

*The Synthesis of Plutonium Trichloride
by Chlorination of Plutonium Dioxide
with Phosgene*

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*The Synthesis of Plutonium Trichloride
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THE SYNTHESIS OF PLUTONIUM TRICHLORIDE BY CHLORINATION OF PLUTONIUM DIOXIDE WITH PHOSGENE

by

Michelle D. Ferran and Michael H. West

ABSTRACT

Both phosgene (COCl_2) and chlorine-carbon tetrachloride ($\text{Cl}_2\text{-CCl}_4$) are effective reagents for the chlorination of low-fired plutonium dioxide (PuO_2), which results in the synthesis of plutonium trichloride (PuCl_3). Results are reported for 145 experiments including those where carbon monoxide-chlorine (CO-Cl_2) was investigated as a possible chlorinating agent for PuO_2 . Phosgene has proven to be a safe compound to adapt to a glove box environment and a simpler reagent to use than $\text{Cl}_2\text{-CCl}_4$. For 112 experiments where COCl_2 was the reagent for chlorination, the average plutonium content of PuCl_3 was 69.32 ± 0.38 wt % and the average chloride content was 29.9 ± 0.6 wt %. The corresponding theoretical values are 69.20 wt % plutonium and 30.8 wt % chloride. Using a Pyrex reactor vessel, PuCl_3 has been synthesized on the 500-g scale. Other supporting facilities, equipment, and instrumentation are described in the report.

Introduction

Synthesis of plutonium trichloride (PuCl_3) has proven important to support numerous pyrochemical operations at Los Alamos National Laboratory (LANL). The pyrochemical processes include the molten salt extraction of americium from plutonium metal [1], electrorefining of plutonium metal, ambient pressure reduction of PuCl_3 [2], and basic research into the molten salt chemistry of the actinides [3]. Currently, Lawrence Livermore National Laboratory is exploring the two-step synthesis of PuCl_3 from plutonium metal with plutonium hydride ($\text{PuH}_{2.7}$) as the intermediate compound [4]. Plutonium hydride is formed by reaction of hydrogen (H_2) with plutonium metal followed by conversion of the

hydride to PuCl_3 by use of 8 vol % hydrogen chloride (HCl) in argon. Rockwell International's Rocky Flats Plant produces cesium hexachloroplutonate, Cs_2PuCl_6 , by aqueous processing technology [5 and 6]. The use of chlorine gas (Cl_2) saturated with carbon tetrachloride (CCl_4) as a reagent for PuCl_3 production was previously reported by LANL [7].

The present report discusses the synthesis of PuCl_3 from low-fired plutonium dioxide (PuO_2) by reaction with phosgene (COCl_2) at 500°C . Work in this area had been previously described by Rasmussen and Hopkins [8], Fullam and Soine [9], and Soine [10]. Phosgene is a relatively simple reagent to use for the production of PuCl_3 and allows for a one-reagent, one-step synthesis. The risks of COCl_2 utilization, although real, are often exaggerated as this compound is routinely employed in universities [11–13] and industry (dyes, pharmaceuticals, herbicides, insecticides, synthetic foams, resins, polymers, and as a chlorinating agent) [14].

Experimental Procedure

Phosgene cylinders are stored in pairs outside the Plutonium Facility (PF-4 Building) in Room 116 of the PF-3 Building. Argon compressed gas cylinders are also stored in Room 116. Room 116 can be heated by steam heat. The COCl_2 cylinders (size 2P) are obtained from Matheson Gas Products in LaPorte, Texas. Phosgene is a liquid at 21°C with a vapor pressure of 10.7 psi at that temperature [14]. The manifold for handling COCl_2 in Room 116 is described in Los Alamos drawing number 26Y-200581, Fig. 1. The COCl_2 gas is transported from Room 116 through 1/4-in. Monel lines to GB (glove box) 439 in Room 429 of the PF-4 Building where chlorination of PuO_2 is performed routinely.

TLD-1 gas detectors for COCl_2 from MDA Scientific, Inc., are located above GB 439 in Room 429 and in Room 116, Fig. 2. These units have both audible and visual alarms which activate at 100 parts per billion COCl_2 , the threshold limit value-time weighted average (TLV-TWA) for this gas. The TLV-TWA is the weighted-average concentration of a compound to which an individual can be exposed for an 8-h workday without adverse effects. Two further audible alarms are located in Room 429 (one in the northwest corner and the other in the north center of the room) to provide additional warning to personnel to evacuate the room in the event of a COCl_2 release (However, none has ever occurred.). If the TLD-1 unit in Room 429 alarms, it automatically closes the electropneumatic solenoid controlling the COCl_2

flow from Room 116 to Room 429 (see Fig. 1). A digital display for the COCl_2 concentration in Room 116 is located outside the room so one does not have to enter the room to ascertain the concentration.

In GB 439 of Room 429, argon or COCl_2 flow to the Pyrex static bed reactor is controlled by mass flow controllers from Teledyne Hastings-Raydist, Fig. 3. The Model CST-M (Stock Number 54-147) is used for COCl_2 (flow range 0–500 standard cm^3/min) and the Model CST-(1K)M is used for argon (flow range 0–1.43 standard L/min). A four-channel power supply is also supplied by Teledyne Hastings-Raydist (Model CPR-4AJ and Stock Number 54-172), Fig. 4.

Tygon tubing (3/16-in. inner diameter by 1/16-in. wall) transports COCl_2 from Monel tubing inside GB 439 to the Pyrex reactor. A Pyrex 28/15 ball joint and socket ground joint with a 90° elbow form the reactor exit. Phosgene enters the reactor at the top and travels downward through the bed of $\text{PuO}_2\text{-PuCl}_3$ on the coarse-fritted disc. Gas exits the reactor through Tygon tubing (3/8-in. diameter by 1/8-in. wall), attached to the elbow, and passes to a multichambered scrubber filled with aqueous 5 M sodium hydroxide (NaOH) in an adjoining glove box, GB 440.

The Pyrex reactor is described in Los Alamos drawing number 26Y-200812 [10], Fig. 5. The Pyrex reactor is fabricated from a 2000-mL Buchner funnel with a coarse-fritted disc (VWR Scientific Number 30295-184). Despite the obvious fragility of glass, the reactor lasts for many temperature cycles.

