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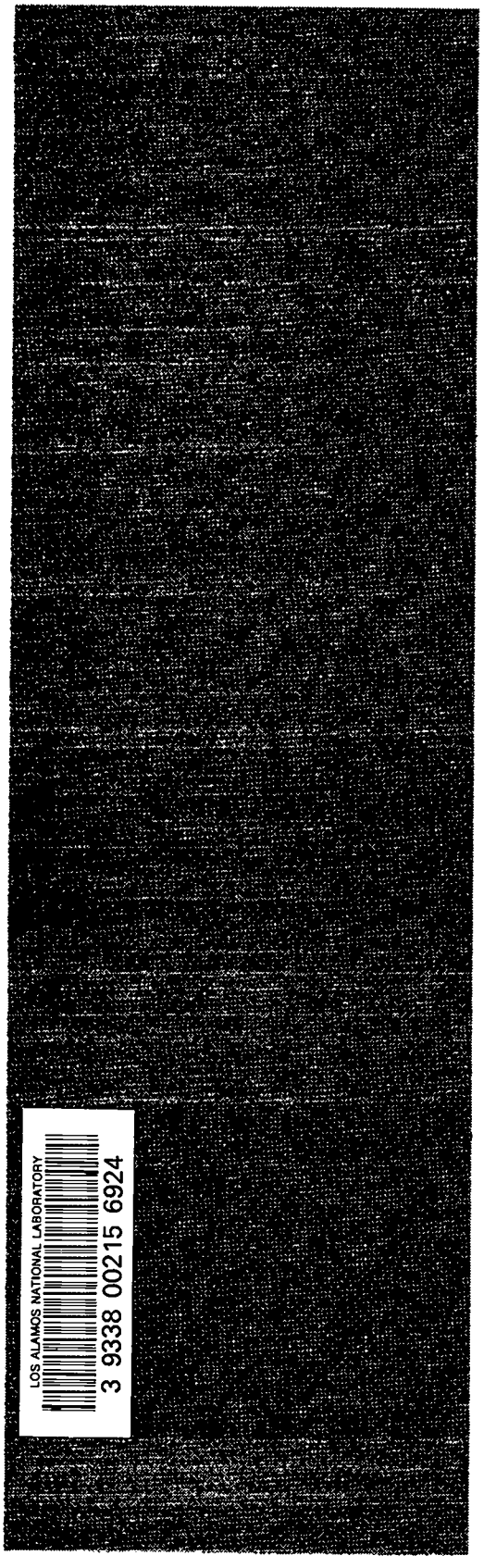

The Synthesis of Plutonium Trichloride by Chlorination of Plutonium Dioxide with Phosgene

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Edited by Jeff Skiby, Group IS-1

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LA-12631-MS

UC-711
Issued: October 1993

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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 (Cl_2-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 Cl_2-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 ($PuH_{2.7}$) 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 PuO_2-$PuCl_3$ 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 cm^3/min overnight, removing traces of $COCl_2$, while the reactor cools. The argon also blankets the $PuCl_3$ 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 $COCl_2$ 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 $PuCl_3$ 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 $PuCl_3$ 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 PuO_2 relative to the remainder of the $PuCl_3$.

In the early stages of this work, a single-chambered scrubber was used for elimination of $COCl_2$ 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 $COCl_2$ 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 scubber unit with fresh caustic then is used for removing further unreacted $COCl_2$. 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 PuO_2 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 PuO_2 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 $PuCl_3$ in the product.

Through experiment PUCL3-138, a stream of Cl_2 saturated with CCl_4 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 $COCl_2$. Therefore, efforts were transferred to the use of $COCl_2$ as a chlorinating agent in December 1987.

The use of CO-Cl_2 mixtures to chlorinate PuO_2 was reported by Rasmussen and Hopkins but no supporting data appeared in the paper other than the fact it was not as reactive as $COCl_2$ [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 PuO_2.

For experiment PUCL3-115, the flow rates of Cl_2 and CO were nominally equal, and a weight increase of 33.7 g was found for 200 g PuO_2 feed. The succeeding chlorination experiment used a Cl_2 to CO flow ratio of 1.5, and the weight gain was 22.2 g for the same quantity of PuO_2 feed as in experiment PUCL3-115. The observed chloride content was 14.7 wt %. (The theoretical weight percent for chloride in $PuCl_3$ is 30.8.) The last work with CO-Cl_2 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 Cl_2 appear to improve the conversion of PuO_2 to $PuCl_3$, based on the increase in weight, but Cl_2-CCl_4 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 PuO_2 feed and chlorination with Cl_2-CCl_4 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 (Al_2O_3). They report results similar for chlorination with $COCl_2$ directly. Thus, the COCl radical is probably the species responsible for chlorination of oxide compounds by $COCl_2$. In addition, Ferran et al. [17] have observed COCl formation at the ionization filament in mass spectrometric studies of gaseous effluents from chlorination of PuO_2 by $COCl_2$.

The reaction of $COCl_2$ with PuO_2 has been shown to proceed according to Equation (1) [8,17]:

$$PuO_2 + 2\,COCl_2 = PuCl_3 + 1/2\,Cl_2 + 2\,CO_2 \quad ; \qquad (1)$$

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

$$PuO_2 + COCl_2 = PuOCl + CO_2 + 1/2\,Cl_2 \quad \text{and} \qquad (2)$$

$$PuOCl + COCl_2 = PuCl_3 + CO_2 \; . \qquad (3)$$

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 $PuCl_3$ product is shown in Table I for experiments PUCL3-118 through PUCL3-138, excluding PUCL3-122 and PUCL3-123, where Cl_2-CCl_4 or CCl_4 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 $COCl_2$ 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 Cl_2-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
Cl_2-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 PuO_2 feed and $PuCl_3$ product.

