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BATTELLE-NORTHWEST FUEL FABRICATION EXPERIENCE
WITH HIGH-EXPOSURE PLUTONIUM

JANUARY 1965

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BATTELLE-NORTHWEST FUEL FABRICATION EXPERIENCE
WITH HIGH-EXPOSURE PLUTONIUM

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BATTELLE-NORTHWEST FUEL FABRICATION EXPERIENCE WITH HIGH-EXPOSURE PLUTONIUM

INTRODUCTION

Over 90% of the plutonium-containing fuel elements evaluated to date in the United States were designed and fabricated at Battelle-Northwest.⁽¹⁾ The major fuel element facility used was the Plutonium Fuels Laboratory (PFL),⁽²⁻⁴⁾ a large-scale laboratory with glove box enclosed equipment for plutonium fuels research and development studies under the Plutonium Recycle Program (PRP).⁽⁵⁾

Standard PFL glove boxes are constructed of stainless steel and have acrylic plastic or safety glass viewing panels.^(4, 6, 7) The boxes provide complete alpha containment but little protection against penetrating photon radiation; however, the glove box exteriors were designed to accommodate supplementary, easily removable, lead glass panels (to 1.27 cm or 0.5 in. overall thickness) for radiation shielding.

Plutonium, a toxic and pyrophoric metal, is detectable in extremely small quantities by modern instruments. It is an emitter of alpha particles and gamma, X, and neutron radiation. The radiation dose rate from plutonium is related to the amount present, the isotopic content, the time since it was purified or separated, and its physical or chemical state (i.e., metal oxide, fluoride, etc.). The plutonium isotopes 238, 239 (most common), 240, and 242 all emit alpha particles (energies to 5.49 Mev) and all undergo spontaneous fission.⁽⁸⁾ The major gamma ray energies (in Mev) for the same isotopes are: 0.0438, 0.052, 0.0453, and 0.045, respectively.^(8, 9) The major X-ray energies (in Mev) for the isotopes are: 0.017, 0.0205, 0.017, and 0.017, respectively. The gamma dose rate is relatively insensitive to the amount of plutonium present, but the neutron dose rate (not a problem to date in PFL) is directly related to the mass present. If materials of low atomic weight (e.g., beryllium, fluorine, chlorine, etc.) are involved with the plutonium, (alpha, neutron) reactions take place and the neutron dose rate can be markedly increased.

Most of the plutonium processed in the PFL since operation was initiated in February 1960 was of the low-exposure type: i. e., the plutonium contained mainly Pu^{239} with up to 8% Pu^{240} and lesser amounts of Pu^{241} and Pu^{242} . Much of the plutonium was used in support of the PRP for fuel element components for the Plutonium Recycle Test Reactor (PRTR). In the 18-month period following January 1962, 34 kg of high-exposure plutonium, mostly Pu^{239} , but with Pu^{240} contents to 30% plus smaller quantities of Pu^{241} , Pu^{242} , and Pu^{238} , was used in the fabrication of fuel elements. (4, 10, 11) In the period since July 1962, an additional 6 kg of high-exposure plutonium (22 to 27 wt% Pu^{240}), the first recycle material from the PRTR, was processed.

The use of high-exposure plutonium was initiated for two reasons: to further the research and development interests in the problems of handling and processing the material, and to gain fuel fabrication experience with plutonium of the recycle type which will be employed in the near future in commercial applications. (12)

This document describes the radiation studies performed and the equipment and process modifications required to permit safe, routine, efficient handling of high-exposure plutonium.

SUMMARY

Of the 237 kg of plutonium processed into more than 17,600 individual thermal reactor fuel components since initial PFL operation in February 1960, 40 kg was high-exposure plutonium, similar to the reactor grade types which will be commercially employed in the near future. Of the 40 kg, more than 8 kg was in the 26 to 30 wt% Pu^{240} range, 25 kg was in the 16 to 25 wt% Pu^{240} range, and the remainder in the 8 to 13 wt% Pu^{240} range. The high-exposure plutonium was essentially all processed during the period January 1962 through June 1963. Nearly half of the high-exposure plutonium went into $\text{UO}_2\text{-PuO}_2$ (typically 0.5 wt% PuO_2) fuel element components, while the remaining plutonium was used in the form of Al-Pu alloy in fuel element components.

The surface dose rate of bare high-exposure plutonium (18 wt% Pu^{240}) was 2.5 to 3.5 times the rate for bare low-exposure plutonium (5.5 wt% Pu^{240}),

when the time since separation for both metal specimens is on the order of 20 to 100 days.

The dose rates from glove boxes where $\text{UO}_2\text{-PuO}_2$ is handled are one-half to one-fourth that of glove boxes where Al-Pu alloy, made from similar high-exposure plutonium, is handled. This difference is attributed to the shielding provided by the uranium. In the case of Al-Pu alloys, where the plutonium contains 16 to 30 wt% Pu^{240} , the radiation levels are a factor of 5 to 10 higher than those from similar alloys made with low-exposure plutonium.

By effective use of lead glass panels, lead-impregnated gloves, portable shielding, and good work habits, the fuel fabrication with high-exposure plutonium was accomplished without excessive operator exposure and, in fact, the exposure was comparable to that obtained with unshielded low-exposure plutonium. Typical exposures for personnel handling low-exposure plutonium were 3.0 R/hr for the hands and 1.0 rem/yr for the body and for personnel handling high-exposure plutonium, 3.0 R/yr for the hands and 0.75 rem/yr for the body.*

The expenditure for the shielding devices, about \$25,000, would most likely have been made even if no high-exposure plutonium had been received. Incorporation of the shielding devices had been seriously considered on some glove boxes for low-exposure plutonium work because the radiation dose rate at the surface of the glove boxes was undesirably high (2-15 mR/hr). The experience at Battelle-Northwest with low-exposure plutonium indicates that a glove box background reading of 2-5 mR/hr will gradually build up from residual plutonium (usually in the form of oxide) left from normal glove box operations. The reading, taken at the acrylic panel surface, is reduced by a factor of about 10 when 0.64 cm thick lead glass shielding is added. The room background reading typically varies from 0.02 to 0.9 mR/hr, depending on the number of glove boxes, arrangements, and shielding.