Phosgene is introduced to the reactor once the internal temperature achieves about 200°C . The PuO_2 feed is under an argon atmosphere until this temperature is attained. The internal temperature is monitored with a Type K (Chromel-Alumel) thermocouple, and the temperature is output to a Honeywell Brown Electronik recorder [Model Number Y153x(67)-V12H-II-III-(101)]. A Lindberg furnace (MK-6015-SV) is controlled by a rheostat (Staco Energy Products Co., Dayton, Ohio, Type 2520CT, Input 240 V, 50/60 Hz, Output 0–280 V, 10 A, and 2.8 kVA). CER-WOOL (Premier Refractories and Chemicals, Inc., 1-in.-thick by 24-in.-wide by 300-in.-long HTZ8 blanket) is used to insulate the annulus between the Pyrex reactor and the furnace; the same type of insulation also was placed above the furnace. A rheostat setting of 65% is used initially to increase the temperature rapidly. The internal temperature is brought to 500°C and held there for approximately 8 h

(rheostat setting of 50%) during the chlorination. At the end of this time, the rheostat is adjusted to zero, and the reactor brought to ambient temperature under argon.

After chlorination is complete, the line is flushed with argon from Room 116 at 100 cm³/min overnight, removing traces of COCl₂, while the reactor cools. The argon also blankets the PuCl₃ product with an inert gas which prevents conversion back to an oxide, plutonium oxychloride (PuOCl), or adsorption of moisture while the product cools to room temperature. The COCl₂ cylinder in Room 116 is closed at the cylinder valve after completion of the daily run as an additional precaution.

If chlorination appears complete the next morning, based on the product being a blue-green color throughout, then the PuCl₃ is removed from the reactor, crushed with a Coors alumina mortar and pestle, and stored in a wide-mouth glass bottle with a phenolic cap containing a paper insert. A 2.2-g sample is typically sent for plutonium, chloride, and x-ray diffraction analyses by Chemical Laser Sciences Division (CLS-1). Once the product has been consumed in other pyrochemical operations, the bottles can be reused by simply wiping with cheesecloth.

If the PuCl₃ contains macroscopic crystals, which are forest green in color, this usually indicates incomplete chlorination. Axler [15] has shown the product containing macroscopic crystals to have larger amounts of unreacted PuO₂ relative to the remainder of the PuCl₃.

In the early stages of this work, a single-chambered scrubber was used for elimination of COCl₂ from the gas stream. For later stages, a multichambered scrubber, described in Los Alamos drawing number 26Y-200615 (not shown in this document), was used for COCl₂ removal. Gaseous effluents from the Pyrex reactor are injected into the bottom chamber and are forced to follow a transverse path through the upper chambers. The scrubber is held at a lesser pressure with respect to the glove box by using the wet vacuum system. The temperature of the aqueous caustic scrubber is monitored continuously and the solution is no longer used when the temperature attains approximately 70°C. An upper limit of 70°C was adopted to avoid overheating the scrubber, which is constructed from Plexiglas. An Omega Model 199-KC-X-X-DSS unit was used for a digital display of the scrubber temperature. A separate scrubber unit with fresh caustic then is used for removing further unreacted COCl₂. The spent caustic is filtered and analyzed for plutonium by an alpha-particle counting method. If the alpha-particle determination is less

than 5×10^9 counts/min/L, then the caustic is discarded through the caustic waste line to the Waste Treatment Facility at LANL TA-50.

Results and Discussion

The batch size typically varied between 200 and 500 g PuO₂ but eventually a batch size of 400 g was used for experiments beyond PUCL3-193 (Appendix). Results for experiments before PUCL3-111 are summarized in an earlier report [7]. The appendix contains data regarding the experiment number, identification of feed PuO₂ blend, feed batch size, quantity of product, chlorinating agent, chlorination temperature, chlorination time in hours, weight percent plutonium in the product, weight percent chloride in the product, chloride to plutonium mole ratio, and weight percent PuCl₃ in the product.

Through experiment PUCL3-138, a stream of Cl₂ saturated with CCl₄ was the chlorinating agent of choice. The apparatus for this reagent is described in an earlier report [7] and is somewhat awkward compared to a single gaseous reagent such as COCl₂. Therefore, efforts were transferred to the use of COCl₂ as a chlorinating agent in December 1987.

The use of CO-Cl₂ mixtures to chlorinate PuO₂ was reported by Rasmussen and Hopkins but no supporting data appeared in the paper other than the fact it was not as reactive as COCl₂ [8]. Chlorine-rich, CO-rich, and equal mixtures of these gases were tried at LANL in order to evaluate the efficiency of this mixture for chlorination of PuO₂.

For experiment PUCL3-115, the flow rates of Cl₂ and CO were nominally equal, and a weight increase of 33.7 g was found for 200 g PuO₂ feed. The succeeding chlorination experiment used a Cl₂ to CO flow ratio of 1.5, and the weight gain was 22.2 g for the same quantity of PuO₂ feed as in experiment PUCL3-115. The observed chloride content was 14.7 wt %. (The theoretical weight percent for chloride in PuCl₃ is 30.8.) The last work with CO-Cl₂ mixtures (PUCL3-117) used a flow ratio of 1/1.5, and the corresponding weight change was 40.2 g. The corresponding weight percent for chloride was 24.6.

Flow rates favoring CO over Cl₂ appear to improve the conversion of PuO₂ to PuCl₃, based on the increase in weight, but Cl₂-CCl₄ is still superior to the former as a chlorinating agent for a specified reaction time and temperature. For example, experiment PUCL3-107 [7] gave a weight change of 49.0 g for 200 g PuO₂ feed and chlorination with Cl₂-CCl₄ at 500°C.

The use of ultraviolet radiation from a mercury vapor lamp to photolytically dissociate molecular chlorine into chlorine radicals, which are likely stabilized by CO with formation of carbonyl chloride (COCl) radical, has been explored by Soleiman and Rao [16] for chlorination of alpha-alumina (Al₂O₃). They report results similar for chlorination with COCl₂ directly. Thus, the COCl radical is probably the species responsible for chlorination of oxide compounds by COCl₂. In addition, Ferran et al. [17] have observed COCl formation at the ionization filament in mass spectrometric studies of gaseous effluents from chlorination of PuO₂ by COCl₂.

The reaction of COCl₂ with PuO₂ has been shown to proceed according to Equation (1) [8,17]:



although the sum of Equations (2) and (3) would lead to the same resulting stoichiometry as Equation (1):



The presence of plutonium oxychloride (PuOCl) in the solid phase would need to be demonstrated to make the latter reaction sequence credible.

