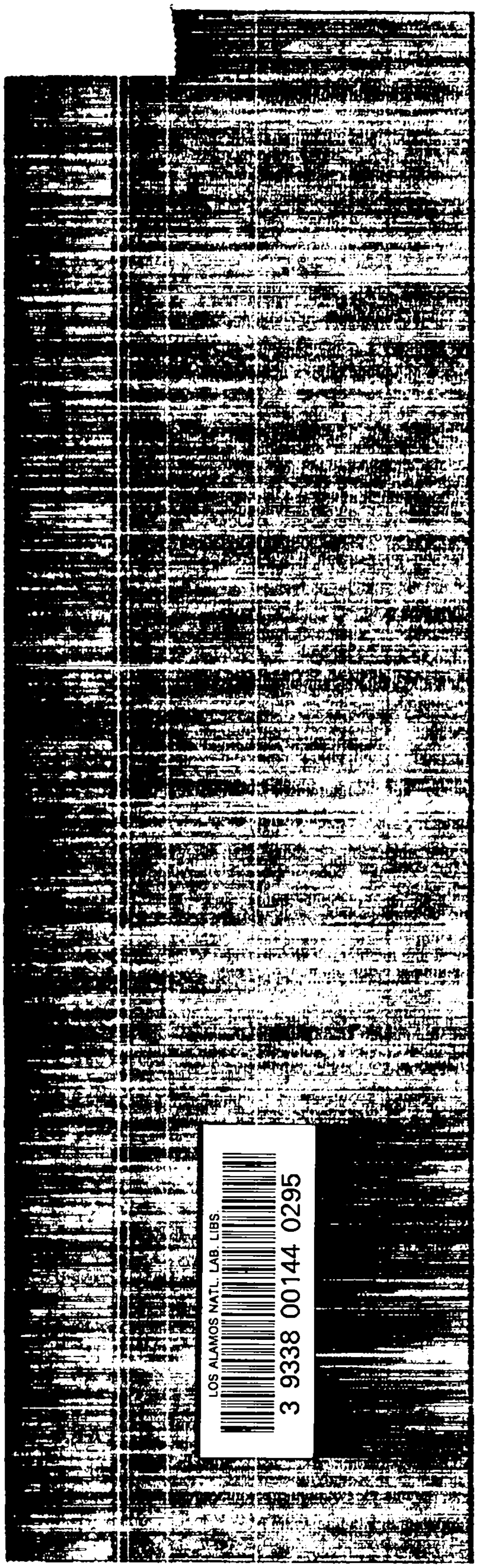
LA-11256



The Chlorination of Plutonium Dioxide

Los Alamos

Los Alamos National Laboratory is operated by the University of California for the United States Department of Energy under contract W-7405-ENG-36.

Edited by Connie Snyder
Photocomposition by Patrick Byrnes

LA-11256

UC-711
Issued: September 1988

The Chlorination of Plutonium Dioxide

Mike H. West
Michelle D. Ferran 0
Keith W. Fife 0

Los Alamos Los Alamos National Laboratory
Los Alamos,New Mexico 87545

THE CHLORINATION OF PLUTONIUM DIOXIDE

by

Mike H. West, Michelle D. Ferran, and Keith W. Fife

ABSTRACT

We investigated the conversion of PuO_2 to $PuCl_3$ with a number of chlorinating agents, sources of PuO_2, and reaction conditions, including temperature. We examined Cl_2, HCl, CCl_4, Cl_2-CCl_4, and HCl-CCl_4 as potential chlorinating agents. Our study showed that Cl_2-CCl_4 was the superior chlorinating agent. Using this agent, low-fired PuO_2—formed by calcining plutonium (III) oxalate at temperatures below 500°C—is more reactive toward chlorination than high-fired PuO_2, which is formed by calcining plutonium (III) oxalate at 850°C. Both the low- and high-fired PuO_2 feed materials are more reactive than when PuO_2 is produced by burning plutonium metal at 400–500°C (foundry oxide). The conversion efficiency of PuO_2 to $PuCl_3$ is relatively constant over a batch-size range of 10–300 g.

I. INTRODUCTION

In recent years, researchers have noted a resurgence of interest in synthesizing $PuCl_3$ for use in pyrochemical processes. Reasons for this renewed interest are broad, encompassing a range of processing activities. A primary driving force has been the desire to substitute $PuCl_3$ for PuF_4 in the traditional bomb-reduction (pressure vessel) process for the large-scale production of plutonium metal. For three decades, concerns have arisen over the $\{\alpha, n\}$ reaction from the PuF_4 molecule and the subsequent high neutron exposure rates for chemical operators.[1] In addition, the $CaCl_2$-CaI_2 slag—formed during the bomb-reduction process in which $PuCl_3$ is reduced by calcium metal in the presence of an iodine booster—has a lower melting point than the analogous slag (CaF_2-CaI_2), which is formed by the reduction of PuF_4. The $CaCl_2$-CaI_2 slag is also more readily dissolved in aqueous plutonium recovery schemes.

Other significant molten salt processes for purifying and producing plutonium metal that could incorporate $PuCl_3$ are the molten salt extraction (MSE) of americium from plutonium metal, electrorefining (ER) of plutonium metal, and the ambient pressure reduction (APR) of $PuCl_3$ to plutonium metal, with calcium metal or other reducing agents in a molten salt diluent matrix. The availability of $PuCl_3$ is also vital to fundamental electrochemical studies of trivalent plutonium in molten chloride salt systems.[2]

Los Alamos researchers believe it is worthwhile to replace $MgCl_2$ with $PuCl_3$ as an oxidant in the MSE and ER processes from the standpoint of eliminating magnesium as a system impurity. Magnesium metal volatilizes during foundry casting of plutonium metal, causing plutonium losses through splattering and damage to the casting equipment and the associated vacuum system.

