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PLUTONIUM HANDLING AND CONTROL PRACTICES
AT PACIFIC NORTHWEST LABORATORY

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OCTOBER, 1966

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RICHLAND, WASHINGTON

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BATTELLE MEMORIAL INSTITUTE

for the

UNITED STATES ATOMIC ENERGY COMMISSION UNDER CONTRACT AT(45-1)-1830

PRINTED BY/FOR THE U. S. ATOMIC ENERGY COMMISSION

3 3679 00060 2765

BNWL-287

UC-41, Health and Safety

PLUTONIUM HANDLING AND CONTROL PRACTICES
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By

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September 1966

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RICHLAND, WASHINGTON

- * A paper presented at the Plutonium Symposium, sponsored by the American Nuclear Society, Atomic Industrial Forum, and the Edison Electric Institute in cooperation with the Atomic Energy Commission, at Washington, D. C., on March 1 and 2, 1966.

Printed in U.S.A. Price \$1.00. Available from the
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PLUTONIUM HANDLING AND CONTROL PRACTICES AT PACIFIC NORTHWEST LABORATORY

INTRODUCTION

One of two major facilities used for plutonium-containing fuels research and development at Battelle-Northwest is the Plutonium Fuels Laboratory (PFL). This facility was specifically designed and equipped for research and development studies involving multikilogram quantities of plutonium and its mixtures, compounds, and alloys. Since initial operation of the PFL in February, 1960, more than 325 kg of plutonium have been fabricated into more than 19,300 individual thermal reactor fuel element components. More than 18% of the plutonium processed has contained 10 to 30% Pu^{240} .

Most of the plutonium processed in the PFL has been fabricated into test fuel element components for the Plutonium Recycle Test Reactor (PRTR). A typical 19 rod cluster PRTR fuel element is shown in Figure 1. The fuel diameter of each rod is about 1.28 cm (0.505 in.), and the fuel length is either 148.6 cm (58.5 in.) or 228.6 cm (90 in.), depending upon the particular design. Fuel fabricated into PRTR rods includes Al-Ni-Pu alloy and PuO_2 in UO_2 , ThO_2 , ZrO_2 , and MgO . Methods developed for metal fuels fabrication include casting, rolling, drawing, machining, and extruding. Methods developed for oxide fuels fabrication include swage compaction, vibrational compaction, high-energy-rate impaction, and pellet pressing, sintering, and

grinding. All of the oxide fuel for PRTR components has contained 0.5 to 5.0 wt% PuO_2 .

FIGURE 1. Typical UO_2 - PuO_2 or Al-Ni-Pu Alloy Fuel Element Used in the Plutonium Recycle Test Reactor (Neg. 0640543-1)

SUMMARY

One of two major facilities used for plutonium fuels research and development studies at Battelle-Northwest is the Plutonium Fuels Laboratory (PFL). This facility was specifically designed and equipped for research and development studies involving multikilogram quantities of plutonium and its mixtures, compounds, and alloys.

As at other sites, the design and operation philosophy of the PFL is one of complete plutonium containment. Primary plutonium containment is provided by glove boxes, secondary containment is provided by individual laboratories, and tertiary containment is supplied by the building proper. Air samples, taken throughout the facility, are constantly being monitored for free contamination.

Personnel exposure caused by X, gamma, and neutron radiation has been primarily limited by inspiring good work habits and housekeeping practices in personnel (i.e., uncontrolled accumulations of plutonium are avoided, plutonium is confined as much as possible, exposure times are limited, and shielding equipment is used whenever necessary).

Rules for the prevention of an inadvertent criticality in the PFL are based upon the criteria that at least two control conditions must fail before criticality is imminent.

RADIATION PROBLEMS ASSOCIATED WITH PLUTONIUM

Plutonium is an emitter of alpha, X, gamma, and neutron radiation (Table I). It is a man-made toxic element, principally produced by the neutron irradiation of U^{238} . The

maximum permissible body burden for Pu^{239} is $0.04 \mu\text{c}$, and the maximum permissible concentration in air (MPC_A) is $2 \times 10^{-12} \mu\text{c}/\text{cm}^3$. For this reason, complete containment of plutonium is essential in any plutonium processing facility.

CONTROL OF INTERNAL DEPOSITION

Nearly all plutonium work performed in the PFL is conducted in glove boxes which provide primary containment. A typical PFL glove box is shown in Figure 2. These boxes are constructed of stainless steel with 0.952 cm (0.375 in.) thick acrylic plastic viewing panels. Either dry air or inert gas is used for static or dynamic operation of the glove boxes. Atmospheres are automatically controlled at a negative pressure by self-sensing supply and exhaust valves. In the event of a glove rupture or other emergency condition, an exhaust rate of up to 212 liters/sec ($450 \text{ ft}^3/\text{min}$) is automatically instituted. High efficiency filters at both the inlet and the exhaust provide complete plutonium containment. All glove box atmospheres are filtered at least three times before being released to the environs.