The average weight percent chloride in PuCl₃ product is shown in Table I for experiments PUCL3-118 through PUCL3-138, excluding PUCL3-122 and PUCL3-123, where Cl₂-CCl₄ or CCl₄ were the chlorinating agents. Similarly, the average weight percent chloride is also shown in Table I for experiments PUCL3-139 through PUCL3-255, excluding PUCL3-222, PUCL3-235, and PUCL3-249, where COCl₂ was

the reagent for chlorination. Use of COCl_2 leads to an improved conversion of PuO_2 to PuCl_3 . With COCl_2 as the reagent of choice, the weight percent plutonium in PuCl_3 is closer to the theoretical value of 69.2 wt % (see Table I).

Table I. Comparison between COCl_2 and $\text{Cl}_2\text{-CCl}_4$ as chlorinating agents for low-fired PuO_2 .

Chlorinating agents	Plutonium weight percent	Chloride weight percent	Number of runs
COCl_2	69.32 ± 0.38	29.9 ± 0.6	112
$\text{Cl}_2\text{-CCl}_4$	69.59 ± 0.27	29.7 ± 0.4	16

An uncertainty of one standard deviation is noted.

For five chlorination experiments, samples of PuCl_3 product were analyzed for impurities by direct current arc-atomic emission spectrometry using the carrier distillation technique. Unfortunately, with relative standard deviations of 50%, this analytical method is not particularly precise. A comparison of trace metal concentrations in PuCl_3 was made to the same trace impurity concentrations in the feed PuO_2 (see Table II). Aluminum was found as a contaminant in PuCl_3 on two occasions: plutonium trichloride was crushed with an Al_2O_3 mortar and pestle, and aluminum is also a constituent of Pyrex. The contamination of the product PuCl_3 was erratic, however. Use of a stainless steel vessel to crush the product is preferable but only in a dry air environment. Silicon is another impurity which was erratically accumulated in the product. For example, it was present at 320 ppm in PUCL3-215 and at 100 and 40 ppm in PUCL3-216 and PUCL3-217, respectively. The PuO_2 feed was identical for these three experiments and contained 70 ppm silicon. The source of silicon is likely the Pyrex reactor although the lack of a corresponding boron impurity in the product is puzzling because Pyrex is a borosilicate glass. Boron trichloride (BCl_3), which could be formed during chlorination of PuO_2 with COCl_2 , is very volatile and this might account for the lack of a boron impurity in PuCl_3 . Formation of aluminum trichloride (AlCl_3), silicon tetrachloride (SiCl_4), and BCl_3 from the respective oxides of aluminum, silicon, and boron is thermodynamically favorable at 527°C [18,19]; however, it is difficult to assess the kinetics.

Table II. Trace impurities in PuO₂ feed and PuCl₃ product.

	Al ppm	B ppm	Si ppm
MBP78ER*	10	<5	370
PUC13-205	10	<5	190
PUC13-216	590	<5	330
MSTPPB53*	8	<5	70
PUC13-215	700	<5	320
PUC13-216	7	<5	100
PUC13-217	<5	<5	40

*PuO₂ feeds to chlorination.

Future Work

Work by Ferran et al. [17] has shown that, for a 400-g feed batch of PuO₂, evolution of carbon dioxide (CO₂) slows considerably after about four hours of chlorination at 500°C. According to Equation (1), CO₂ is a product of the chlorination reaction. Because chlorination is not complete for this batch size until 8 h have elapsed, stirring the feed during part or all of the chlorination may enhance the chlorination rate. A reactor was designed and constructed with a stirrer but has not yet been tested (Los Alamos drawing number 26Y-199917, not shown in this document).

Currently, direct chlorination of molten plutonium metal also is being pursued as an alternate means of producing PuCl₃. Monitoring of the effluent gases for unreacted chlorine and a video camera for viewing of the high-temperature reactions are part of this research project [20].