	Al ppm	B ppm	Si ppm
MBP78ER*	10	<5	370
PUCl3-205	10	<5	190
PUCl3-216	590	<5	330
MSTPPB53*	8	<5	70
PUCl3-215	700	<5	320
PUCl3-216	7	<5	100
PUCl3-217	<5	<5	40

*PuO_2 feeds to chlorination.

Future Work

Work by Ferran et al. [17] has shown that, for a 400-g feed batch of PuO_2, evolution of carbon dioxide (CO_2) slows considerably after about four hours of chlorination at 500°C. According to Equation (1), CO_2 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 $PuCl_3$. 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

The authors are grateful to the excellent support given this project by the Mechanical and Electronics Support Division (MEC-10) Glass Shop personnel, including Bill Fox and Max Newman. Lorenzo Jaramillo of the Nuclear Materials Technology Division (NMT-3) designed and tested the multichambered scrubber. Wayne Smythe of NMT-3 designed and installed the original Cl_2 and $COCl_2$ gas delivery systems. Don Temer, Tom Marshall, and R. Brad Roof of CLS-1 supervised the analysis of $PuCl_3$ for plutonium, chloride, and major crystalline components by x-ray diffraction, respectively.

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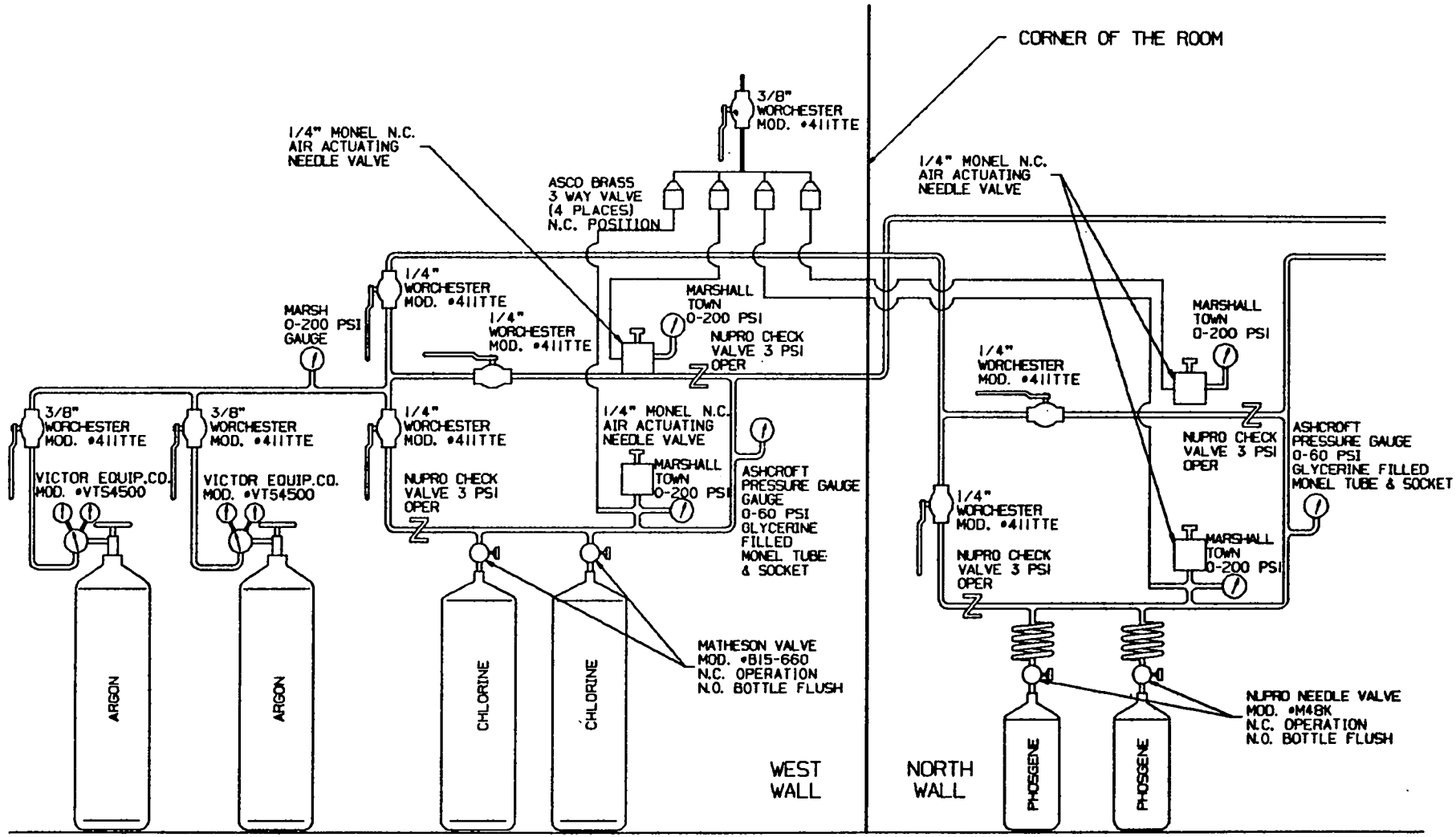

Fig. 1. Phosgene and chlorine manifolds in room 116, PF-3 Building at TA-55. (Los Alamos drawing number 26Y-200581)