* The occupational exposure limits are 75 rem/yr for the hands and 5 rem/yr for the whole body; however, administrative controls are based on 60 rem/yr for the hands and 4 rem/yr for the whole body.

From the experience in PFL and for the quantities and types of plutonium employed during 1962-1964, it is believed that there is little or no incremental operational cost involved in going from low- to high-exposure plutonium processing and no remote handling type operations are required.

PART 1

FUEL FABRICATION IN THE PLUTONIUM FUELS LABORATORY

A. Fuel Element Components and Plutonium Processed to Date

More than 17,600 individual thermal reactor fuel element components containing over 237 kg of plutonium have been processed (Table I) through the Plutonium Fuels Laboratory since initial operation in February 1960. Included in the above quantity was 39.9 kg of high-exposure plutonium (to 30% Pu²⁴⁰), of which 34 kg was handled in the period January 1962 to June 1963 and the remainder in the period July 1963 to January 1964. During the period February 1960 to January 1962, nearly all the fuel element components were prepared with Al-Pu alloy which contained low-exposure plutonium. From June 1963 to the present, the bulk of the fuel element components were of the UO₂-PuO₂ type and contained low-exposure plutonium. A detailed list of the high-exposure plutonium processed mainly during the period January 1962 to June 1963 is shown in Table II. Nearly half of the high-exposure plutonium went into UO₂-PuO₂ (typically 0.5 wt% PuO₂) fuel element components, while the remaining plutonium was used in the form of Al-Pu alloy in fuel element components.

TABLE I
PLUTONIUM PROCESSED AND FUEL ELEMENT COMPONENTS
PRODUCED IN THE PLUTONIUM FUELS LABORATORY

<u>Calendar Year</u>	<u>Total Plutonium Used, kg</u>	<u>Fuel Element Components Produced, Total Rods*</u>	<u>Nominal Pu²⁴⁰ Content, in Pu, %</u>	<u>Major Fuel Core Type</u>
1960	110.9	1382/~ 1300	≤ 8	Al-Pu Alloy
1961	27.4	1301/PRTR 1064, PRCF 19	≤ 8	Al-Pu Alloy
1962	29.6**	2235/PRTR 1083, PCTR 1000, PRCF 1000, PRCF 152	8-30	UO ₂ -PuO ₂
1963 (1st Half)	7.8**	551/PRTR 551	8-30	UO ₂ -PuO ₂
1963 (2nd Half)	11.8	608/PRTR 570	≤ 8***	UO ₂ -PuO ₂
1964	49.8	11,587/PRTR 627, EBWR 1674, PCTR 250, PRCF 201	≤ 8****	UO ₂ -PuO ₂
Totals	237.3****	17,664/PRTR 3895, EBWR 1674, PRCF 372, PCTR 1250		

* Total = total number of finished components produced/rods = rods (included in total) of a particular type. Fuel rods were made for the Plutonium Recycle Test Reactor (PRTR), (5, 13-18) the Experimental Boiling Water Reactor (EBWR), (19) the Physical Constants Test Reactor (PCTR), and the Plutonium Recycle Test Reactor Critical Facility (PRCF).

** Of the 29.6 + 7.8 = 37.4 kg of plutonium handled, about 34 kg was high-exposure type. See Table II for details. About 22 of the 34 kg went into UO₂-PuO₂ fuel.

*** Small quantities of plutonium (5.6 kg total) with Pu²⁴⁰ contents to 27% also processed, mainly into UO₂-PuO₂ fuel.

**** The 237.3 kg of plutonium, contained in finished components, was present in ~3000 kg of Al-Pu alloy. About 300 kg of plutonium was actually processed in the PFL.

TABLE II
HIGH-EXPOSURE PLUTONIUM
HANDLED IN THE PLUTONIUM FUELS LABORATORY

Plutonium, gm	Isotope Assay, wt%				
	Pu ²³⁹	Pu ²⁴⁰	Pu ²⁴¹	Pu ²⁴²	(Pu ²³⁸)
416.26	90.65	8.41	0.90	0.044	(0.011)
408.19	87.96	10.69	1.27	0.074	(0.013)
453.26	87.16	11.37	1.39	0.076	(0.013)
451.05	86.69	11.57	1.44	0.096	(0.013)
420.57	86.79	11.66	1.46	0.086	(0.013)
466.56	85.69	12.62	1.60	0.092	(0.014)
463.73	85.16	13.08	1.67	0.094	(0.014)
611.49	84.80	13.42	1.68	0.100	(0.014)
1524.63	84.48	13.61	1.80	0.109	(0.014)
1705.67	83.14	14.75	1.96	0.142	(0.014)
1855.24	81.53	16.04	2.24	0.181	(0.014)
1791.13	80.99	16.47	2.34	0.190	(0.015)
590.33	80.92	16.54	2.34	0.189	(0.014)
1501.33	80.47	16.94	2.39	0.201	(0.015)
523.45	79.90	17.27	2.59	0.235	
1426.51	79.99	17.29	2.51	0.208	(0.015)
1343.88	79.69	17.44	2.62	0.246	
1264.36	79.22	17.77	2.74	0.266	
1241.13	78.97	17.96	2.79	0.273	
1366.90	78.97	17.98	2.78	0.272	
1275.62	78.80	18.10	2.82	0.277	
1565.71	78.32	18.51	2.87	0.297	
597.36	76.54	19.93	3.19	0.335	(0.016)
1669.22	76.30	20.07	3.28	0.345	
1271.70	76.42	20.16	3.08	0.328	
1792.83	75.79	20.47	3.38	0.359	
1757.38	75.87	20.60	3.20	0.329	
570.40	75.22	20.99	3.40	0.395	(0.018)
1520.00**	73.04	22.49	3.81	0.66	
239.60**	~77.5	~22.5			
223.60**	~87.0	~23.0			
523.21	69.14	25.96	4.09	0.804	(0.040)
654.27	68.88	26.20	4.10	0.810	(0.038)
620.10**	66.87	27.08	5.06	0.99	
3076.90**	66.53	27.33	5.15	0.99	
623.38	65.67	27.65	5.49	1.18	(0.043)
413.41	64.60	28.56	5.67	1.17	(0.040)
632.88	64.40	29.13	5.34	1.14	(0.040)
631.56	64.07	29.29	5.47	1.17	(0.046)
588.93	62.92	29.91	5.93	1.24	(0.042)
484.45	62.69	29.99	6.00	1.17	(0.040)
187.89	<86.87	13.13			
502.05	<83.96	16.04			