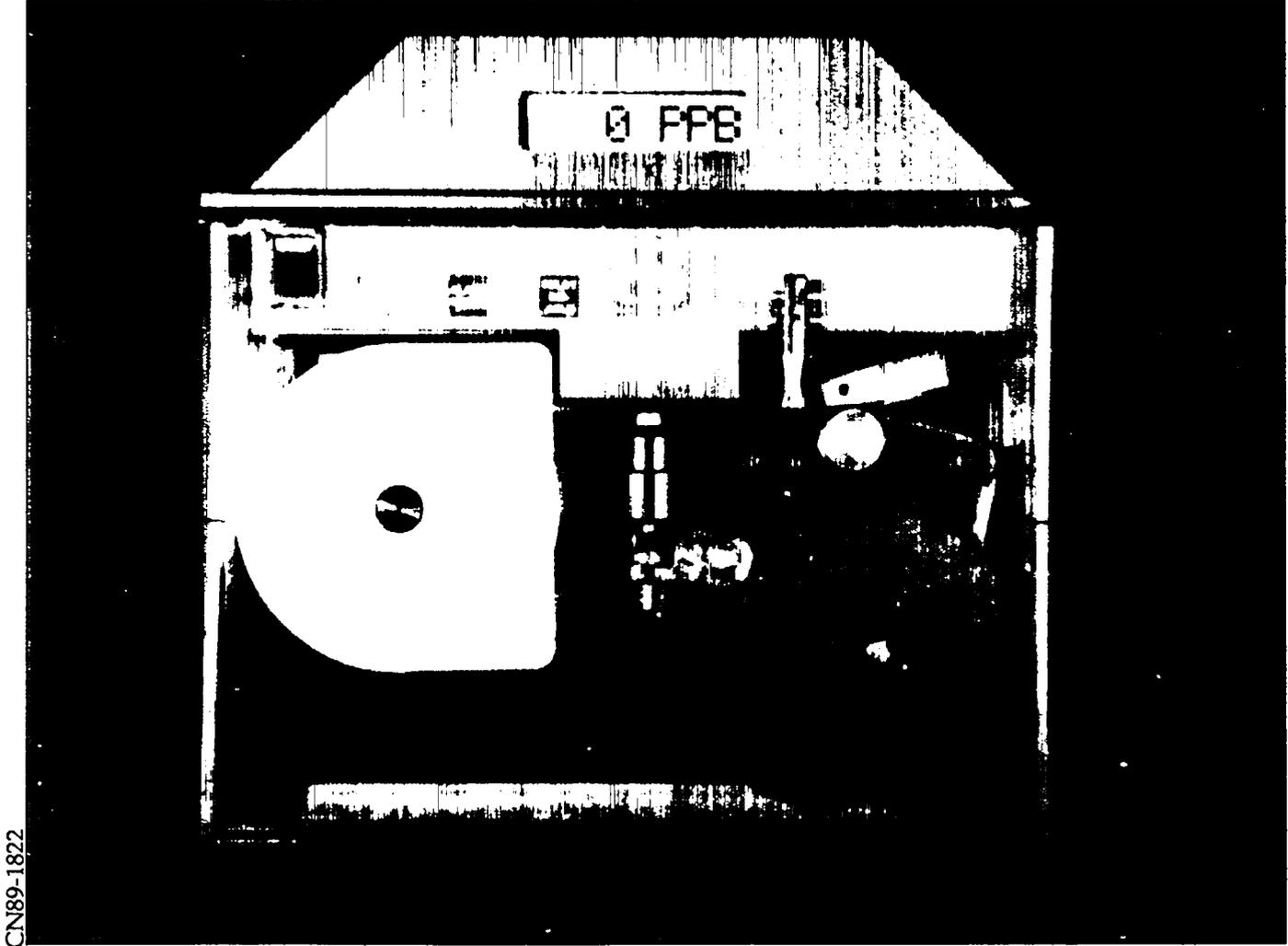
Acknowledgments

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Fig. 2. MDA's TLD-1 toxic gas detector for phosgene with Chemcassette.

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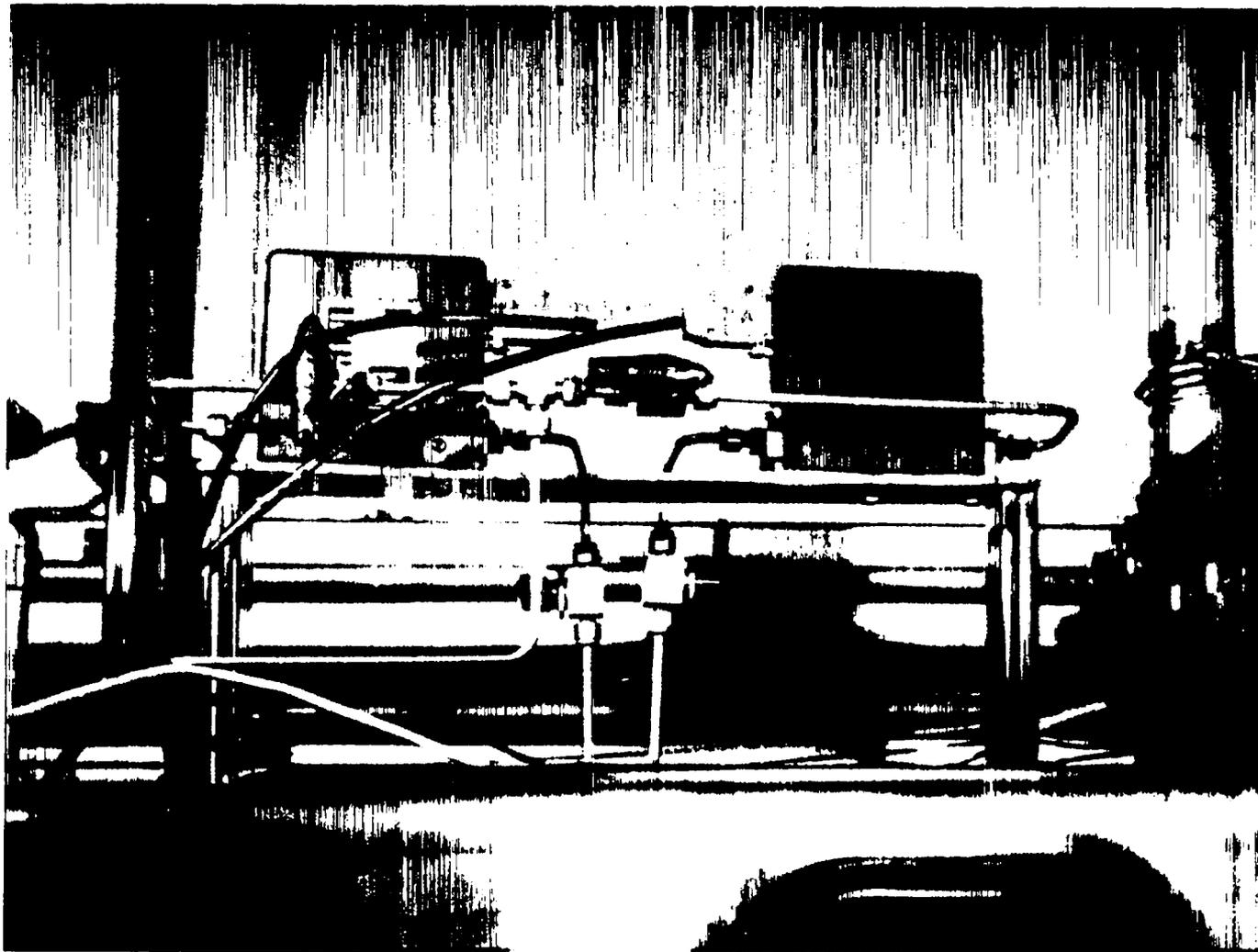


Fig. 3. Flow controllers for phosgene and argon.