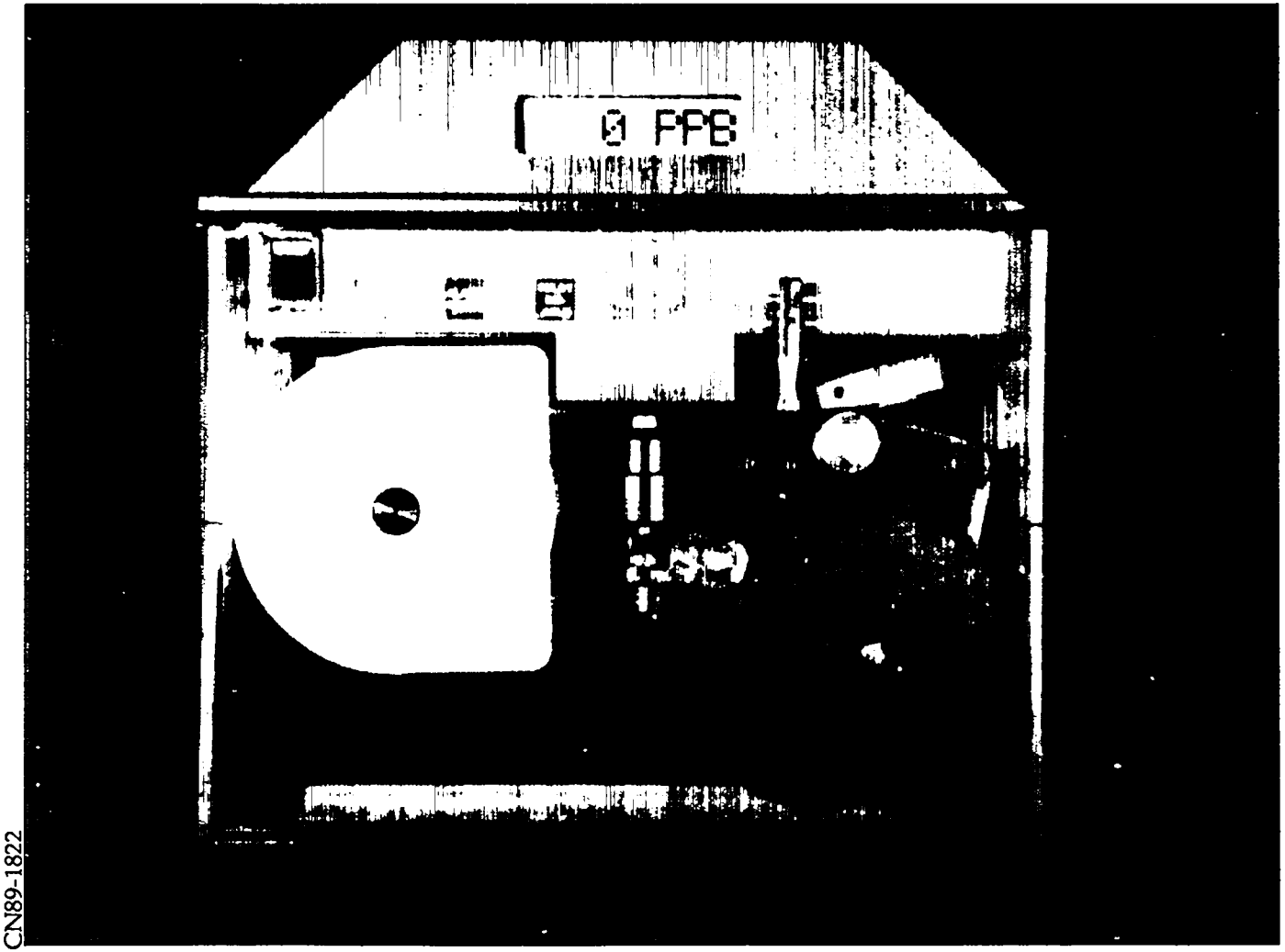

Fig. 2. MDA's TLD-1 toxic gas detector for phosgene with Chemcassette.