Magnesium also participates in an $\{\alpha, n\}$ reaction with plutonium and americium, leading to a potentially serious neutron exposure problem. Another reason for using $PuCl_3$ in the MSE and ER processes is the large distribution coefficient for the partitioning of americium between the metal and salt phases in the presence of $PuCl_3$.[3]

The eutectic point for the $CaCl_2$-$PuCl_3$ phase diagram occurs at 40 mole % $PuCl_3$ and 610°C.[4] Compound formation is not observed for this system. Thus, it may be possible to solubilize 0.4 moles $PuCl_3$ (138 g) in 0.6 moles $CaCl_2$ (66.6 g) at temperatures above 610° C and reduce the $PuCl_3$ to plutonium metal with calcium metal reductant at temperatures much lower than those employed in the conventional direct oxide reduction (DOR) process (>800°C).[5] This procedure is the APR process. Calcium chloride will also be an effective solvent for CaO produced by the reduction of unconverted PuO_2 present in $PuCl_3$. In theory, the $CaCl_2$ salt phase is recycled to subsequent reductions as a result of the small CaO content and the increased purity resulting from the calcium reduction step. For plutonium metal, we are also considering performing the ER process in the vessel used for the APR process.

The development of synthetic routes for $PuCl_3$ production dates to the Manhattan Project efforts at Los Alamos and the Metallurgical Laboratory of the University of Chicago, which is now Argonne National Laboratory.[6,7] In the Soviet Union, Budayev and Volsky used CCl_4 to chlorinate PuO_2, which was produced by calcining plutonium oxalate.[8] Rasmussen and Hopkins[1] chlorinated low-fired PuO_2 with $COCl_2$ at temperatures up to 500°C. A screw calciner operated at 250–350°C produced the PuO_2 that subsequently flowed countercurrent to $COCl_2$ in a vibrating tube chlorinator. These workers also investigated HCl-H_2 and CO-Cl_2 as potential chlorinating agents but found them less reactive toward PuO_2 than $COCl_2$. Another potential source of $PuCl_3$ relies upon the substoichiometric oxidation of plutonium metal by $ZnCl_2$ in a KCl salt diluent. The oxidation process is a modification to the three-step pyroredox process for upgrading impure plutonium metal to a feed suitable for electrorefining.[9] It remains to be seen whether trace quantities of zinc metal are deleterious to the use of K_3PuCl_6 in other processes. Reavis developed a two-step synthesis for preparing $PuCl_3$ that involved forming $PuH_{2.7}$ at 25–250°C from the reaction between plutonium metal and H_2. This reaction was then followed by a subsequent reaction with HCl at 450°C for producing $PuCl_3$.[10] The reversible reaction between H_2 and plutonium metal is vital to forming a large surface area for the subsequent reaction with HCl.

Boreham et al. reported forming $PuCl_3$ from plutonium (III) and (IV) oxalates by the reaction with HCl or $COCl_2$.[11] Temperatures near 500°C were optimal for hydrochlorinating the plutonium (III) and (IV) oxalates. Nance at Los Alamos also successfully hydrochlorinated plutonium (III) oxalate.[12] Becker and Soine studied the influence of various molten salt systems on the dissolution of PuO_2 by Cl_2 or HCl gas sparge.[13] Maximum dissolution was observed for 0.3 LiCl-0.3 KCl-0.4 CsCl (mole fractions precede each salt component). The amount of PuO_2 dissolved and the concentration of tetravalent plutonium formed in the salt phase relative to the trivalent species depended directly upon the mole fraction of CsCl.

RESULTS

Recent Los Alamos research focused on evaluating prospective chlorinating agents for synthesizing $PuCl_3$ from PuO_2.

A. Small-Scale Chlorination Experiments

The first chlorinating agents investigated for PuO_2 were Cl_2 and HCl. Neither gas proved capable of chlorinating PuO_2 at temperatures up to 545°C (HCl) or 790°C (Cl_2). The thermodynamic data of Glassner suggest that chlorinating PuO_2 with HCl is not feasible.[14]

$$PuO_2 + 4HCl = PuCl_3 + 2H_2O + 1/2Cl_2, \tag{1}$$

where ΔG_{1000K} = 29 kcal/mole. Both low-fired and high-fired PuO_2 were studied in the case of Cl_2 and low-fired PuO_2 only with HCl. The inertness of PuO_2 toward HCl contrasts with AmO_2, which reacts with this reagent at 600°C to form $AmCl_3$.[15]

The first successful chlorinating agent for PuO_2 examined was CCl_4, which decomposes at elevated temperatures to a variety of compounds including C_6Cl_6, C_2Cl_4, C_2Cl_6, CO_2, CO, Cl_2, $COCl_2$, and C.[8] Argon was saturated with CCl_4 by bubbling the argon stream through CCl_4 contained in a gas-washing bottle (Fig. 1). The Ar-CCl_4 stream passed over PuO_2, which was contained in a quartz vessel heated by a Lindberg 55031 resistance furnace. The temperature of the reactor was monitored continuously using a type-K thermocouple protected by a quartz tube sealed at one end. Gases exiting the quartz reactor through a side arm were scrubbed by 5–6 M KOH.

The reaction proposed by Fullam and Soine[16] for the reaction of CCl_4 with PuO_2 is

$$PuO_2(s) + CCl_4(g) = PuCl_3(s) + CO_2(g) + 1/2Cl_2(g). \tag{2}$$

However, Fig. 2 shows that the $PuCl_3$ product formed by this reaction was a dark solid, suggesting carbon contamination from CCl_4 pyrolysis.

Therefore, the use of Cl_2 gas saturated with CCl_4 was explored as a means of synthesizing $PuCl_3$ unadulterated by carbon. The Cl_2 was saturated with CCl_4 by the technique described for argon. Figure 3 shows that the $PuCl_3$ resulting from chlorinating PuO_2 appeared relatively free of carbon contamination from CCl_4 pyrolysis.