41,248.32

* Approximately 39.6 kg of the plutonium was processed into fuel element components. The remaining 1.6 kg of unused material is in the 20% Pu²⁴⁰ class.

** First recycle plutonium from PRTR.

B. Fuel Element Fabrication Capabilities

Plutonium fuels of various types (Figure 1) were fabricated since 1955 by one or more techniques.⁽¹⁹⁻²⁶⁾ Metallic fuel elements were prepared by casting and machining, injection casting, extrusion, coextrusion cladding, and roll cladding. Ceramic fuel elements were made by die compaction and sintering, vibrational compaction, and swage compaction. UO_2 - PuO_2 fuel materials were densified by high-energy-rate impactation (Figure 2) for the first time in December 1962.⁽²⁷⁾

Fuel element lengths to 366 cm (12 ft) and diameters to 15 cm (6 in.) can be processed in the PFL. Equipment is available to produce one 19-rod cluster (Figure 3) of the Plutonium Recycle Test Reactor (PRTR) type per day (single 8-hr shift) using either metallic or ceramic fuels.⁽¹³⁻¹⁸⁾ The fuel cores in the rods are nominally 1.27 cm (0.5 in.) in diameter and 224 cm (88 in.) long. Typical fuels were Al-2.0 wt% Ni-1.8 wt% Pu alloy and UO_2 (natural)-0.5, 0.75, and 1.0 wt% PuO_2 . During November-December 1964, fabrication of UO_2 (natural)-2.0 wt% PuO_2 fuel rods of the same diameter but shorter length was initiated. The 19-rod clusters are like the one in Figure 3, except the elements are now 76 cm (30 in.) shorter. The plutonium content per rod was about 12 gm in the Al-Ni-Pu alloy case, 12-25 gm in the UO_2 -0.5, 0.75, and 1.0 wt% PuO_2 cases, and about 33 gm in the case of the shorter rods with UO_2 -2.0 wt% PuO_2 fuel.

Starting in 1964, fuel rods were fabricated for the joint program of Battelle-Northwest and Argonne National Laboratory to study the burnup characteristics of plutonium-enriched oxide fuel elements in the Experimental Boiling Water Reactor (EBWR).^(12, 19, 27) The vibrationally-compacted EBWR rods each contain 830 gm of impacted UO_2 (depleted)-1.5 wt% PuO_2 fuel (the plutonium has 8 wt% Pu^{240}), have low-nickel type Zircaloy-2 cladding, are 1.1 cm in diameter and 148 cm long, and currently number over 2000.

Work was also initiated in 1964 on 160 Saxton Reactor fuel rods, an important job in that satisfactory irradiation behavior of the rods should encourage greater commercial use of plutonium in thermal power reactors.⁽¹²⁾ The vibrationally-compacted Saxton Reactor rods each contain 530 gm

FIGURE 1

Variety of Fuel Elements and Components
Fabricated at Battelle-Northwest with Plutonium-Containing Cores.
The 19-Rod Cluster at Top is 230 cm (90 in.) Long.

FIGURE 2

High-Energy Impactation Unit. Capacity of the Unit is 145,000 ft-lb.
Neg. 0631046-1, 0641271-3

FIGURE 3

Typical $\text{UO}_2\text{-PuO}_2$ or Al-Ni-Pu Alloy Fuel Element Used in
the Plutonium Recycle Test Reactor.

Rods are 1.43 cm (0.563 in.) in Diameter and 230 cm (90 in.) Long.

The Element is 8.25 cm (3.25 in.) in Diameter.

Neg. 0640543

of impacted UO_2 (natural)-6.6 wt% PuO_2 fuel (plutonium has 8.6 wt% Pu^{240}),
have Zircaloy-4 cladding, and are 1.0 cm in diameter and 99 cm long.

PART 2

RADIATION PROBLEMS ASSOCIATED WITH PLUTONIUM FUELS

There has been considerable effort directed toward the solution of the radiological problems encountered while working with plutonium. Not only is one confronted with dose rates that may require personnel shielding, but also some mechanism is required to assure that no plutonium is present in the atmosphere at work locations. These two conditions lead to two types of radiological control, i.e., the control of internal deposition, and control of external radiations (see Table III).

A. Exposure by Internal Plutonium Deposition

The principal means of internal deposition is by inhalation of plutonium contaminated air and by entry through minor wounds. In the case of the latter, entry is almost exclusively limited to the hands. Under no conditions is there any planned exposure to airborne contaminants and, therefore, all depositions are the result of unplanned occurrences.