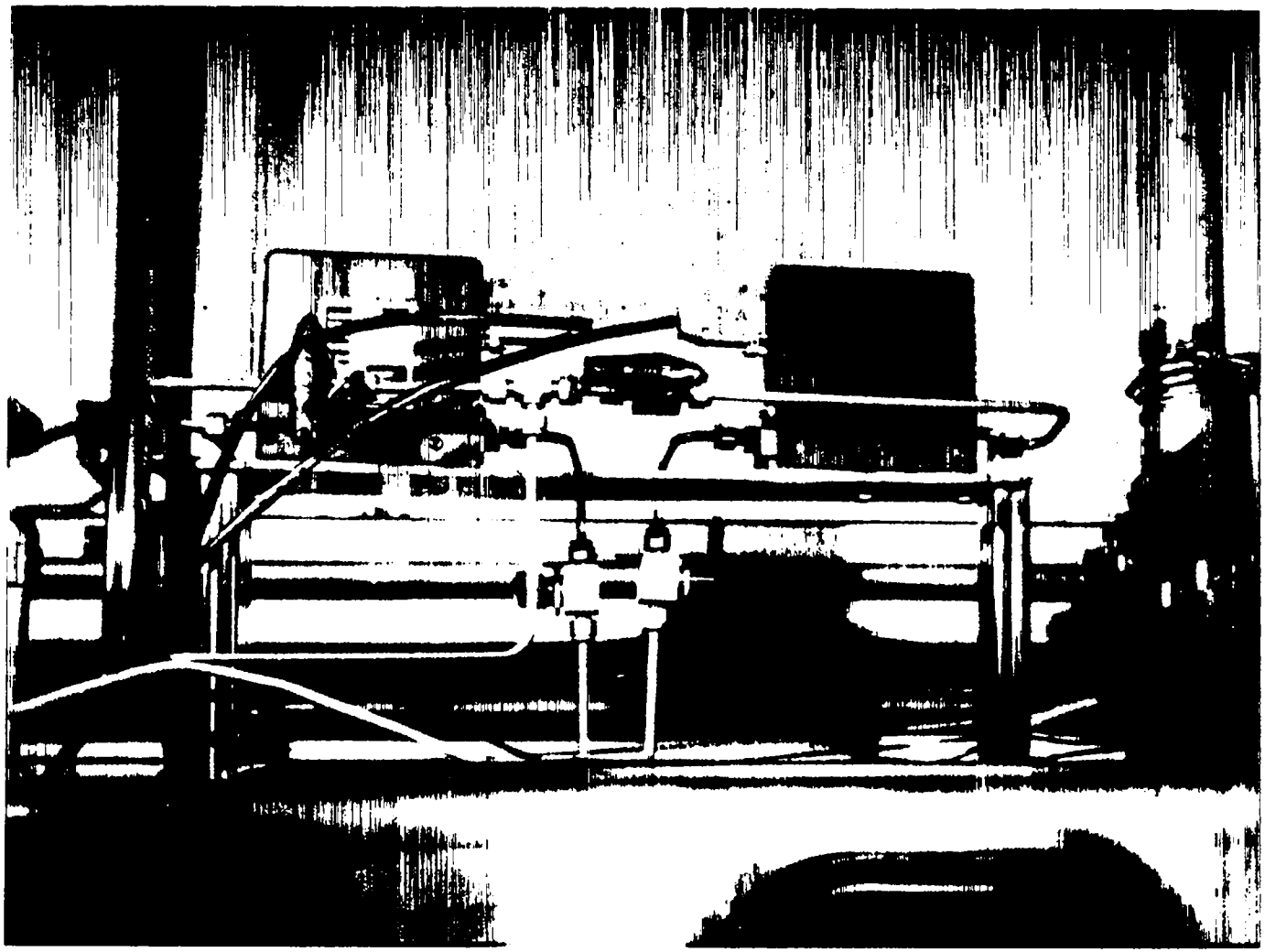

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Fig. 3. Flow controllers for phosgene and argon.