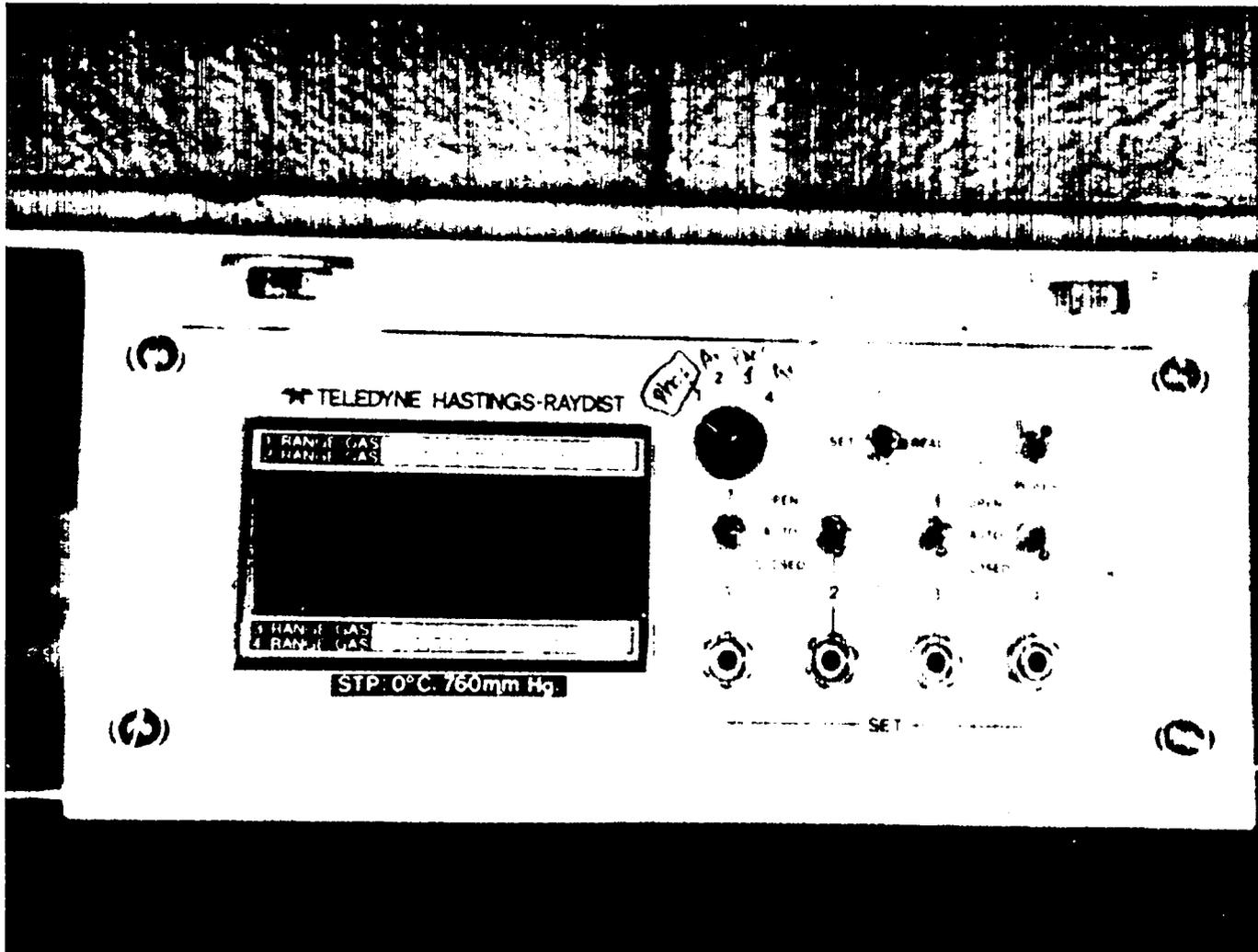


Fig. 4. Teledyne Hastings-Raydist mass flow controller power supply.

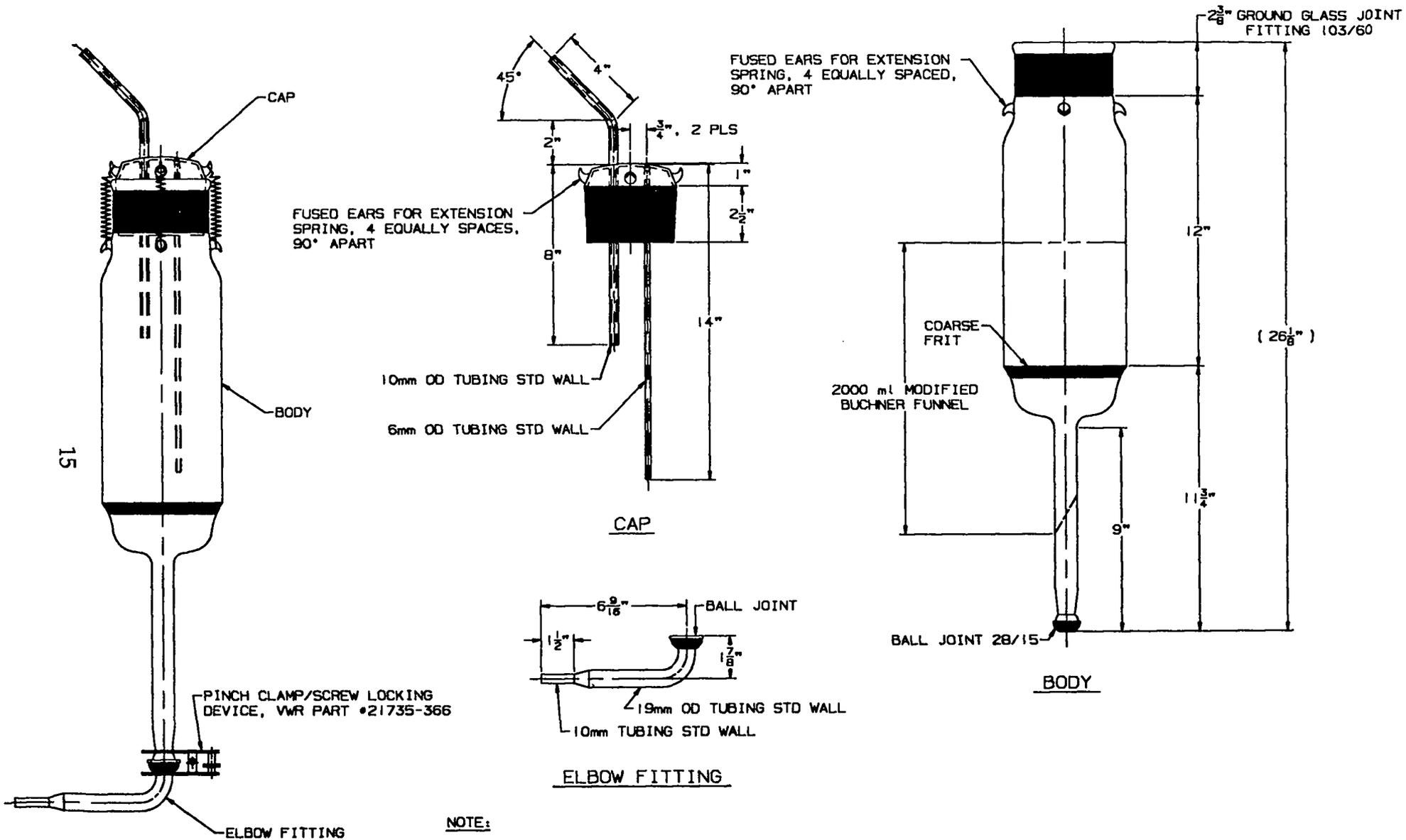


Fig. 5. Pyrex reactor for plutonium trichloride synthesis. (Los Alamos drawing number 26Y-200812)