During subsequent experiments with Cl_2-CCl_4, we investigated the influence of both chlorination temperatures and calcination temperatures for plutonium (III) oxalate on the extent of $PuCl_3$ formation from PuO_2 at the 10-g scale. Plutonium (III) oxalate is typically calcined at temperatures <500°C to produce what is commonly referred to as low-fired PuO_2. Figure 4 shows that the synthesis of $PuCl_3$ is more complete as the chlorination reaction temperature approaches 500°C. Through x-ray powder diffraction analyses we demonstrated that unreacted PuO_2 is present in $PuCl_3$. Low-fired PuO_2 is more reactive towards Cl_2-CCl_4 at all temperatures than high-fired PuO_2. High-fired PuO_2 is prepared by calcining the low-fired product at 800–900°C. This process prepares PuO_2 for use in the Los Alamos DOR operation during which plutonium metal is prepared by reducing

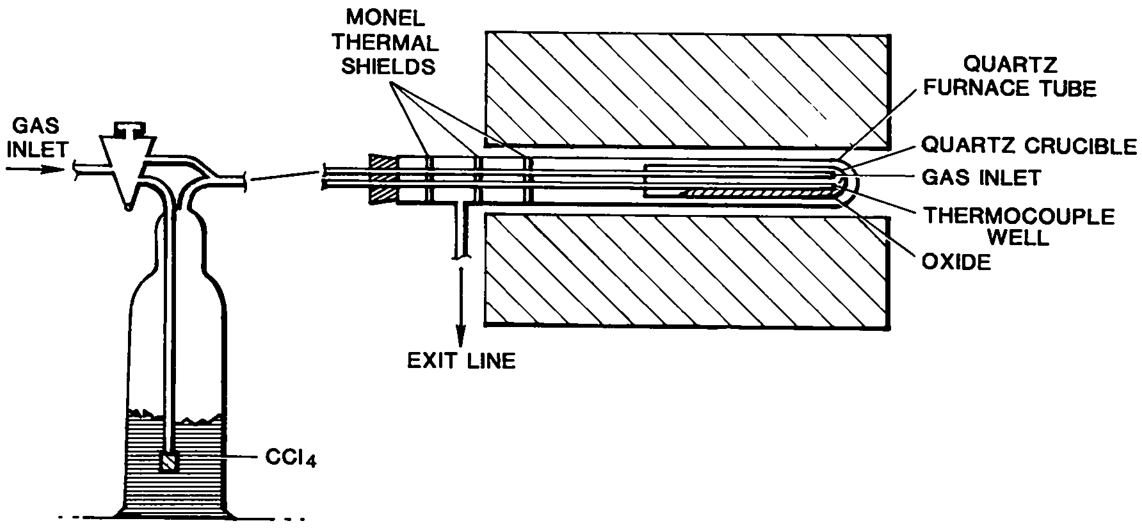


Fig. 1. Quartz reactor for exploratory chlorination experiments with PuO_2.

Fig. 2. Plutonium trichloride from a small-scale chlorination of PuO_2 with CCl_4. Carbon contamination of $PuCl_3$ results in a black coloration of the sample.

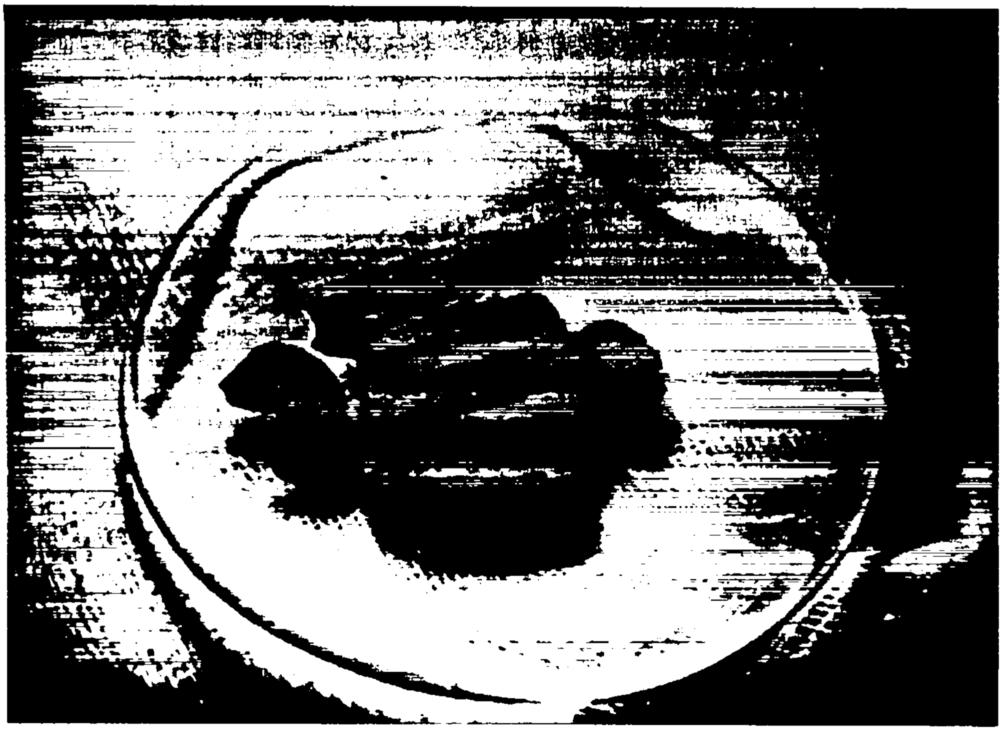


Fig. 3. Plutonium trichloride from a small-scale chlorination of PuO_2 with Cl_2-CCl_4. The absence of carbon during the process results in a lighter coloration of the sample, which contrasts Fig. 2.

the oxide with calcium metal in molten $CaCl_2$. By decreasing the chlorination reactivity of PuO_2 as it is calcined at increasingly higher temperatures (800–900°C), we substantiate the work of Bjorkland and Staritsky.[17] They found that as the calcination temperature increases, the reactivity of PuO_2 toward aqueous HCl-KI decreases. Correspondingly, the x-ray diffraction pattern of the oxide intensifies, indicating a more stable crystalline state.

Figure 5 shows that a similar series of experiments was performed using $HCl\text{-}CCl_4$ with nearly identical results. However, the conversion of low-fired PuO_2 to $PuCl_3$ is not as complete with $HCl\text{-}CCl_4$ as with $Cl_2\text{-}CCl_4$. In addition, carbonaceous deposits are present after chlorination with $HCl\text{-}CCl_4$.

Foundry oxide was also chlorinated with $Cl_2\text{-}CCl_4$ but not until the reactor temperature approached 700°C. Foundry oxide is unreactive toward concentrated HNO_3-dilute HF solutions. Bjorkland and Staritsky reported a 2.40 refractive index for oxide prepared from the metal at 170°C.[17] An identical value for the refractive index was obtained for oxide prepared from plutonium (IV) oxalate at 1000°C. The lack of reactivity is related to a stable PuO_2 crystal structure formed upon burning plutonium metal in air.