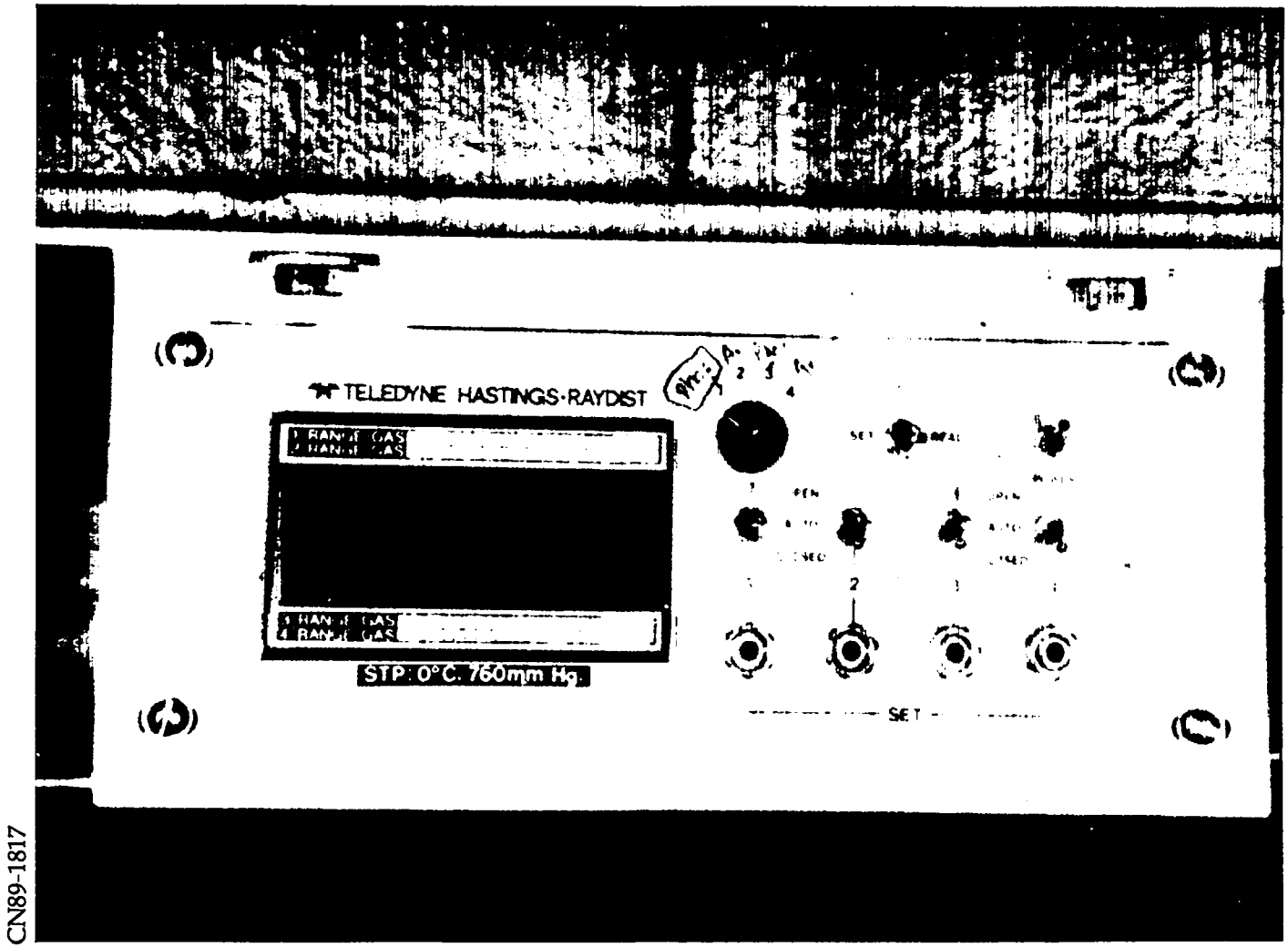
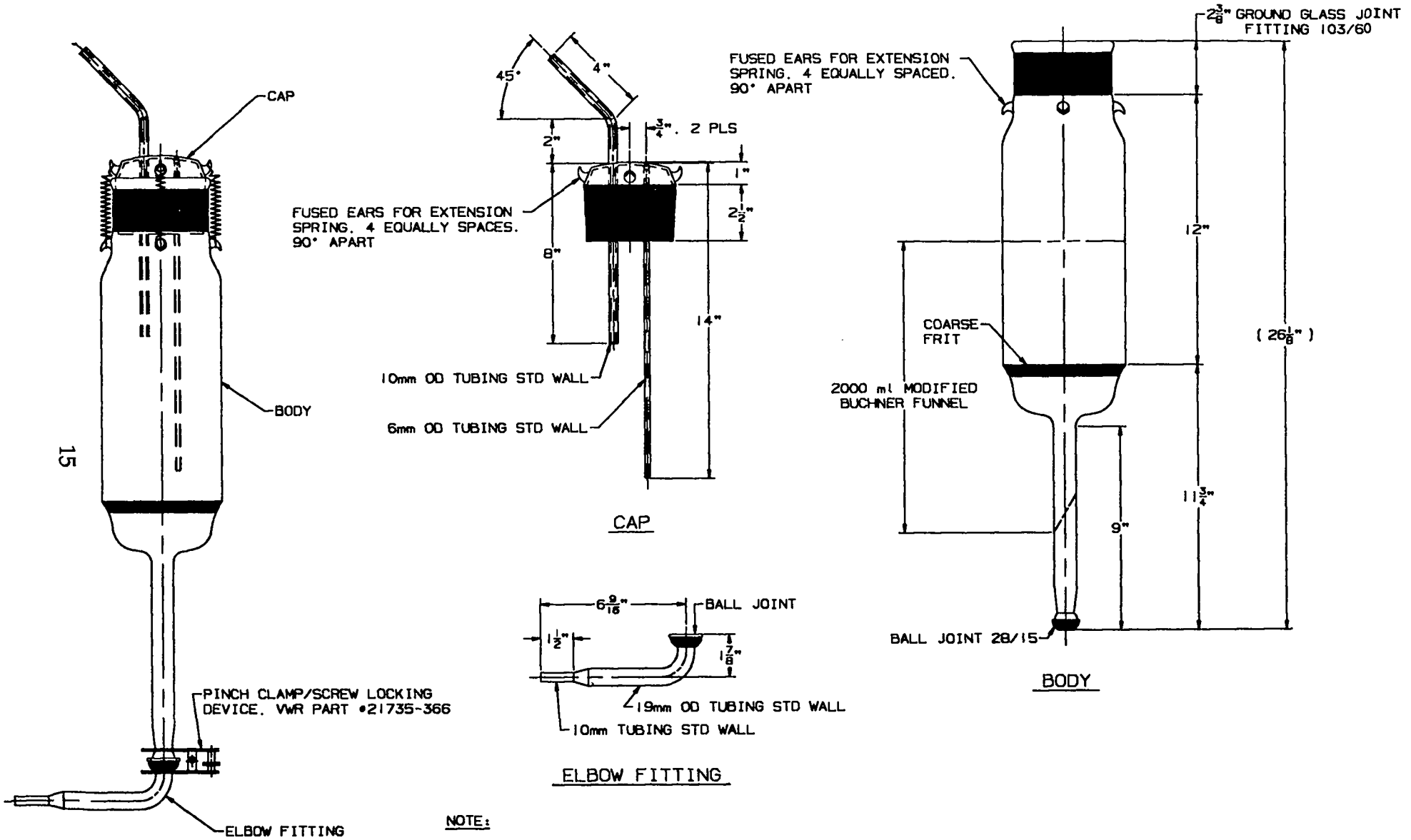


Fig. 4. Teledyne Hastings-Raydist mass flow controller power supply.



NOTE:

