vapor pressure. As a result, a decrease in the rate of reaction would result in a decrease of the rate of magnesium metal vapor leaving the melt and, consequently, less splattering of the slag. The reduced splattering with slag added is indirect evidence of less reaction between the molten metal and the crucible material.

**Table 1. Crucible Charges and Results of Small Scale Reductions.**

| Experiment No | Charge Weight (g) PuF$_4$ | PuF$_3$ | I$_2$ | Ca | CaF$_2$ Slag | Theoretical Yield of Pu Metal (g)[a] | Weight of Pu Metal Button (g) | Percent Yield | Temperature of Reaction[b] (melting point in °C) 1563 Fe | 1769 Pt | 1900 V | 2415 Nb |
|---|---|---|---|---|---|---|---|---|---|---|---|---|
| 71-1 | 74.17 | | 26.8 | 29.9 | | 56.7 | 54.0 | 95.1 | M | M | M | NM |
| 71-2 | 74.17 | | 26.8 | 29.9 | 18.7[c] | 56.7 | 54.0 | 95.1 | NM | NM | NM | |
| 71-3 | 63.04 | 11.13 | 26.8 | 29.9 | | 56.7 | 54.0 | 95.1 | M | M | M | |
| 71-4 | 63.04 | 11.13 | 26.8 | 29.9 | 18.7 | 56.7 | 53.5 | 94.4 | NM | NM | NM | |
| 71-5 | 58.9 | PuO$_2$ 14.0 | 26.8 | 29.9 | | 56.7 | 43.0 | 75.6 | M | M | NM | |

[a]These weights are based on plutonium analyses of the plutonium compounds.
[b]The abbreviations used are: M for metals which melt, and NM for metals that do not melt.
[c]This is equivalent to 25 wt% of the PuF$_4$ weight.

**Table 2. Effect of Slag Addition on Metal Yield.**

2.5-kg scale reductions with slag added.

| Date | Theoretical Yield (g) | Plutonium Metal (g) | Percent Yield |
|---|---|---|---|
| 2-20-68 | 2500 | 2459 | 98.4 |
| 2-22-68 | 2500 | 2478 | 99.1 |
| 2-22-68 | 2500 | 2448 | 97.9 |
| 2-23-68 | 2500 | 2491 | 99.6 |
| 2-23-68 | 2500 | 2447 | 97.6 |
| | Theoretical grams 12500 | Total grams 12323 | Average 98.6 |

2.0-kg scale production runs without calcium fluoride slag added

| Date | Theoretical Yield (g) | Plutonium Metal (g) | Percent Yield |
|---|---|---|---|
| 2-20-68 | 2000 | 1880 | 94.0 |
| 2-20-68 | 2000 | 1978 | 98.9 |
| 2-20-68 | 2000 | 1978 | 98.9 |
| 2-20-68 | 2000 | 1732 | 86.6 |
| 2-20-68 | 2000 | 1956 | 97.8 |
| 2-22-68 | 2000 | 1944 | 97.2 |
| 2-22-68 | 2000 | 1746 | 87.3 |
| 2-22-68 | 2000 | 1706 | 85.3 |
| 2-22-68 | 2000 | 1838 | 91.9 |
| 2-22-68 | 2000 | 1956 | 97.8 |
| 2-22-68 | 3000 | 1866 | 93.3 |
| 2-23-68 | 2000 | 1714 | 85.7 |
| 2-23-68 | 2000 | 1968 | 98.4 |
| 2-23-68 | 2000 | 1822 | 91.1 |
| 2-23-68 | 2000 | 1724 | 86.2 |
| 2-23-68 | 2000 | 1950 | 97.5 |
| 2-23-68 | 2000 | 1890 | 94.5 |
| 2-23-68 | 2000 | 1898 | 94.9 |
| | Theoretical grams 36000 | Total grams 33546 | Average 93.3 |

This factor is discountable in the small scale reductions because tantalum vessels were used.

## CONCLUSIONS

A simple technique was developed to estimate the temperature in bomb reductions of plutonium tetrafluoride. The maximum temperatures in both small and large scale reductions were found to exceed 1900°C. The addition to small scale reductions, of calcium fluoride slag which was generated during previous reductions, did not affect the metal yield. The addition of slag to large scale reductions provided an average metal yield of 98.6% for the 2.5-kg scale as compared to an average metal yield of 93.3% for the 2.0-kg scale without slag added.

Splattering of molten reduction products was virtually eliminated by slag addition, probably by decreasing the rate of reaction between the magnesium oxide vessels and molten plutonium metal.

## REFERENCE

1. Plutonium Handbook, Volume 2, Chapter 15, page 564, by J. M. Cleveland (edited by O. J. Wick).