Plutonium trichloride, formed by chlorinating PuO_2, was transported from the main body of PuO_2 toward the monel heat reflectors shown in Fig. 1 and was deposited as a mass of green needles (Fig. 6). The chlorination rate appeared to depend on the transport of $PuCl_3$ from the reaction site. Plutonium trichloride has a negligible vapor pressure at 700°C (0.00029 mm),[18] but $PuCl_4$ has a substantial vapor pressure at the same temperature (3.7 mm).[19] Researchers believe the $PuCl_3$ is thus formed as in Eq. (2) and transported as in Eqs. (3) and (4).

$$PuCl_3(s) + 1/2Cl_2(g) = PuCl_4(g) \quad @ \quad 700°C \qquad (3)$$

$$PuCl_4(g) = PuCl_3(s) + 1/2\ Cl_2(g) \quad @ \quad < 700°C. \qquad (4)$$

The vapor-phase absorption spectrum of $PuCl_4$ at 925°C reported by Gruen and DeKock provides additional evidence for its existence in the vapor phase.[20]

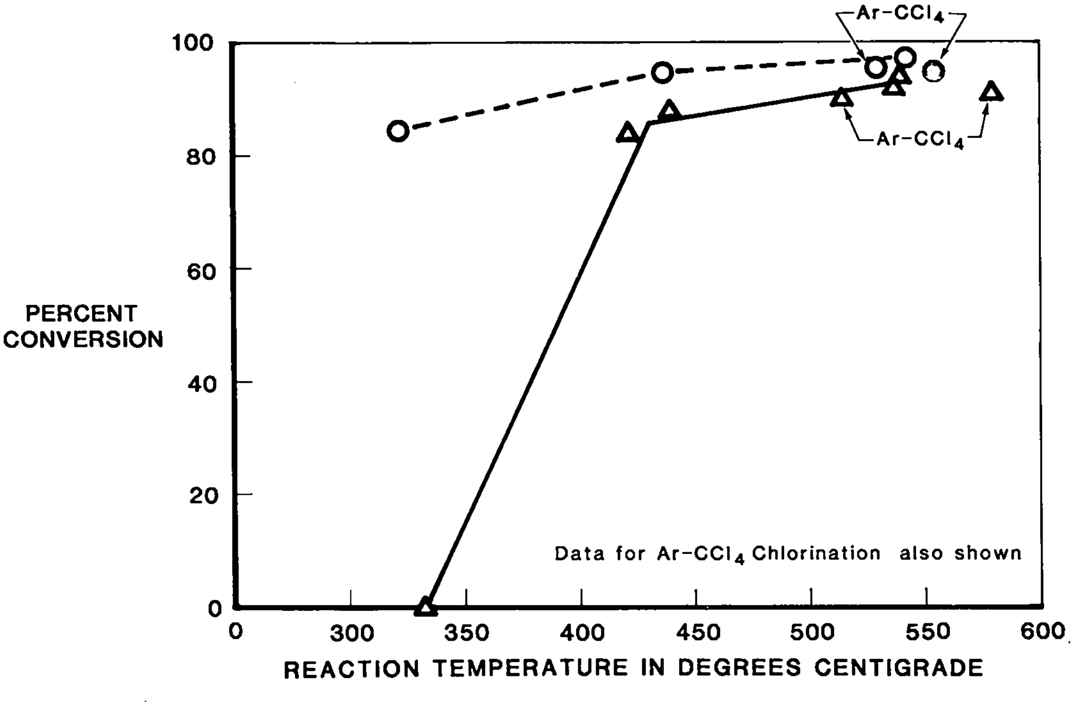


Fig. 4. Chlorination of low-fired (○) and high-fired (△) PuO_2 with $Cl_2\text{-}CCl_4$ as a function of temperature.

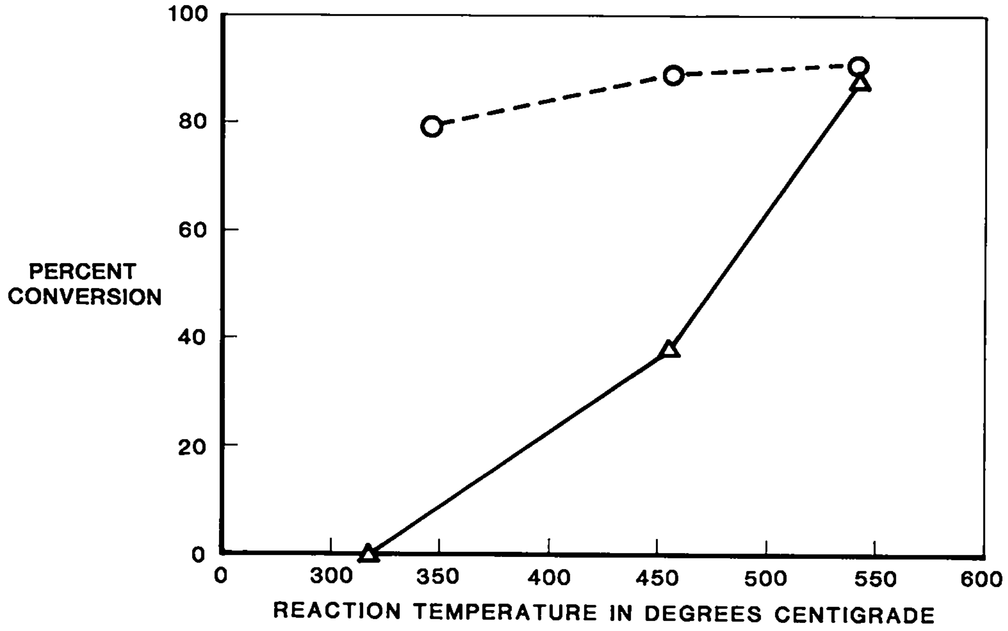


Fig. 5. Chlorination of low-fired (⊙) and high-fired (△) PuO_2 with HCl-CCl_4 as a function of temperature.