9. ALL GLASSWARE TO BE PYREX.

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	PuCl3 wt % (based on chloride analysis)
PUCL3-111	MPB22ERC1	400.0	501.5	Cl2/CCl4	500	13 1/2	69.08	29.8	2.91	96.8
PUCL3-112	MPB22ERC1	500.0	625.9	Cl2/CCl4	500	14	69.66	29.6	2.86	96.1
PUCL3-113	MPB22ERC1	500.0	627.5	Cl2/CCl4	500	14 1/6	69.41	29.9	2.90	97.1
PUCL3-114	MPB22ERC1	200.0	250.9	CO-Cl2	500	6 1/6	68.99	29.6	2.89	96.1
				Cl2/CCl4	500	11 1/2				
PUCL3-115	MPB22ERC1	200.0	233.7	CO-Cl2	500	7 1 /6				
PUCL3-115 Rerun	MPB22ERC1	233.2	250.1	Cl2/CCl4	500	2 5/6	68.30	29.3	2.89	95.1
PUCL3-116	MPB22ERC1	200.0	222.2	CO-Cl2	500	6	76.21	14.7	1.30	47.7
PUCL3-116 Rerun	MPB22ERC1	219.2	248.4	Cl2/CCl4	500	6 1/3	67.71	29.2	2.91	94.8
PUCL3-117	MPB22ERC1	200.0	240.2	CO-Cl2	500	10 2/3	71.29	24.6		79.9
PUCL3-117 Rerun	MPB22ERC1	240.2	249.4	Cl2/CCl4	500	3 1/3		29.8		96.8
PUCL3-118	MPB22ERC1	445.9	560.1	Cl2/CCl4	500	13 1/6	69.47	29.5	2.86	95.8
PUCL3-119	MPB22ERC3	200.0	250.0	Ar/CCl4	500	6 5/6	69.94	29.5	2.84	95.8
PUCL3-120	MPB22ERC3	200.0	249.4	Cl2/CCl4	500	6 5/6	70.20	29.0	2.78	94.2
PUCL3-121	MPB22ERC3	300.0	375.2	Cl2/CCl4	500	7	69.69	29.3	2.83	95.1
PUCL3-122	MPB22ERC3	400.0	463.3	Cl2/CCl4	500	6 5/6	75.71	19.3	1.72	62.7
PUCL3-122 Rerun	MPB22ERC3	460.5	495.4	Cl2/CCl4	500	6 1/2	69.57	29.4	2.85	95.5
PUCL3-123	MPB22ERC3	400.0	477.3	Cl2/CCl4	500	6 5/6	71.93	24.9	2.33	80.9
PUCL3-123 Rerun	MPB22ERC3	474.5	497.2	Cl2/CCl4	500	5 2/3	69.64	29.6	2.87	96.1
PUCL3-126	MPB22ERC3	400.0	501.5	Cl2/CCl4	500	11 1/6	69.42	29.9	2.90	97.1
PUCL3-127	MPB22ERC3	534.3	667.9	Cl2/CCl4	500	20 1/2	69.92	29.9	2.88	97.1
PUCL3-128	MPB36ERC4	200.0	241.4	Cl2/CCl4	500	6 2/3	69.53	29.7	2.88	96.4
PUCL3-129	MPB36ERC4	400.0	501.7	Cl2/CCl4	500	14	69.38	30.0	2.91	97.4
PUCL3-131	MPB36ERC4/MPB22ERC4	400.0	498.2	Cl2/CCl4	500	12	69.71	29.4	2.84	95.5
PUCL3-133	MPB36ERC4	400.0	501.5	Cl2/CCl4	500	18 1/6	69.47	29.9	2.90	97.1
PUCL3-134	MPB36ERC4	400.0	502.6	Cl2/CCl4	500	16 1/2	69.03	30.3	2.96	98.4
PUCL3-136	MPB22ERC2	200.0	249.8	Cl2/CCl4	500	10 1/2	69.57	29.3	2.84	95.1
PUCL3-137	MPB22ERC2	200.0	251.5	Cl2/CCl4	500	10 1/6	69.37	30.1	2.93	97.7
PUCL3-138	MPB22ERC2	200.0	250.6	Cl2/CCl4	500	8 1/2	69.51	30.3	2.94	98.4
PUCL3-139	MPB22ERC2	200.0	251.2	COCl2	500	6 1/2	69.44	30.2	2.93	98.1
PUCL3-140	MPB22ERC2	200.0	251.6	COCl2	500	5 1/2	69.52	30.4	2.95	98.7
PUCL3-141	MPB22ERC2	300.0	377.4	COCl2	500	8 1/6	68.99	30.2	2.95	98.1
PUCL3-142	MPB22ERC2	400.0	501.3	COCl2	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	PuCl3 wt % (based on chloride analysis)
PUCL3-144	MPB22ERC2/KHCB1C3	300.0	376.6	COCl2	500	7 5/6	69.09	30.4	2.95	98.7
PUCL3-149	MPB43ERC3	300.0	379.8	COCl2	500	9 1/3	69.02	30.2	2.95	98.1
PUCL3-150	MPB43ERC3	300.0	378.6	COCl2	500	9 2/3	69.03	30.3	2.96	98.4
PUCL3-151	MPB43ERC3	300.0	378.7	COCl2	500	9	69.29	30.0	2.95	97.4
PUCl3-153	MPB43ERC3	200.0	252.1	COCl2	500	7 1/2	68.85	31.0	3.04	100.7
PUCL3-154	MPB43ERC3	200.0	252.2	COCl2	500	7 5/6	69.25	30.3	2.95	98.4
PUCL3-155	MPB43ERC3	300.0	378.7	COCl2	500	10 1/6	69.16	29.9	2.91	97.1
PUCL3-156	MPB43ERC3	300.0	378.7	COCl2	500	11	69.13	29.9	2.92	97.1
PUCL3-157	MPB43ERC3	300.0	378.6	COCl2	500	9 2/3	69.33	30.0	2.92	97.4