The amount of plutonium present as airborne contaminants is determined by a continuous air sampling program. If at any time the amount of airborne activity exceeds the maximum permissible concentration in air (MPC_A) which is $2 \times 10^{-12} \mu\text{c}/\text{cm}^3$, respiratory protection is required. The effectiveness of the air sampling program is verified by a bioassay program. There has never been any significant plutonium deposition in PFL workers. Minimum detection is about 1% of the maximum permissible body burden ($0.04 \mu\text{c}$) for Pu^{239} when quarterly bioassay samples are collected.

Plutonium depositions through external wounds and scratches are limited primarily by good housekeeping practices and the cultivation of good working habits by the personnel. Sharp objects and tools are eliminated wherever possible in all hoods and glove boxes.

TABLE III
GAMMA AND X RADIATIONS FROM SELECTED ISOTOPES OF THE ACTINIDES

Isotope	Half-Life	Gamma Ray		Energy, kev	Yield, %	e/ λ Ratio*
		Energy, kev	Yield, %			
Pu ²³⁸	86.4 yr	43.8	0.038	17	11	740
		99	8×10^{-3}	17	0.035	11
		150	1×10^{-3}			
		203	4×10^{-6}			
		760	5×10^{-5}			
		810				
		875				
Pu ²³⁹	2.436×10^4 yr	37	1.6×10^{-3}	13.6	1.2	
		52	6.4×10^{-3}	17.4	1.4	
		120	9.6×10^{-4}	20.5	3.3	
		207	3.2×10^{-4}			
		340	4.9×10^{-4}			
		380	9.6×10^{-4}			
		420	6.4×10^{-4}			
Pu ²⁴⁰	6.58×10^3 yr	45.3	0.009	17	4	
Pu ²⁴¹	13.0 yr	145	2.8×10^{-4}			
Pu ²⁴²	3.79×10^5 yr		0.01	17	4.1	
Pu ²⁴³	4.98 hr	12.2				
U ²³⁷	6.75 day	26.4				
		33.2				
		43.5				
		59.6	36			
		65	2.3			
		114				
		165	3.6	17	0.19	0.13
		208	24	17	22	2.3
		267	0.9	17	0.22	0.63
		332	1.4	17	0.22	0.04
		335	0.2	17	0.016	0.2
		368				
Am ²⁴¹	4.58×10^2 yr	371				
		26.4	2.5	17	6.5	7
		33.2	0.15	17	14.3	240
		43.5	0.073			
		59.6	36	17	21.1	1.5
		99	0.023			
		103	0.019			
		113	2.5×10^{-3}			
		130	3.8×10^{-3}			
		159	2.9×10^{-4}			
		210	5.8×10^{-4}			
		270	7.6×10^{-5}			
		330	2.5×10^{-4}			
		370	1.5×10^{-4}			

* Electron to gamma photon emission ratio

B. External Exposure

External exposure occurs to both the whole body and the hands. The magnitude of exposure is related to several factors. These are:

- The amount of material present
- The isotopic content
- The time since it was purified or separated
- Its physical or chemical state, i. e., metal, oxide, etc.

Dose rates associated with handling of large quantities of plutonium have been calculated and measured. The following equation⁽⁹⁾ can be used to calculate the surface dose rate due to X and gamma rays from an infinitely thick slab of plutonium metal. Plutonium is considered infinitely thick to its own energy spectrum, if, when made thicker, the dose rate remains constant.

$$D_s = 960 P^{238} + 0.98 P^{239} + 4.7 P^{240} + \{0.19 t + 17 [1 - e^{-0.102t}] \} P^{241} + 0.29 P^{242}, \quad (1)$$

where D_s = surface dose rate, rad/hr

P^i = percent of the i^{th} isotope of plutonium present

t = time since separation, days.

Close examination of Equation (1) reveals the importance of isotopic composition and time since separation. This equation was solved for three different isotopic compositions and for two different times since separation and the results are presented in Table IV.

Surface dose rate measurements were made on plutonium metal, PuO_2 and UO_2 - PuO_2 mixtures. The results of some of these measurements are summarized in Table V, along with overall attenuation factors of 0.030 cm (0.012 in.) plastic, 0.043 cm (0.017 in.) thick neoprene and 0.076 cm (0.030 in.) thick, lead-loaded, neoprene gloves.

TABLE IV
SURFACE DOSE RATES OF PLUTONIUM METAL
AS A FUNCTION OF COMPOSITION AND TIME SINCE SEPARATION

<u>Composition, wt%</u>	<u>Time Since Separation, days</u>	<u>Surface Dose Rate, rad/hr</u>
Pu ²³⁹ - 94	20	1.3
Pu ²⁴⁰ - 5.5		
Pu ²⁴¹ - 0.5		
Pu ²³⁹ - 94	100	1.4
Pu ²⁴⁰ - 5.5		
Pu ²⁴¹ - 0.5		
Pu ²³⁹ - 90.451	20	1.5
Pu ²⁴⁰ - 8.578		
Pu ²⁴¹ - 0.923		
Pu ²⁴² - 0.048		
Pu ²³⁹ - 90.451	100	1.6
Pu ²⁴⁰ - 8.578		
Pu ²⁴¹ - 0.923		
Pu ²⁴² - 0.048		
Pu ²³⁹ - 71	20	3.2
Pu ²⁴⁰ - 18		
Pu ²⁴¹ - 9		
Pu ²⁴² - 2		
Pu ²³⁹ - 71	100	4.8
Pu ²⁴⁰ - 18		
Pu ²⁴¹ - 9		
Pu ²⁴² - 2		