Fig. 6. Crystals of $PuCl_3$ from chlorination of foundry PuO_2 with Cl_2-CCl_4 at 700°C.

B. Large-Scale Chlorination Experiments with Cl_2-CCl_4

The initial work to scale up the batch chlorination experiments used a 3-in. (outer diameter) quartz vessel with a coarse porosity frit (Fig. 7), on which rest the PuO_2 feed; a 3-in. (inner diameter) Lindberg furnace (MK-3012) heated the reaction vessel. A gas inlet tube and thermocouple well, constructed from quartz, penetrated a silicone rubber stopper placed in a standard taper 24/40 ground joint at the vessel's top. Later, the rubber stopper was replaced with a Pyrex male ground joint with a thermocouple well and gas inlet tube, whereas a ball joint and elbow fitted with Tygon formed the exit line. Chlorine saturated with CCl_4 flowed into the top of the vessel through the inlet tube and exited through Tygon tubing at the vessel's bottom. Once the reaction neared completion, excess CCl_4 condensed in the off gas line. Fullam and Soine observed this phenomenon for the stirred-bed batch chlorinator.[16] Figure 8 shows that volatile chlorides, such as $FeCl_3$ and pyrolysis products of CCl_4, collect below the frit in the cool region of the furnace. The preliminary experiments used a 50-g batch size of PuO_2. The reactivity of the low-fired PuO_2 partially determined the time required for complete chlorination. We needed to double the Cl_2 flow rate through CCl_4 to achieve complete chlorination of 100-g batches of PuO_2 within a normal working day (one 8-hour shift). Figure 9 shows a 175-g batch size of $PuCl_3$ resting on the frit.

Table I illustrates the considerable variation in reactivity toward chlorination even among low-fired PuO_2 blends. The data in Table I also illustrate comparable reactivity for high-fired and some low-fired PuO_2 samples. Plutonium dioxide was originally derived by calcining plutonium (III) and (IV) oxalates.

Table I. Chlorination Reactivity of Some Selected Low-Fired and High-Fired PuO_2 Blends

Oxide Source	Pu (%)	Cl (%)	Reaction Time (h) at 500°C	Batch Size (g)
MSTPPB9C05 (a)	70.94	27.5	5 1/6	100.0
PUTHR2KFC5 (b)	70.71	27.1	6 2/3	50.0
MSTPPB334C (a)	70.17	28.3	13 5/6	200.0

(a) Low-fired PuO_2.
(b) High-fired PuO_2.

The MSTPPB PuO_2 blends, which are low-fired oxides, are normally suitable for fluorination to PuF_4 with HF for the synthesis of plutonium metal by the bomb-reduction process because fluorination also requires active or low-fired PuO_2.

Fifty-gram batches from blended, low-fired PuO_2 were chlorinated for periods between 1 and 6 hours at 500°C and 600°C. Chlorination appeared more rapid at the higher temperature, requiring approximately 3 hours, but work at 600°C requires using quartz, which is considerably more expensive, harder to fabricate, and more difficult to shape than Pyrex. For these reasons, chlorination at 500°C was chosen with approximately 4 hours required for a complete reaction (Fig. 10).

To achieve more rapid chlorination of PuO_2 and, concurrently, to increase the batch size, we constructed a Pyrex vessel from a 600-mL Büchner-type, fritted disc funnel. A standard taper 55/50 ground joint sealed the top of the reactor with an 18/7 ball joint and elbow forming an exit line. With this reactor vessel and a standard 6-in. (inner diameter) Lindberg furnace (MK-6015-S-V), up to 300-g batches of PuO_2 have been successfully chlorinated at 500°C in less than 7 hours. Table II summarizes the extent of chlorination for batch sizes ranging from 10 to 300 g.

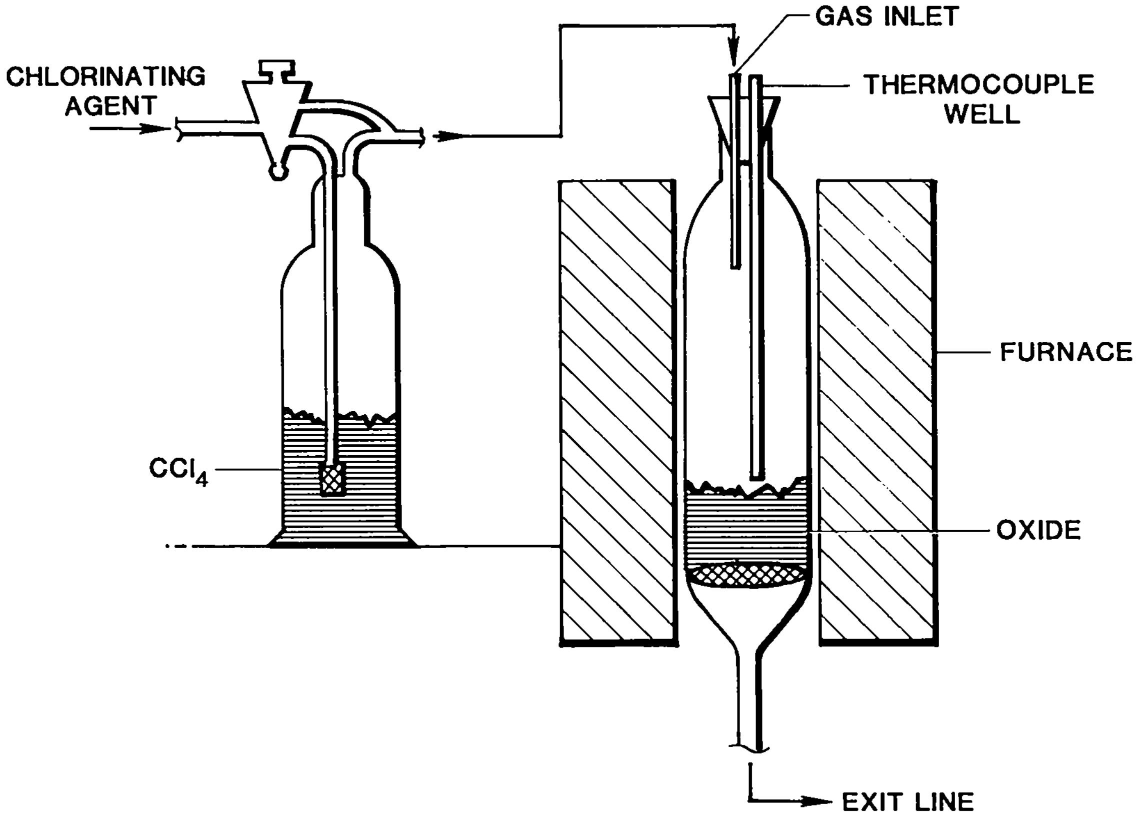


Fig. 7. Apparatus for chlorination experiments with PuO_2 (50–100 g).