APPENDIX Plutonium Trichloride Synthesis

Run Number	Feed Lot ID	Feed in grams	Product In grams	Chlorinating Agent	Temperature	Time at 500 C	Plutonium wt %	Chloride wt %	Cl/Pu mole ratio	PuCl ₃ wt % (based on chloride analysis)
PUCL3-111	MPB22ERC1	400.0	501.5	Cl ₂ /CCl ₄	500	13 1/2	69.08	29.8	2.91	96.8
PUCL3-112	MPB22ERC1	500.0	625.9	Cl ₂ /CCl ₄	500	14	69.66	29.6	2.86	96.1
PUCL3-113	MPB22ERC1	500.0	627.5	Cl ₂ /CCl ₄	500	14 1/6	69.41	29.9	2.90	97.1
PUCL3-114	MPB22ERC1	200.0	250.9	CO-Cl ₂	500	6 1/6	68.99	29.6	2.89	96.1
				Cl ₂ /CCl ₄	500	11 1/2				
PUCL3-115	MPB22ERC1	200.0	233.7	CO-Cl ₂	500	7 1/6				
PUCL3-115 Rerun	MPB22ERC1	233.2	250.1	Cl ₂ /CCl ₄	500	2 5/6	68.30	29.3	2.89	95.1
PUCL3-116	MPB22ERC1	200.0	222.2	CO-Cl ₂	500	6	76.21	14.7	1.30	47.7
PUCL3-116 Rerun	MPB22ERC1	219.2	248.4	Cl ₂ /CCl ₄	500	6 1/3	67.71	29.2	2.91	94.8
PUCL3-117	MPB22ERC1	200.0	240.2	CO-Cl ₂	500	10 2/3	71.29	24.6		79.9
PUCL3-117 Rerun	MPB22ERC1	240.2	249.4	Cl ₂ /CCl ₄	500	3 1/3		29.8		96.8
PUCL3-118	MPB22ERC1	445.9	560.1	Cl ₂ /CCl ₄	500	13 1/6	69.47	29.5	2.86	95.8
PUCL3-119	MPB22ERC3	200.0	250.0	Ar/CCl ₄	500	6 5/6	69.94	29.5	2.84	95.8
PUCL3-120	MPB22ERC3	200.0	249.4	Cl ₂ /CCl ₄	500	6 5/6	70.20	29.0	2.78	94.2
PUCL3-121	MPB22ERC3	300.0	375.2	Cl ₂ /CCl ₄	500	7	69.69	29.3	2.83	95.1
PUCL3-122	MPB22ERC3	400.0	463.3	Cl ₂ /CCl ₄	500	6 5/6	75.71	19.3	1.72	62.7
PUCL3-122 Rerun	MPB22ERC3	460.5	495.4	Cl ₂ /CCl ₄	500	6 1/2	69.57	29.4	2.85	95.5
PUCL3-123	MPB22ERC3	400.0	477.3	Cl ₂ /CCl ₄	500	6 5/6	71.93	24.9	2.33	80.9
PUCL3-123 Rerun	MPB22ERC3	474.5	497.2	Cl ₂ /CCl ₄	500	5 2/3	69.64	29.6	2.87	96.1
PUCL3-126	MPB22ERC3	400.0	501.5	Cl ₂ /CCl ₄	500	11 1/6	69.42	29.9	2.90	97.1
PUCL3-127	MPB22ERC3	534.3	667.9	Cl ₂ /CCl ₄	500	20 1/2	69.92	29.9	2.88	97.1
PUCL3-128	MPB36ERC4	200.0	241.4	Cl ₂ /CCl ₄	500	6 2/3	69.53	29.7	2.88	96.4
PUCL3-129	MPB36ERC4	400.0	501.7	Cl ₂ /CCl ₄	500	14	69.38	30.0	2.91	97.4
PUCL3-131	MPB36ERC4/MPB22ERC4	400.0	498.2	Cl ₂ /CCl ₄	500	12	69.71	29.4	2.84	95.5
PUCL3-133	MPB36ERC4	400.0	501.5	Cl ₂ /CCl ₄	500	18 1/6	69.47	29.9	2.90	97.1
PUCL3-134	MPB36ERC4	400.0	502.6	Cl ₂ /CCl ₄	500	16 1/2	69.03	30.3	2.96	98.4
PUCL3-136	MPB22ERC2	200.0	249.8	Cl ₂ /CCl ₄	500	10 1/2	69.57	29.3	2.84	95.1
PUCL3-137	MPB22ERC2	200.0	251.5	Cl ₂ /CCl ₄	500	10 1/6	69.37	30.1	2.93	97.7
PUCL3-138	MPB22ERC2	200.0	250.6	Cl ₂ /CCl ₄	500	8 1/2	69.51	30.3	2.94	98.4
PUCL3-139	MPB22ERC2	200.0	251.2	COCl ₂	500	6 1/2	69.44	30.2	2.93	98.1
PUCL3-140	MPB22ERC2	200.0	251.6	COCl ₂	500	5 1/2	69.52	30.4	2.95	98.7
PUCL3-141	MPB22ERC2	300.0	377.4	COCl ₂	500	8 1/6	68.99	30.2	2.95	98.1
PUCL3-142	MPB22ERC2	400.0	501.3	COCl ₂	500	10 5/6	68.91	30.2	2.95	98.1