TABLE V
MEASURED SURFACE DOSE RATES AND ATTENUATION FACTORS

Material	Isotopic Composition	Plutonium, wt%	Dose Rate, rad/hr	Attenuation Factors*		
				0.030 cm (0.012 in.) Plastic	0.043 cm (0.017 in.) Neoprene	0.076 cm (0.030 in.) Lead-Loaded Neoprene
PuO ₂ -UO ₂	Pu ²³⁹ - Unknown,	1	0.24	1.56		
PuO ₂ -UO ₂	Pu ²⁴⁰ - 8%,	3	0.30	1.50		
PuO ₂ -UO ₂	Pu ²⁴¹ - Unknown,	5	0.37	1.47		
PuO ₂ -UO ₂	For all four cases	10	0.45	1.40		
PuO ₂ -UO ₂	Pu ²³⁹ - Unknown,	3	0.24	1.50		
	Pu ²⁴⁰ - 2.2%,					
PuO ₂ -UO ₂	Pu ²³⁹ - Unknown,	3	0.30	1.50		
	Pu ²⁴⁰ - 8%,					
	Pu ²⁴¹ - Unknown					
PuO ₂ -UO ₂	Pu ²³⁹ - Unknown,	3	0.37	1.50		
	Pu ²⁴⁰ - 16.5%,					
	Pu ²⁴¹ - Unknown					
PuO ₂ -UO ₂	Pu ²³⁹ - Unknown,	3	0.47	1.50		
	Pu ²⁴⁰ - 27.3%,					
	Pu ²⁴¹ - Unknown					
PuO ₂ **	Pu ²³⁹ - 90.451,	100	1.57	1.31	2.2	5.9
	Pu ²⁴⁰ - 8.578,					
	Pu ²⁴¹ - 0.923,					
	Pu ²⁴² - 0.048					
Pu**	Pu ²³⁹ - 90.451,	100	1.69	1.30	2.6	5.8
	Pu ²⁴⁰ - 8.578,					
	Pu ²⁴¹ - 0.923,					
	Pu ²⁴² - 0.048					

* Attenuation Factor = (surface dose rate of bare source)/(surface dose rate of material-covered source).

** Both the PuO₂ and Pu were separated about July 15, 1964.

Unfortunately, the history of only one source was well enough known to make any direct comparison between experiment and theory. Measurements were made on the plutonium metal in Table V on October 28, 1964. From Equation (1), the calculated dose rate is 1.6 rad/hr as compared to 1.7 rad/hr measured. This is in excellent agreement.

It was found that the effectiveness of common shielding material used in hoods or glove boxes, i. e., plastic and gloves, remains about the same for any composition when isotope Pu^{239} dominates; however, it will change if the amount of isotopes Pu^{240} through Pu^{242} become dominant. Unfortunately, energy spectrum measurements are not readily available at any composition other than for ordinary production material nor are any studies of spectral changes with time currently available.

C. Hand Exposure

The type of hood glove used is a significant factor in determining hand exposure. The most common glove in use at the PFL is the 0.076 cm (0.030 in.) thick neoprene type. However, 1.52 cm (0.060 in.) thick lead-loaded, neoprene gloves have been used occasionally with limited success. If hand exposures were to approach the annual control guide, currently 60 rem/yr, the thicker gloves would be used.

D. Whole Body Exposure

The PFL has no extraordinary shielding material in place to control whole body exposure. When similar high-exposure plutonium is employed in the fuels, the dose rates from glove boxes where $\text{UO}_2\text{-PuO}_2$ is handled are one-half to one-fourth that of glove boxes where Al-Pu alloy is handled. This difference is attributed to the shielding provided by uranium. In the case of Al-Pu alloys where the plutonium contains 16 to 30 wt% Pu^{240} , the radiation levels (Figure 4) are a factor of 5 to 10 higher than those obtained with similar alloys, but made with low-exposure plutonium. While it is true that surface dose rates are quite high, the dose rates which exist at work locations can be controlled by the use of plastic, iron, or lead in the construction of the enclosure. In fact, most of the glove boxes are constructed with plastic panels and dose rates at the surface of the glove boxes are in the

2 to 15 mR/hr range. Unshielded fuel element storage areas for low-exposure plutonium may have dose rates as high as 20 mR/hr. Dose rates in unshielded plutonium storage vaults may reach 50 mR/hr,⁽²⁸⁾ primarily because of the quantity of material present (to 100 kg). If gamma or X-ray dose rates at work locations ever become a problem, 0.64 to 1.27 cm (1/4 to 1/2 in.) of lead glass would probably be adequate to reduce the dose rate to acceptable levels.

FIGURE 4

Whole Body Dose Rates, Measured Under Actual Working Conditions with a CP, at the Glove Box Working Face Through 0.76 (0.030 in.) Thick Neoprene Gloves (the average whole body dose rate experienced during these operations was less than 2 mrem/hr due to the attenuation provided by the glove box materials and lead-glass panels.) [Al-2.6 wt% Pu alloy fuel rods within typical fabrication glove-boxes; 16.5% Pu²⁴⁰; 2.33% Pu²⁴¹; rods 2.54 cm (100 in.) long, 1.27 cm (0.5 in.) diam]

E. Exposure Due to Neutrons

Dose rates due to neutrons are dependent on the isotopic composition of plutonium and the mass of material present. This is particularly true with very high exposure plutonium fuels when the amounts of Pu^{238} , Pu^{240} , and Pu^{242} are appreciable. Exposure due to neutrons has been an infrequent problem in the PFL. Experience to date has shown that the annual whole body exposure from neutrons has averaged approximately 1/10 the annual exposure from X- and gamma rays.⁽²⁸⁾ However, it might be necessary to install neutron shielding if appreciable quantities of high exposure plutonium were to be processed, especially if on a routine basis. If a process existed within a fuel fabrication facility where plutonium existed as PuF_4 or some combination involving materials of low atomic weight, one could expect a large increase in neutron dose rates because of the alpha-neutron reactions. It would then, in all probability, become necessary to use neutron shielding such as water, paraffin, or other material that could be conveniently used in the form of a wall.