PUCL3-158	MPB43ERC3	300.0	378.4	COCl2	500	9 1/3	69.33	30.1	2.93	97.7
PUCL3-159	MPB43ERC3	245.9	310.5	COCl2	500	7 5/6	69.05	30.1	2.94	97.7
PUCL3-160	MPB56ERC4	300.0	375.4	COCl2	500	9 2/3	69.22	30.2	2.94	98.1
PUCL3-161	MPB56ERC4	300.0	375.7	COCl2	500	8 1/2	69.07	30.2	2.95	98.1
PUCL3-162	MPB56ERC4	300.0	374.3	COCl2	500	7 2/3	69.81	29.4	2.84	95.5
PUCL3-163	MPB56ERC4	300.0	373.5	COCl2	500	7 2/3	69.42	29.2	2.84	94.8
PUCL3-164	MPB56ERC4	300.0	371.5	COCl2	500	8	70.36	28.7	2.75	93.2
PUCL3-165	MPB56ERC4	300.0	373.6	COCl2	500	8 1/3	69.70	28.9	2.80	93.8
PUCL3-166	MPB56ERC4	300.0	373.7	COCl2	500	9 1/6	70.03	29.1	2.80	94.5
PUCL3-167	MPB56ERC4	363.8	453.0	COCl2	500	11	69.67	29.6	2.86	96.1
PUCL3-168	MPB59ERC4	300.0	373.4	COCl2	500	9 1/6	69.74	29.5	2.85	95.8
PUCL3-169	PUT63HFC1	200.0	252.7	COCl2	500	7	69.08	30.3	2.96	98.4
PUCL3-170	PUT63HFC1	199.7	252.3	COCl2	500	7	69.24	30.5	2.97	99.0
PUCL3-171	MPB59ERC4	300.0	375.0	COCl2	500	12 1/6	69.01	29.8	2.91	96.8
PUCL3-172	MPB56ERC3/KHCB2C4	296.6	369.3	COCl2	500	10 5/6	68.49	29.7	2.92	96.4
PUCL3-173	MPB56ERC3	300.0	373.3	COCl2	500	11 1/6	69.73	29.2	2.82	94.8
PUCL3-174	MPB56ERC3	200.0	251.0	COCl2	500	7	69.01	30.0	2.93	97.4
PUCL3-175	MPB56ERC3	300.0	376.0	COCl2	500	11	69.09	30.4	2.97	98.7
PUCL3-176	MPB56ERC3	300.0	371.0	COCl2	500	9	69.99	28.8	2.77	93.5
PUCL3-177	MPB56ERC3	300.0	373.9	COCl2	500	9 2/3	68.89	29.5	2.89	95.8
PUCL3-178	MPB56ERC3	300.0	374.0	COCl2	500	9 1/6	68.95	29.5	2.88	95.8
PUCL3-179	MPB56ERC3	350.0	436.9	COCl2	500	11 2/3	69.71	29.5	2.85	95.8
PUCL3-180	MPB56ERC3	350.4	438.3	COCl2	500	10 1/3	69.26	30.0	2.92	97.4
PUCL3-181	MPB56ERC2	200.0	251.0	COCl2	500	5 2/3	68.62	30.1	2.96	97.7
PUCL3-182	MPB56ERC2	300.0	373.1	COCl2	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	PuCl3 wt % (based on chloride analysis)
PUCL3-187	MPB56ERC2	200.0	251.2	COCl2	500	6 1/2	68.79	30.4	2.98	98.7
PUCL3-188	MPB56ERC2	300.0	376.6	COCl2	500	8 1/6	68.98	30.5	2.98	99.0
PUCL3-189	MPB56ERC2	350.4	440.0	COCl2	500	8 1/6	69.10	30.5	2.98	99.0
PUCL3-190	MPB74ERC4	300.0	378.6	COCl2	500	8 1/6	69.15	30.4	2.96	98.7
PUCL3-191	MPB74ERC4	350.0	435.4	COCl2	500	8 1/6	69.24	28.8	2.80	93.5
PUCL3-192	MPB74ERC4	350.0	440.7	COCl2	500	8 1/2	68.69	30.2	2.96	98.1
PUCL3-193	MPB74ERC4	350.0	440.2	COCl2	500	8 1/3	69.09	30.5	2.98	99.0
PUCL3-194	MPB74ERC4	400.0	502.6	COCl2	500	8 1/3	69.40	30.4	2.95	98.7
PUCL3-195	MPB74ERC4	400.0	502.3	COCl2	500	8 1/6	69.01	30.5	2.98	99.0
PUCL3-196	MPB74ERC4	400.0	503.6	COCl2	500	8 1/6	69.00	30.3	2.96	98.4
PUCL3-197	MPB75ERC1	400.0	502.1	COCl2	500	8 1/6	69.04	30.2	2.95	98.1
PUCL3-198	MPB75ERC1/MPB74ERC4	400.0	502.4	COCl2	500	8 1/6	68.50	30.5	3.00	99.0
PUCL3-199	MPB75ERC1	400.0	499.6	COCl2	500	8 1/3	69.80	29.5	2.85	95.8
PUCL3-200	MPB75ERC1	400.0	502.2	COCl2	500	8 1/2	69.26	30.2	2.94	98.1
PUCL3-201	MPB75ERC1	400.0	500.9	COCl2	500	8 1/3	69.03	29.6	2.89	96.1
PUCL3-202	MPB75ERC1	400.0	501.7	COCl2	500	8 1/6	68.99	30.3	2.96	98.4
PUCL3-203	MPB78ERC4	400.0	504.4	COCl2	500	9 1/6	69.19	30.5	2.96	99.0
PUCL3-204	MPB78ERC4/MPB75ERC1	400.0	503.1	COCl2	500	8 1/6	69.40	30.5	2.97	99.0
PUCL3-205	MPB78ERC4	400.0	504.1	COCl2	500	8 1/6	69.30	30.2	2.94	98.1
PUCL3-206	MPB78ERC4	400.0	504.8	COCl2	500	8 1/3	69.22	30.4	2.96	98.7