APPENDIX Plutonium Trichloride Synthesis

Run Number	Feed Lot ID	Feed in grams	Product in grams	Chlorinating Agent	Temperature	Time at 500 C	Plutonium wt %	Chloride wt %	Cl/Pu mole ratio	PuCl ₃ wt % (based on chloride analysis)
PUCL3-144	MPB22ERC2/KHCB1C3	300.0	376.6	COCl ₂	500	7 5/6	69.09	30.4	2.95	98.7
PUCL3-149	MPB43ERC3	300.0	379.8	COCl ₂	500	9 1/3	69.02	30.2	2.95	98.1
PUCL3-150	MPB43ERC3	300.0	378.6	COCl ₂	500	9 2/3	69.03	30.3	2.96	98.4
PUCL3-151	MPB43ERC3	300.0	378.7	COCl ₂	500	9	69.29	30.0	2.95	97.4
PUCL3-153	MPB43ERC3	200.0	252.1	COCl ₂	500	7 1/2	68.85	31.0	3.04	100.7
PUCL3-154	MPB43ERC3	200.0	252.2	COCl ₂	500	7 5/6	69.25	30.3	2.95	98.4
PUCL3-155	MPB43ERC3	300.0	378.7	COCl ₂	500	10 1/6	69.16	29.9	2.91	97.1
PUCL3-156	MPB43ERC3	300.0	378.7	COCl ₂	500	11	69.13	29.9	2.92	97.1
PUCL3-157	MPB43ERC3	300.0	378.6	COCl ₂	500	9 2/3	69.33	30.0	2.92	97.4
PUCL3-158	MPB43ERC3	300.0	378.4	COCl ₂	500	9 1/3	69.33	30.1	2.93	97.7
PUCL3-159	MPB43ERC3	245.9	310.5	COCl ₂	500	7 5/6	69.05	30.1	2.94	97.7
PUCL3-160	MPB56ERC4	300.0	375.4	COCl ₂	500	9 2/3	69.22	30.2	2.94	98.1
PUCL3-161	MPB56ERC4	300.0	375.7	COCl ₂	500	8 1/2	69.07	30.2	2.95	98.1
PUCL3-162	MPB56ERC4	300.0	374.3	COCl ₂	500	7 2/3	69.81	29.4	2.84	95.5
PUCL3-163	MPB56ERC4	300.0	373.5	COCl ₂	500	7 2/3	69.42	29.2	2.84	94.8
PUCL3-164	MPB56ERC4	300.0	371.5	COCl ₂	500	8	70.36	28.7	2.75	93.2
PUCL3-165	MPB56ERC4	300.0	373.6	COCl ₂	500	8 1/3	69.70	28.9	2.80	93.8
PUCL3-166	MPB56ERC4	300.0	373.7	COCl ₂	500	9 1/6	70.03	29.1	2.80	94.5
PUCL3-167	MPB56ERC4	363.8	453.0	COCl ₂	500	11	69.67	29.6	2.86	96.1
PUCL3-168	MPB59ERC4	300.0	373.4	COCl ₂	500	9 1/6	69.74	29.5	2.85	95.8
PUCL3-169	PUT63HFC1	200.0	252.7	COCl ₂	500	7	69.08	30.3	2.96	98.4
PUCL3-170	PUT63HFC1	199.7	252.3	COCl ₂	500	7	69.24	30.5	2.97	99.0
PUCL3-171	MPB59ERC4	300.0	375.0	COCl ₂	500	12 1/6	69.01	29.8	2.91	96.8
PUCL3-172	MPB56ERC3/KHCB2C4	296.6	369.3	COCl ₂	500	10 5/6	68.49	29.7	2.92	96.4
PUCL3-173	MPB56ERC3	300.0	373.3	COCl ₂	500	11 1/6	69.73	29.2	2.82	94.8
PUCL3-174	MPB56ERC3	200.0	251.0	COCl ₂	500	7	69.01	30.0	2.93	97.4
PUCL3-175	MPB56ERC3	300.0	376.0	COCl ₂	500	11	69.09	30.4	2.97	98.7
PUCL3-176	MPB56ERC3	300.0	371.0	COCl ₂	500	9	69.99	28.8	2.77	93.5
PUCL3-177	MPB56ERC3	300.0	373.9	COCl ₂	500	9 2/3	68.89	29.5	2.89	95.8
PUCL3-178	MPB56ERC3	300.0	374.0	COCl ₂	500	9 1/6	68.95	29.5	2.88	95.8
PUCL3-179	MPB56ERC3	350.0	436.9	COCl ₂	500	11 2/3	69.71	29.5	2.85	95.8
PUCL3-180	MPB56ERC3	350.4	438.3	COCl ₂	500	10 1/3	69.26	30.0	2.92	97.4
PUCL3-181	MPB56ERC2	200.0	251.0	COCl ₂	500	5 2/3	68.62	30.1	2.96	97.7
PUCL3-182	MPB56ERC2	300.0	373.1	COCl ₂	500	9 1/3	69.25	28.8	2.82	93.5

APPENDIX Plutonium Trichloride Synthesis

Run Number	Feed Lot ID	Feed in grams	Product in grams	Chlorinating Agent	Temperature	Time at 500 C	Plutonium wt %	Chloride wt %	Cl/Pu mole ratio	PuCl ₃ wt % (based on chloride analysis)
PUCL3-187	MPB56ERC2	200.0	251.2	COCl ₂	500	6 1/2	68.79	30.4	2.98	98.7
PUCL3-188	MPB56ERC2	300.0	376.6	COCl ₂	500	8 1/6	68.98	30.5	2.98	99.0
PUCL3-189	MPB56ERC2	350.4	440.0	COCl ₂	500	8 1/6	69.10	30.5	2.98	99.0
PUCL3-190	MPB74ERC4	300.0	378.6	COCl ₂	500	8 1/6	69.15	30.4	2.96	98.7
PUCL3-191	MPB74ERC4	350.0	435.4	COCl ₂	500	8 1/6	69.24	28.8	2.80	93.5
PUCL3-192	MPB74ERC4	350.0	440.7	COCl ₂	500	8 1/2	68.69	30.2	2.96	98.1
PUCL3-193	MPB74ERC4	350.0	440.2	COCl ₂	500	8 1/3	69.09	30.5	2.98	99.0
PUCL3-194	MPB74ERC4	400.0	502.6	COCl ₂	500	8 1/3	69.40	30.4	2.95	98.7
PUCL3-195	MPB74ERC4	400.0	502.3	COCl ₂	500	8 1/6	69.01	30.5	2.98	99.0
PUCL3-196	MPB74ERC4	400.0	503.6	COCl ₂	500	8 1/6	69.00	30.3	2.96	98.4
PUCL3-197	MPB75ERC1	400.0	502.1	COCl ₂	500	8 1/6	69.04	30.2	2.95	98.1
PUCL3-198	MPB75ERC1/MPB74ERC4	400.0	502.4	COCl ₂	500	8 1/6	68.50	30.5	3.00	99.0
PUCL3-199	MPB75ERC1	400.0	499.6	COCl ₂	500	8 1/3	69.80	29.5	2.85	95.8
PUCL3-200	MPB75ERC1	400.0	502.2	COCl ₂	500	8 1/2	69.26	30.2	2.94	98.1
PUCL3-201	MPB75ERC1	400.0	500.9	COCl ₂	500	8 1/3	69.03	29.6	2.89	96.1
PUCL3-202	MPB75ERC1	400.0	501.7	COCl ₂	500	8 1/6	68.99	30.3	2.96	98.4
PUCL3-203	MPB78ERC4	400.0	504.4	COCl ₂	500	9 1/6	69.19	30.5	2.96	99.0
PUCL3-204	MPB78ERC4/MPB75ERC1	400.0	503.1	COCl ₂	500	8 1/6	69.40	30.5	2.97	99.0
PUCL3-205	MPB78ERC4	400.0	504.1	COCl ₂	500	8 1/6	69.30	30.2	2.94	98.1
PUCL3-206	MPB78ERC4	400.0	504.8	COCl ₂	500	8 1/3	69.22	30.4	2.96	98.7
PUCL3-207	MPB78ERC4	400.0	504.6	COCl ₂	500	8 2/3	69.19	29.9	2.91	97.1
PUCL3-208	MPB78ERC4	400.0	504.3	COCl ₂	500	8 1/6	69.24	30.2	2.94	98.1
PUCL3-209	MPB80ERC4	400.0	505.0	COCl ₂	500	8 1/3	69.25	30.3	2.95	98.4
PUCL3-210	MPB78ERC4/MPB80ERC4	400.0	504.7	COCl ₂	500	8 1/6	69.35	30.1	2.93	97.7
PUCL3-211	MPB80ERC4	400.0	504.8	COCl ₂	500	8 1/6	69.28	30.4	2.96	98.7
PUCL3-212	MPB80ERC4	400.0	498.3	COCl ₂	500	11 5/6	69.97	29.4	2.83	95.5
PUCL3-213	MPB80ERC4	400.0	504.4	COCl ₂	500	8 1/6	69.27	30.5	2.97	99.0
PUCL3-214	MPB80ERC4	410.7	517.6	COCl ₂	500	8 1/6	69.30	30.5	2.97	99.0
PUCL3-215	MSTPPB53C4	400.0	503.5	COCl ₂	500	8 1/6	69.27	30.8	3.00	100.0
PUCL3-216	MSTPPB53C4	400.0	503.3	COCl ₂	500	8 2/3	69.35	30.5	2.96	99.0
PUCL3-217	MSTPPB53C4	400.0	503.0	COCl ₂	500	8 1/6	68.81	30.0	2.95	97.4
PUCL3-218	MSTPPB53C4	400.0	503.0	COCl ₂	500	8 1/6	69.05	30.0	2.93	97.4
PUCL3-219	MSTPPB53C4	400.0	507.6	COCl ₂	500	8 1/6	69.16	30.3	2.95	98.4
PUCL3-220	MPB84ERC4	400.0	499.2	COCl ₂	500	8 1/3	69.12	30.1	2.94	97.7