Equation (2) is an equation which can be used to calculate the surface dose rate, due to neutrons, of a spherical mass of plutonium. The equation was first presented by Roesch⁽²⁹⁾ and subsequently modified by Faust.⁽⁹⁾

$$D_s = M^{1/3} [0.927 P^{238} + 8.24 \times 10^{-6} P^{239} + 0.281 P^{240} + 0.467 P^{242}] \quad (2)$$

where D_s = dose rate in rem/hr

M = mass of plutonium in grams

P^i = percent of the i^{th} isotope of plutonium present

F. Actual Personnel Exposures

Table VI summarizes the whole body and hand exposures for a group of 15 persons employed within the PFL. The year 1962, while included in the overall summary, is listed separately for comparison purposes for it was in that year that the bulk of the experience was gained in processing high-exposure plutonium. The differences between the exposures in the time periods are insignificant.

The maximum allowable exposure values at Battelle-Northwest are 75 rem/yr for the hands and 5 rem/yr for the whole body; however, administrative controls limit the maximum exposures to 60 rem/yr for the hands and 4 rem/yr for the body.

TABLE VI
AVERAGE PERSONNEL EXPOSURE

<u>Year</u>	<u>Hand</u>	<u>Whole Body Penetrating Dose</u>	<u>Type of Plutonium Handled</u>
1960-1963	3.0 R/yr	1 rem/yr	Mainly low-exposure
1962	3.0 R/yr	0.75 rem/yr	Primarily high-exposure

PART 3

PLUTONIUM FUELS LABORATORY (308 BUILDING)

A. Background Information

Plutonium fuels have been fabricated at Battelle-Northwest since 1955. The initial fabrication efforts were conducted within an existing plutonium metallurgical laboratory⁽²⁾ that did not contain adequate space for the large-scale fuel fabrication equipment required to meet the PRP needs. Construction of a fuel fabrication laboratory, the PFL, was started in mid-1958 and completed in late 1959. This facility, located in the PRTR complex about 6 miles north of Richland, Washington, was initially known as the Plutonium Fuels Pilot Plant (PFPP).

The PFL is a versatile laboratory with glove box enclosed equipment for plutonium fuels research, development of new fabrication processes, and fabrication of experimental reactor fuels containing plutonium.^(2, 4) Commercially available equipment of standard or near-standard type is used to the greatest extent possible for the processes in the PFL. Metalworking equipment has required no more than minor modifications for glove box use, and this is also generally true of oxide processing equipment. The equipment is installed within glove boxes or incorporated into the glove box frame.

The basic design philosophy of the PFL is that of containment and directional ventilation control.⁽²⁾ Primary containment of plutonium in the PFL is within the glove boxes, secondary containment within individual laboratories, and ultimate containment within the building proper. Limitation and control of any contamination spread is effectively accomplished by this design.

B. Description of the Building

The PFL (Figures 5, 6, and 7) consists of a two story process area having a gross floor area of 1900 m^2 ($20,000 \text{ ft}^2$) on each floor, and an attached, one story service and office wing of 650 m^2 (7000 ft^2). Approximately 740 m^2 (8000 ft^2) are required on the second floor for ventilation equipment and maintenance shops. The process portion of the building is of reinforced concrete and masonry construction. Reinforced concrete columns support the slab type second floor and structural steel columns support the steel-framed roof. Nonload bearing wall panels are constructed of cinder blocks and all block surfaces exposed to the building interior are plastered to provide a smooth, continuous surface which is easily decontaminated. A systematic arrangement of ventilation supply and exhaust ducts allows a good deal of freedom in room and equipment arrangement. Electrical switchgear and service piping headers are located in galleries that are isolated from the zones with potential contamination.

C. Building Ventilation

Contamination spread in the building is minimized by the directional ventilation system (Figure 8) which maintains the negative building pressure and provides up to 15 air changes per hour. The high-volume, low-velocity system is designed to be very stable without splitters and to maintain building balance with a changing total air flow.

Air balance is maintained to direct the flow from zones of low contamination potential to zones of highest potential. The air in the corridors of the process area is at less than atmospheric pressure (differential is 0.13 cm or 0.05 in. of water). Air in the process area laboratories is also at less than atmospheric pressure (differential is 0.25 cm or 0.10 in. of water).

FIGURE 5

Section Views of the Plutonium Fuels Laboratory.
Two-Story Portion is 43 m (142 ft) Square.

FIGURE 6

Cut-Away View of the First Floor of the Plutonium Fuels Laboratory.
Neg. 02125-1, 38854-1

FIGURE 7

Cut-Away View of the Second Floor of the Plutonium Fuels Laboratory.

FIGURE 8

Schematic Ventilation Flow Diagram for Plutonium Fuels Laboratory.
Neg. 0640157-2, 29899-1

Washed, filtered air is supplied through ceiling diffusers and exhausted through high efficiency filters located at floor level. The downward sweep of air has been demonstrated to be effective in confining accidental contamination spills to a small area. Exhaust air from the laboratory is filtered a minimum of two times through high efficiency filters. All exhaust air is monitored for radioactive contamination between filters and before release to the atmosphere. Automatic back-draft dampers prevent the escape of contaminated air through supply ducts not equipped with high efficiency filters, but enable the building to breathe through filtered exhaust lines in case of complete failure of ventilation equipment.

D. Glove Boxes

Typical PFL glove box construction is shown in Figure 9. The boxes are constructed of stainless steel with acrylic plastic panels. Plastic glove ports are ordinarily cemented to the panels. Aluminum or lead tape is used on inside joints (in some cases outside joints too) of glove boxes to assist in maintaining clean box interiors, to reduce the background radiation level

FIGURE 9

Typical Plutonium Fuels Laboratory Glove Box Construction
Neg. 02125-2

stemming from the joint crevices, and to keep the rubber gasket in the acrylic panel-box joint as contamination free as possible. Glove boxes are essentially leak free.