Fig. 8. Condensation of volatile chlorides and CCl_4 pyrolysis products at the reactor exit. The reddish deposits represent the products of the $FeCl_3$ distilling process. The whitish deposits present in the condensate represent CCl_4 pyrolysis products.

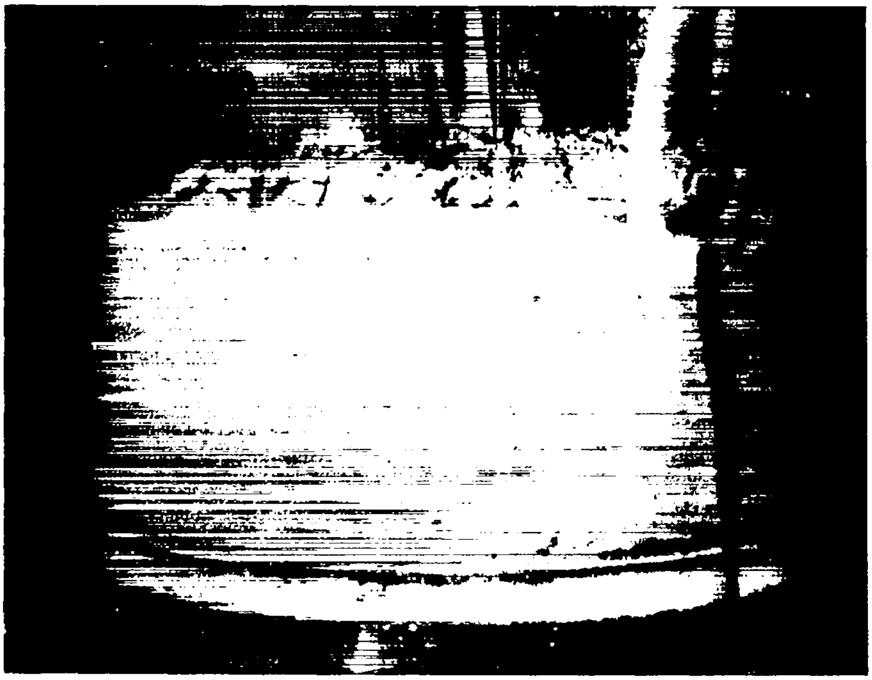


Fig. 9. Plutonium trichloride from a large-scale chlorination experiment (175 g PuO_2) using Cl_2-CCl_4.

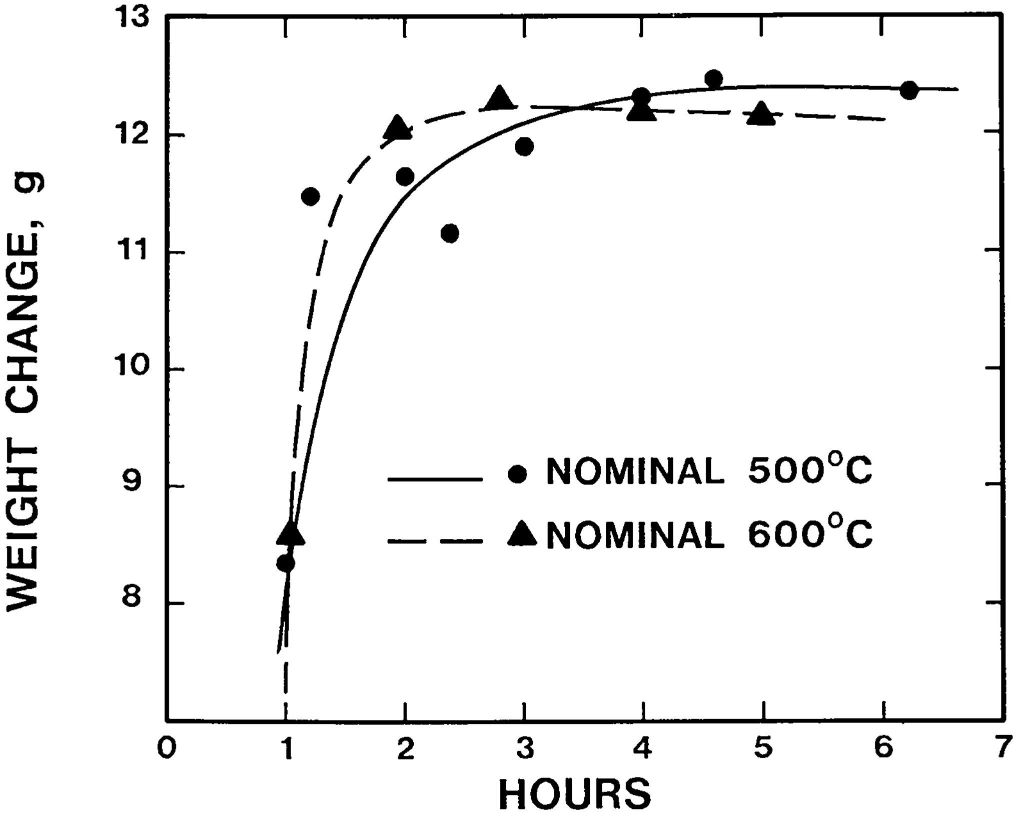


Fig. 10. Weight change (g) versus chlorination time at 500°C and 600°C.

Table II. The Effect of Batch Size on the Chlorination of PuO_2 with Cl_2-CCl_4

PuO_2 Batch Size (g)	$PuCl_3$ (%)	Number of Runs
10.0	95.4	2
50.0	94.5	2
75.0	94.2	1
100.0	94.8	14
125.0	94.6	3
150.0	95.1	1
200.0	95.2	8
300.0	94.6	6

The conversion efficiency is essentially constant at 95% as the batch size increases by an order of magnitude. The Appendix summarizes all chlorination experiments up to and including the 300-g batch size.