PUCL3-207	MPB78ERC4	400.0	504.6	COCl2	500	8 2/3	69.19	29.9	2.91	97.1
PUCL3-208	MPB78ERC4	400.0	504.3	COCl2	500	8 1/6	69.24	30.2	2.94	98.1
PUCL3-209	MPB80ERC4	400.0	505.0	COCl2	500	8 1/3	69.25	30.3	2.95	98.4
PUCL3-210	MPB78ERC4/MPB80ERC4	400.0	504.7	COCl2	500	8 1/6	69.35	30.1	2.93	97.7
PUCL3-211	MPB80ERC4	400.0	504.8	COCl2	500	8 1/6	69.28	30.4	2.96	98.7
PUCL3-212	MPB80ERC4	400.0	498.3	COCl2	500	11 5/6	69.97	29.4	2.83	95.5
PUCL3-213	MPB80ERC4	400.0	504.4	COCl2	500	8 1/6	69.27	30.5	2.97	99.0
PUCL3-214	MPB80ERC4	410.7	517.6	COCl2	500	8 1/6	69.30	30.5	2.97	99.0
PUCL3-215	MSTPPB53C4	400.0	503.5	COCl2	500	8 1/6	69.27	30.8	3.00	100.0
PUCL3-216	MSTPPB53C4	400.0	503.3	COCl2	500	8 2/3	69.35	30.5	2.96	99.0
PUCL3-217	MSTPPB53C4	400.0	503.0	COCl2	500	8 1/6	68.81	30.0	2.95	97.4
PUCL3-218	MSTPPB53C4	400.0	503.0	COCl2	500	8 1/6	69.05	30.0	2.93	97.4
PUCL3-219	MSTPPB53C4	400.0	507.6	COCl2	500	8 1/6	69.16	30.3	2.95	98.4
PUCL3-220	MPB84ERC4	400.0	499.2	COCl2	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	PuCl3 wt % (based on chloride analysis)
PUCL3-226	MPB84ERC4	351.7	438.5	COCl2	500	8	69.22	30.1	2.93	97.7
PUCL3-227	MPB85ERC4	400.0	504.0	COCl2	500	8 1/3	69.27	30.0	2.92	97.4
PUCL3-228	MPB85ERC4	400.0	503.6	COCl2	500	8 1/2	69.33	30.0	2.96	98.7
PUCL3-229	MPB85ERC4	400.0	503.8	COCl2	500	8 2/3	69.31	30.4	2.96	98.7
PUCL3-230	MPB85ERC4	400.0	504.3	COCl2	500	8 1/6	69.42	30.4	2.93	98.1
PUCL3-231	MPB93ERC5	400.0	502.2	COCl2	500	9 1/3	69.44	30.2	2.97	99.4
PUCL3-232	MPB93ERC5/MPB85ERC4	400.0	497.3	COCl2	500	8 1/6	69.74	30.6	2.84	95.5
	MPB56ERC4A							29.4		
PUCL3-233	MPB94ERC5	400.0	501.3	COCl2	500	9	69.45	30.3	2.94	98.4
PUCL3-234	MPB93ERC5/MPB94ERC5	400.0	501.9	COCl2	500	8 2/3	69.08	30.2	2.95	98.1
PUCL3-236	MPB94ERC5	400.0	500.5	COCl2	500	9 1/6	69.46	29.7	2.88	96.4
PUCL3-237	MPB95ERC5	400.0	504.9	COCl2	500	8 1/3	69.31	30.4	2.96	98.7
PUCL3-238	MPB94ERC5/MPB95ERC5	400.0	502.7	COCl2	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	COCl2	500	10	70.34	29.0	2.78	94.2
PUCL3-240	MPB97ERC5	444.3	501.2	COCl2	500	8 1/3	70.06	28.9	2.78	93.8
PUCL3-241	MPB100ERC4	400.0	501.3	COCl2	500	7 5/6	69.25	30.5	2.97	99.0
PUCL3-242	MPB100ERC4	400.0	497.8	COCl2	500	7 2/3	69.43	29.7	2.89	96.4
PUCL3-243	MPB98ERC5	400.0	496.1	COCl2	500	10 1/3	69.33	29.8	2.90	96.8
PUCL3-244	MPB104ERC4	400.0	494.9	COCl2	500	9	70.35	28.6	2.74	92.9
PUCL3-245	MPB98ERC5/MPB100ERC4	400.0	502.0	COCl2	500	9	69.50	30.0	2.91	97.4
	MPB104ERC4	400.0								
PUCL3-246	MPB104ERC4	400.0	496.8	COCl2	500		68.98	29.6	2.89	96.1
PUCL3-247	MPB101ERC4	400.0	496.8	COCl2	500	9	69.94	28.6	2.76	92.9
PUCL3-248	MPB101ERC4/MPB104ERC4	400.0	496.8	COCl2	500	9	69.96	29.1	2.80	94.5
PUCL3-250	MPB101ERC4	400.0	494.6	COCl2	500	9 1/6	70.22	29.4	2.80	95.5
PUCL3-251	MPB101ERC4/MSTPPB62C5	400.0	501.0	COCl2	500	8	69.14	30.5	2.97	99.0
PUCL3-252	MSTPPB62C5	400.0	499.2	COCl2	500	10	69.78	29.8	2.89	96.8
PUCL3-253	MSTPPB62C5/MSTPPB63C5	400.0	500.6	COCl2	500	8 1/3	69.45	30.3	2.94	98.4
PUCL3-254	MSTPPB63C5	400.0	500.8	COCl2	500	8 1/6	69.20	29.4	2.86	95.5
PUCL3-255	MSTPPB63C5	386.6	478.8	COCl2	500	8 1/2	70.42	26.9	2.57	87.4

* U.S. GOVERNMENT PRINTING OFFICE 1993-0-774-036/87036

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