Boxes are designed for specific needs, and design and construction costs are minimized by using well-developed standard details and specifications. Individual boxes as long as 14 m (46 ft) are used. Modules can also be joined to form a continuous line of any desired length or to modify existing boxes. A flanged section module was added to the glove box in Figure 10 at a later date to increase the box length as shown in Figure 11.

FIGURE 10

Glove Box Enclosed Swage. Covers, in Place Over Most Upper Glove Ports, Aid in Reducing Glove Box In-Leakage.

Neg. 32024-4

FIGURE 11

Flanged Section of Glove Box Enclosed Swage.
This Section (Center of Photograph) Permits Addition
of Glove Box Modules to Change Box Length.

Neg. 062101-6

Standard PFL glove boxes provide complete alpha containment, but little protection against penetrating photon radiation; however, the glove box exteriors were designed to provide the necessary features to accommodate supplementary, easily removable lead glass panels (to 1.27 cm or 0.5 in. overall thickness) for photon shielding.

E. Glove Box Ventilation

Dry air or inert atmospheres are used in the glove boxes and selection is made by manual valve operation. Variable purge rates to 71 1/sec ($150 \text{ ft}^3/\text{min}$) may be obtained. Box atmospheres are automatically controlled at a negative pressure by self-sensing supply and exhaust valves. An exhaust rate of about 212 1/sec ($450 \text{ ft}^3/\text{min}$) is automatically attainable

under emergency conditions. Exhaust air from the glove boxes is filtered at least three times through high-efficiency filters before being released to the atmosphere. Air samples are taken between filters and monitored for plutonium contamination.

Glove box atmospheres are at a lower pressure (differential is 2.5 cm or 1 in. of water) than the process area laboratories. The pressure differential provides a sufficient margin of safety so that the ventilation system can react to very sudden volume changes and prevent glove box pressurization, a condition where the pressure inside the box is greater than the pressure outside. A pressure differential greater than 2.5 cm (1 in.) of water is usually avoided since it causes undue operator fatigue by making work with gloves difficult.

PART 4

MODIFICATIONS TO ACCOMMODATE HIGH-EXPOSURE PLUTONIUM

There were two major reasons in 1960-1961 for seriously considering the modification of certain pieces of equipment and process steps in the PFL. One reason was the anticipated arrival of high-exposure plutonium. The second was the undesirably high radiation level (2-15 mR/hr) of specific glove boxes, due to the amount of material being handled and/or the background (2-5 mR/hr) from the glove boxes after extended usage with low-exposure plutonium. The glove boxes with notable background radiation levels were primarily those in which plutonium was present as oxide, either introduced as oxide or by the oxidation of metal, and rather uniformly spread in a very thin layer throughout the glove box.

A. Lead Glass

For the foregoing reasons, 0.64 cm (0.25 in.) thick, X-ray type, lead glass panels were installed on the exterior of the acrylic plastic panels (Figures 12 and 13). One lead glass panel is equivalent to about 1.6 cm (0.06 in.) of lead, based on 105-keV photon radiation measurements. The lead glass panels were designed for easy installation and removal by means of clips.

B. Lead-Impregnated Gloves

Lead-impregnated gloves were employed to reduce dose rates. Two thicknesses of lead-impregnated gloves were initially considered. One was 0.76 mm (0.030 in.) and the second, 1.52 mm (0.060 in.). The equivalent lead thickness, based on 0.087 Mev-Cd¹⁰⁹ measurements were about 0.1 mm (0.004 in.) and 0.36 mm (0.014 in.), respectively. Only the thinner type were actually used and then at only specific stations. The 1.52 mm (0.060 in.) thick lead-impregnated gloves were quite difficult to use since the manual dexterity of an operator is seriously impaired—it is an effort just to bring one's fingers together—and technicians tire easily because of the inherent weight.

The nonleaded gloves normally used for low-exposure plutonium work are nominally 0.76 mm (0.030 in.) thick, though thinner (nominally 0.38 mm or 0.015 in.) gloves are used for delicate jobs.

The approximate cost of the gloves is shown below:

<u>Type</u>	<u>Total Glove Thickness</u>		<u>Cost, \$/pair</u>
	<u>mm</u>	<u>in.</u>	
Lead-Impregnated	0.76	0.030	23-27
Lead-Impregnated	1.52	0.060	~80
Nonleaded	0.76	0.030	12-20
Nonleaded	0.38	0.015	10-20

The average lifetime of the lead-impregnated gloves during PFL use is about 1 yr or about the same as that of standard nonleaded gloves. Some of the newer gloves made from tougher compounds should last longer and also be more ozone resistant.

C. Other Modifications

As a general policy, a conscientious effort was made to shield materials being stored (or not being worked on) and to reduce actual handling times by personnel. Lead-impregnated rubber sheets (or aprons) were used effectively as portable shielding both inside and outside the glove boxes. Small lead-lined boxes similar to the type used for storing X-ray film provided shielded storage units which could be easily rolled from room to room.

When Al-Pu alloys containing high-exposure plutonium were inserted or removed from a glove box, operating personnel wore lead-impregnated aprons if radiation levels were undesirably high. In addition, tongs helped to reduce exposures (Figures 14 and 15).

FIGURE 14

Container of PuO_2 Being Positioned with Tongs for Heat Treatment.
Neg. 32024-23

FIGURE 15

Burning of Plutonium Metal to Obtain PuO_2 .

Neg. 32024-16

D. Total Expenditures

For the Al-Pu alloy fuel fabrication process,^(13, 14) the following glove boxes were equipped with lead-type panels and gloves: (a) casting facilities (Figures 13 and 16-18),^(7, 21) (b) extrusion press (Figure 19), (c) rod straightener (Figure 12), (d) large portable fuel rod storage and transfer unit (Figure 20), (e) draw bench (Figure 21), (f) cut-off saw (Figure 12), and (g) lathe (Figure 22). The process flow diagrams are given in the Appendix.