III. FUTURE WORK

We must continue our experimental work to evaluate other potential chlorinating agents including CO-Cl_2, photosensitized CO-Cl_2 mixtures, and $COCl_2$. (Phosgene was the optimum chlorinating agent for PuO_2 reported by the Hanford Atomic Products Operation.)[1]

Once researchers select an optimum reagent for production of $PuCl_3$, we must evaluate construction materials under reactor-like conditions to ensure they are corrosion-resistant.[21]. The most corrosion-resistant materials will be used for constructing large-scale chlorination vessels comparable to those used on the metal preparation line. Large-scale production of $PuCl_3$ will enable researchers to more rapidly evaluate potential uses of $PuCl_3$. We continue our efforts to incorporate and evaluate $PuCl_3$ in the processes of MSE, APR, ER, and bomb reduction.

ACKNOWLEDGMENTS

The authors wish to thank James G. Reavis and Robert L. Nance of Los Alamos National Laboratory and Fred D. Fisher of Westinghouse Hanford Company for encouragement and helpful advice during the early stages of this work. Don Temer, Tom Marshall, and Brad Roof of CLS-1 have been indispensable to this project, providing plutonium, chloride, and x-ray diffraction analyses for $PuCl_3$.

REFERENCES

1. M. J. Rasmussen and H. H. Hopkins, "Preparing Plutonium via the Chloride Process," *Industrial and Engineering Chemistry*, 53(6), 453 (1961).
2. L. E. McCurry, G. M. M. Moy, and D. F. Bowersox, "Electrochemistry of Plutonium in Molten Halides," Los Alamos National Laboratory document LA-UR-87-1385 (1987).
3. G. D. Bird and M. H. West, Los Alamos National Laboratory, unpublished data (1987).

4. J. A. Leary, "Temperature-Composition Diagrams of Pseudo-Binary Systems Containing Plutonium (III) Halides," Los Alamos Scientific Laboratory report LA-2661 (January 22, 1962).

5. L. J. Mullins and C. L. Foxx, "Direct Reduction of $^{238}PuO_2$ and $^{239}PuO_2$ to Metal," Los Alamos National Laboratory report LA-9073 (February 1982).

6. C. S. Garner, S. E. Bakes, I. B. Johns, G. B. Moulton, and B. Weinstock, "The Preparation of Plutonium Trichloride," Los Alamos Scientific Laboratory report LA-112 (July 24, 1944).

7. B. M. Abraham, B. B. Brody, N. R. Davidson, F. Hageman, I. Karle, J. J. Katz, and M. J. Wolf, "Preparation and Properties of Plutonium Chlorides and Oxychlorides," *The Transuranium Elements Research Papers*, Paper 6.7, G. T. Seaborg, J. J. Katz, and W. M. Manning, Eds. (McGraw-Hill, Inc., New York, 1949), pp. 740–758.

8. I. V. Budayev and A. N. Volsky, "The Chlorination of Uranium Dioxide and Plutonium Dioxide" in *Proceedings of the Second United Nations International Conference on the Peaceful Uses of Atomic Energy*, September 1–13, 1958, Geneva, Switzerland, Vol. 28, Paper 2195, 316 (1959).

9. J. A. McNeese, D. F. Bowersox, and D. C. Christensen, "Recovery of Plutonium by Pyroredox Processing," Los Alamos National Laboratory report LA-10457 (September 1985).

10. J. G. Reavis, K. W. R. Johnson, J. A. Leary, A. N. Morgan, A. E. Ogard, and K. A. Walsh, "The Preparation of Plutonium Halides for Fused Salt Studies," *Extractive and Physical Metallurgy of Plutonium and its Alloys*, W. D. Wilkinson, Ed. (Interscience Publishers, New York, 1959), p. 316.

11. D. Boreham, J. H. Freeman, E. W. Hooper, I. L. Jenkins, and J. L. Woodhead, "The Preparation of Plutonium Trichloride," *J. Inorg. Nucl. Chem.* **16**, 154 (1960).

12. R. L. Nance, Los Alamos National Laboratory, unpublished data (1960).

13. C. R. Becker and T. S. Soine, "Screening Tests for Próposed Molten Salt Electrolytes," General Electric Company, Hanford Atomic Products Operation report HW-77591 (May 10, 1963).

14. A. Glassner, "The Thermodynamic Properties of the Oxides, Fluorides, and Chlorides to 2500 K," Argonne National Laboratory report ANL-5750 (1957).

15. J. H. Burns and J. R. Peterson, "Refinement of the Crystal Structure of $AmCl_3$," *Acta Cryst.* **B26**, 1886 (1970).

16. H. J. Fullam and T. S. Soine, "Chlorination Reaction of Plutonium Oxide Prepared in a Screw Calciner," General Electric Company, Hanford Atomic Products Operation report RL-SEP-673 (December 1965).

17. C. W. Bjorkland and E. Staritzky, "Some Observations on the Reactivity of Plutonium Dioxide," Los Alamos Scientific Laboratory report LA-1869 (November 1, 1954).

18. T. E. Phipps, R. C. Seifert, G. W. Sears, and O. C. Simpson, "The Vapor Pressure of Plutonium Halides," *J. Chem. Phys.* **18**(3), 713 (1950).

19. R. Benz, "Thermodynamics of $PuCl_4$ from Transpiration Data," *J. Inorg. Nucl. Chem.* **24**, 119 (1962).

20. D. M. Gruen and C. W. DeKock, "Electronic Absorption Spectra of Gaseous $PuCl_3$, $PuBr_3$, and $PuCl_4$," *J. Inorg. Nucl. Chem.* **29**, 2569 (1967).

21. C. E. C. Rense, K. W. Fife, D. F. Bowersox, and M. D. Ferran, "Materials Compatibility During the Chlorination of Molten $CaCl_2$-CaO Salts," Los Alamos National Laboratory report LA-10700-MS (January 1987).