APPENDIX Plutonium Trichloride Synthesis

Run Number	Feed Lot ID	Feed in grams	Product In grams	Chlorinating Agent	Temperature	Time at 500 C	Plutonium wt %	Chloride wt %	Cl/Pu mole ratio	PuCl ₃ wt % (based on chloride analysis)
PUCL3-226	MPB84ERC4	351.7	438.5	COCl ₂	500	8	69.22	30.1	2.93	97.7
PUCL3-227	MPB85ERC4	400.0	504.0	COCl ₂	500	8 1/3	69.27	30.0	2.92	97.4
PUCL3-228	MPB85ERC4	400.0	503.6	COCl ₂	500	8 1/2	69.33	30.0	2.96	98.7
PUCL3-229	MPB85ERC4	400.0	503.8	COCl ₂	500	8 2/3	69.31	30.4	2.96	98.7
PUCL3-230	MPB85ERC4	400.0	504.3	COCl ₂	500	8 1/6	69.42	30.4	2.93	98.1
PUCL3-231	MPB93ERC5	400.0	502.2	COCl ₂	500	9 1/3	69.44	30.2	2.97	99.4
PUCL3-232	MPB93ERC5/MPB85ERC4	400.0	497.3	COCl ₂	500	8 1/6	69.74	30.6	2.84	95.5
	MPB56ERC4A							29.4		
PUCL3-233	MPB94ERC5	400.0	501.3	COCl ₂	500	9	69.45	30.3	2.94	98.4
PUCL3-234	MPB93ERC5/MPB94ERC5	400.0	501.9	COCl ₂	500	8 2/3	69.08	30.2	2.95	98.1
PUCL3-236	MPB94ERC5	400.0	500.5	COCl ₂	500	9 1/6	69.46	29.7	2.88	96.4
PUCL3-237	MPB95ERC5	400.0	504.9	COCl ₂	500	8 1/3	69.31	30.4	2.96	98.7
PUCL3-238	MPB94ERC5/MPB95ERC5	400.0	502.7	COCl ₂	500	8 1/3	69.37	30.3	2.95	98.4
	MPB96ERC5/MPB97ERC5		495.9			8 1/3				
PUCL3-239	MPB97ERC5	400.0	551.1	COCl ₂	500	10	70.34	29.0	2.78	94.2
PUCL3-240	MPB97ERC5	444.3	501.2	COCl ₂	500	8 1/3	70.06	28.9	2.78	93.8
PUCL3-241	MPB100ERC4	400.0	501.3	COCl ₂	500	7 5/6	69.25	30.5	2.97	99.0
PUCL3-242	MPB100ERC4	400.0	497.8	COCl ₂	500	7 2/3	69.43	29.7	2.89	96.4
PUCL3-243	MPB98ERC5	400.0	496.1	COCl ₂	500	10 1/3	69.33	29.8	2.90	96.8
PUCL3-244	MPB104ERC4	400.0	494.9	COCl ₂	500	9	70.35	28.6	2.74	92.9
PUCL3-245	MPB98ERC5/MPB100ERC4	400.0	502.0	COCl ₂	500	9	69.50	30.0	2.91	97.4
	MPB104ERC4	400.0								
PUCL3-246	MPB104ERC4	400.0	496.8	COCl ₂	500		68.98	29.6	2.89	96.1
PUCL3-247	MPB101ERC4	400.0	496.8	COCl ₂	500	9	69.94	28.6	2.76	92.9
PUCL3-248	MPB101ERC4/MPB104ERC4	400.0	496.8	COCl ₂	500	9	69.96	29.1	2.80	94.5
PUCL3-250	MPB101ERC4	400.0	494.6	COCl ₂	500	9 1/6	70.22	29.4	2.80	95.5
PUCL3-251	MPB101ERC4/MSTPPB62C5	400.0	501.0	COCl ₂	500	8	69.14	30.5	2.97	99.0
PUCL3-252	MSTPPB62C5	400.0	499.2	COCl ₂	500	10	69.78	29.8	2.89	96.8
PUCL3-253	MSTPPB62C5/MSTPPB63C5	400.0	500.6	COCl ₂	500	8 1/3	69.45	30.3	2.94	98.4
PUCL3-254	MSTPPB63C5	400.0	500.8	COCl ₂	500	8 1/6	69.20	29.4	2.86	95.5
PUCL3-255	MSTPPB63C5	386.6	478.8	COCl ₂	500	8 1/2	70.42	26.9	2.57	87.4

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