FIGURE 16

Glove Box Used for Melting and Casting in Air or Inert Gas,
Burning of Plutonium to Form PuO_2 , and Treatment of PuO_2 .

FIGURE 17

Melting and Casting Glove Box (in Figure 16)
After Lead-Glass Panels Installed.

FIGURE 18

Tilt-Pour Furnace in Operation
Al-Pu Alloy Being Poured
into Graphite Mold. Furnace
Tilting Manually Controlled
Outside Glove Box. Filled Mold
Moved with Tongs.

FIGURE 19

Extrusion Press Before Lead-Glass Panels Installed.
Press Capacity is 254 tonnes (280 tons).

FIGURE 20

Portable Storage and Transfer Unit
Before Double Lead-Glass Panels Installed.
Pump and Tank Maintain Negative Pressure
in Unit When it is Detached from Other Glove Boxes.

FIGURE 21

Glove Box Enclosed Draw Bench.
Technician Working in Portable Storage
and Transfer Unit Which is Attached
by Plastic Bag to Draw Bench Box.

FIGURE 22

Glove Box Enclosed Lathe.

The Al-7.5 wt% Pu Alloy Billet Contains Plutonium with 9 wt% Pu²⁴⁰.

Physically mixed UO₂-PuO₂ fuel (Figure 23) has been used in the PFL, and for this process^(15, 16) all glove boxes involved in the steps through blending (of the UO₂ and PuO₂) were modified. The nominal fuel composition during the 18-month period of high-exposure plutonium handling was UO₂ (natural)-0.5 wt% PuO₂ with the plutonium having an average Pu²⁴⁰ content in the 16 wt% range.

The 0.64 cm (0.25 in.) thick lead glass panels, lead-impregnated gloves, and internal shields were installed at the PuO₂ preparation glove
Neg. 0630686-1

boxes (Figures 13-17), which were previously used as the stations for Al-Pu casting, at the mixing station (Figure 24), and the blending station (Figure 25).

FIGURE 23

Incremental Loading of PRTR Fuel Rod with UO_2 -0.5 wt% PuO_2 Fuel with Vacuum Dispensers. The Funnel is Attached to Upper End of Rod. The Plutonium Contained about 16 wt% Pu^{240} . Coarse and Medium UO_2 Particles are in the Larger Can; the Blended Fine Particles of UO_2 and PuO_2 are in the Other.

Neg. 32024-6

FIGURE 24

Mixing of Prew weighed Quantities of PuO_2 (light colored powder) and UO_2 . Alternate Layers of UO_2 are "Salted" Uniformly with PuO_2 and Manually Stirred as the First Mixing Step. The Mixture is then Further Processed in a Twin-Shell Blender.

FIGURE 25

Glove Box with Twin-Shell Blending Equipment for UO_2 - PuO_2 .
Neg. 32024-5, 38581-10-CN

The X-ray type lead glass panels used are 0.64 cm (0.25 in.) thick and typically cost* about \$216/m² or \$20/ft². Single panels were used on most modified PFL glove boxes but double panels, installed in the same fashion used for single panels, were used on two glove boxes (portable storage and transfer unit and rod straightener). Using a stack of more than two of the above thin panels creates a visibility problem because of the numerous interfaces. If more than 1.27 cm (1/2 in.) of lead glass is needed, the PFL glove box design could be slightly altered (done in about 1962) to make provisions for using cast, high-density, lead glass panels. Such panels, 2.5 cm (1 in.) thick, cost about \$1100/m² or \$100/ft².

The 0.64 cm (0.25 in.) lead glass panels used in the PFL have metal-bound edges and have performed well for 5 yr. The few replacements made were for cases in which the glass was inadvertently broken by mishandling when off the glove boxes. It is probable that glass panels like those used in PFL should have a life expectancy of at least 10 yr.

The total expenditure for all shielding devices (glass, gloves, sheets, aprons, storage boxes, etc.) was about \$25,000 and of this only \$1600 was solely due to high-exposure plutonium requirements. A total of about 82 m² (880 ft²) of 0.64 cm (0.25 in.) thick lead glass has been used and of this amount only 7.4 m² (80 ft²) can be strictly attributed to high-exposure plutonium needs. The lead glass panels were installed on glove boxes which had a total of about 300 glove ports. Approximately 30 pairs of the 0.076 cm (0.030 in.) lead-impregnated gauntlet gloves were obtained for use on these glove boxes; however, only half of these gloves were used.

* The cost is for an average panel and includes procurement of the glass and fabrication (at Battelle-Northwest) of metal edging and cut-outs for glove ports.

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APPENDIX

<u>Process Flow Diagrams</u>	<u>Figure Number</u>
Aluminum-Plutonium Fuel Element Fabrication Process	A-1
Vibratory Compaction Fabrication Process	A-2
Swage Compaction Fabrication Process	A-3
Fabrication Process for Vibrationally-Compacted and Swage-Compacted UO_2 - PuO_2 Fuel Elements for PRTR	A-4

FIGURE A-1

Mark I-G

Aluminum-Plutonium Alloy Fuel Element Fabrication Process, (13, 14)
Neg. 0650305-2

FIGURE A-2

Mark I-L

UO₂-PuO₂ Vibratory Compaction Fabrication Process.^(24, 26)

This Process Used in the PFL up to November 1963.

FIGURE A-3

Mark I-M

UO₂-PuO₂ Swage Compaction Fabrication Process^(24, 25)

This Process Used in the PFL up to November 1963.

Neg. 0650305-3

FIGURE A-4

Fabrication Processes for Vibrationally-Compacted (Mark I-L) and Swage-Compacted (Mark I-M) $\text{UO}_2 - \text{PuO}_2$ Fuel Elements for PRTR. These Processes in Use in the PFL Since November 1963. (15, 16, 30-32)

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