Appendix

Summary of Plutonium and Chloride Analyses for Chlorination Experiments with Cl_2-CCl_4

	initial weight (g)	final weight (g)	oxide batch	chlorinating agent	Pu, %	Cl, %	$PuCl_3$ (based on % Cl)	Cl/Pu
PUCL3-5P	9.4	11.6	MSTPPBMWC1	CCl_4	68.98	29.2	94.8	2.85
PUCL3-6P	10.1	12.7			69.37	29.4	95.5	2.86
PUCL3-7P	10.0	12.4	PUTHR2KFC5		70.39	28.1	91.2	2.69
PUCL3-8P	10.0	12.4			70.34	27.7	89.9	2.65
PUCL3-9P	9.9	12.3		CCl_4-Cl_2	69.06	28.9	93.8	2.82
PUCL3-17P	10.0	12.5			69.18	29.9	97.0	2.91
PUCL3-20P	10.0	12.3			69.87	28.3	91.8	2.73
PUCL3-23P	10.0	12.4	MSTPPBMWC1	HCl-CCl_4	67.77	28.1	91.2	2.79
PUCL3-24P	10.0	12.6	PUTHR2KFC5		69.69	27.0	87.7	2.61
PUCL3-31P	10.0	12.4	MSTPPBMWC1	CCl_4-Cl_2	69.53	28.0	90.9	2.71
PUCL3-32P	20.0	24.7			69.55	27.5	89.3	2.67
PUCL3-33P	30.0	37.0			70.57	27.9	90.6	2.66
PUCL3-39P	50.0	62.0			68.64	29.1	94.5	2.86
PUCL3-40P	50.0	62.3			68.95	29.1	94.5	2.85
PUCL3-45P	75.0	94.0			69.18	29.0	94.2	2.83
PUCL3-46P	100.0	124.6			67.26	29.3	95.1	2.94
PUCL3-47P	125.0	155.6			68.27	29.7	96.4	2.93
PUCL3-52P	50.0	60.4	PUTHR2KFC5		70.71	27.1	88.0	2.58
PUCL3-54P	100.0	123.2	MSTPPBCO5		70.94	27.5	89.3	2.61
PUCL3-55P	100.0	125.1	KHCB1C3		69.41	29.6	96.1	2.88
PUCL3-56P	125.0	156.3			69.76	28.8	93.5	2.78
PUCL3-57P	125.0	156.2			68.12	29.1	94.5	2.88
PUCL3-58P	150.0	187.3			69.20	29.3	95.1	2.85
PUCL3-59P	175.0	218.0			69.98	28.6	92.9	2.76
PUCL3-60P	100.0	124.5	KHCB2C5		69.81	29.9	97.1	2.89
PUCL3-68P	100.0	124.1			69.79	29.0	94.2	2.80
PUCL3-78P	100.0	124.6	KHCB1C3		69.76	29.5	95.8	2.85
PUCL3-79P	100.0	124.2	KHCB2C1		69.95	29.0	94.2	2.79
PUCL3-80P	100.0	123.6			69.76	29.2	94.8	2.82
PUCL3-81P	100.0	123.6			69.92	29.1	94.5	2.81
PUCL3-82P	100.0	124.1			69.96	29.1	94.5	2.80
PUCL3-83P	100.0	124.4			69.80	28.9	93.8	2.79
PUCL3-84P	100.0	124.5			69.85	29.2	94.8	2.82
PUCL3-85P	100.0	124.2			69.96	28.8	93.5	2.78
PUCL3-86P	100.0	124.2			69.47	29.1	94.5	2.82
PUCL3-87P	100.0	124.4			69.50	29.1	94.5	2.82

Appendix (cont.)

	initial weight (g)	final weight (g)	oxide batch	chlorinating agent	Pu, %	Cl, %	$PuCl_3$ (based on % Cl)	Cl/Pu
PUCL3-89P	200.0	248.6			69.67	29.4	95.5	2.84
PUCL3-90P	200.0	249.6			69.53	29.3	95.1	2.84
PUCL3-92P	250.0	311.2			69.60	29.4	95.5	2.85
PUCL3-93P	250.0	308.3			69.72	29.1	94.5	2.81
PUCL3-94P	300.0	373.7			69.78	29.4	95.5	2.84
PUCL3-95P	300.0	373.0			69.74	29.4	95.5	2.84
PUCL3-96P	201.9	251.8		KHCB2C4 + KHCB2C1	69.66	29.2	94.8	2.83
PUCL3-97P	200.0	249.6		KHCB2C4	69.47	29.5	95.8	2.86
PUCL3-98P	300.0	372.8			69.71	29.0	94.1	2.80
PUCL3-100P	300.0	372.7			69.87	28.9	93.8	2.79
PUCL3-104P	200.0	248.7		KHCB2C4	69.79	29.1	94.5	2.81
PUCL3-105P	200.0	248.5			69.50	29.3	95.1	2.84
PUCL3-106P	200.0	249.1			69.65	29.6	96.1	2.87
PUCL3-107P	200.0	249.0			69.72	29.3	95.1	2.83
PUCL3-108P	200.0	247.3		MSTPPB334C	70.17	28.3	91.9	2.72
PUCL3-109P	300.0	371.5		KHCB2C4	69.94	29.2	94.8	2.81
PUCL3-110P	300.0	374.4		MPB22ERC1	69.46	29.4	95.5	2.85

Printed in the United States of America
Available from
National Technical Information Service
US Department of Commerce
5285 Port Royal Road
Springfield, VA 22161

Microfiche (A01)

Page Range	NTIS Price Code	Page Range	NTIS Price Code	Page Range	NTIS Price Code	Page Range	NTIS Price Code
001-025	A02	151-175	A08	301-325	A14	451-475	A20
026-050	A03	176-200	A09	326-350	A15	476-500	A21
051-075	A04	201-225	A10	351-375	A16	501-525	A22
076-100	A05	226-250	A11	376-400	A17	526-550	A23
101-125	A06	251-275	A12	401-425	A18	551-575	A24
126-150	A07	276-300	A13	426-450	A19	576-600	A25
						601-up*	A99

*Contact NTIS for a price quote.

Los Alamos
Los Alamos National Laboratory
Los Alamos, New Mexico 87545