Secondary plutonium containment is provided by individual laboratories, and a third containment is provided by the building proper. Air samples, taken from individual laboratories and other locations throughout the building, are constantly being monitored for airborne activity. Further control of airborne contamination is accomplished with a building ventilation system that directs air flow from zones having

TABLE I. Gamma and X Radiations from Common Plutonium Isotopes⁽²⁾

Isotope	Half-Life	Gamma Ray		X-Ray		e/γ Ratio*
		Energy, keV	Yield, %	Energy, keV	Yield, %	
Pu ²³⁸	86.4 yr	43.8	3.8×10^{-2}	17	11	740
		99	8×10^{-3}	17	3.5×10^{-2}	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	9×10^{-3}	17	4	
Pu ²⁴¹	13.0 yr	145	2.8×10^{-4}			
Pu ²⁴²	3.79×10^5 yr		1×10^{-2}	17	4.1	
Pu ²⁴³	4.98 hr	12.2				

* Electron to gamma emission ratio

FIGURE 2. Typical Plutonium Fuels Laboratory Glove Box (Neg. 02125-1)

the lowest contamination probability to zones having the highest. Past experience has shown that this ventilation system is not only effective for reducing spread of contamination within an individual laboratory, but it limits costly plant shutdowns due to contamination spread between adjacent laboratories.

Plutonium deposition through open wounds and scratches is limited by inspiring good work habits and house-keeping practices in personnel. Sharp objects and tools are eliminated from work areas whenever possible, and personnel with known open wounds and scratches are required to obtain professional medical attention before conducting any work in areas containing plutonium. Because of the nature of some glove box operations, occasional glove penetrations are expected. To limit contamination spread resulting from such failures, personnel are required to survey their surgical gloves immediately upon removal of their hands from the glove box gloves. Additional surveys of hands and feet by an alpha-sensitive monitor are required of all personnel before leaving an individual laboratory or the process area proper.

A precautionary check for plutonium deposition in personnel is made by collecting and analyzing bioassay samples quarterly. Minimum detection by this method is about 1% of the maximum permissible body burden.⁽¹⁾

CONTROL OF EXTERNAL EXPOSURE

Whole body and hand exposure to PFL personnel is primarily caused by ion-

izing radiation (i.e., X and gamma radiation) from the plutonium. The magnitude of exposure is related to the geometry of the source, the amount of the material present, the isotopic composition, the time since separation, and the chemical or physical state (i.e., oxide, metal, etc.)^(1,2) The isotopic composition is dependent upon the irradiation parameters of the reactor that produces the plutonium, and will, therefore, vary among fuels produced in different reactors.

Theoretical equations for calculating surface dose rates caused by X and gamma radiation from infinite slabs of plutonium metal isotopes have been derived by Faust.⁽²⁾ A graph of the dose rates expected from various concentrations of isotopes Pu^{238} , Pu^{239} , Pu^{240} , Pu^{241} , and Pu^{242} is given in Figure 3. The dose rate from Pu^{241} increases with time because of the buildup of Am^{241} and U^{237} . Since the dose rate from Pu^{241} is significantly time dependent, it appears to be advantageous to fabricate plutonium containing a considerable amount of Pu^{241} as soon after separation as is possible. These equations have not been experimentally verified in the laboratory, but they do provide a guide for choosing ample shielding.

The highest Pu^{238} content of any plutonium processed in the PFL to date was 0.046%. The Pu^{239} , Pu^{240} , Pu^{241} , and Pu^{242} isotopic compositions of this material were 64.07, 29.29, 5.47, and 1.17%, respectively.⁽¹⁾ No excessive exposure was experienced with 631 g of this material when it was processed in a standard PFL glove box. Some attenuation of X and gamma radia-

$$\text{Pu}^{238}: D_s = 960f_{238}$$

$$\text{Pu}^{239}: D_s = 0.98f_{239}$$

$$\text{Pu}^{240}: D_s = 4.7f_{240}$$

$$\text{Pu}^{241}: D_s = [0.19t + 17(1 - e^{-0.102t})]f_{241}$$

$$\text{Pu}^{242}: D_s = 0.29f_{242}$$

Legend

D_s = Surface dose rate, rad/hr

f_i = Fraction by weight of the i th isotope of plutonium

t = Time since separation, days

FIGURE 3. Calculated X and Gamma Ray Surface Dose Rates from Plutonium Isotopes(2)

tion was provided by the 0.952 cm (0.375 in.) thick acrylic plastic viewing panels, but most of the shielding was accomplished by installing 0.64 cm (0.25 in.) thick lead-glass panels to the glove box exteriors. Dose rates to the hands were reduced by using 0.76 mm (0.030 in.) thick lead-impregnated gloves. Based on a 0.087 MeV- Cd^{109} measurement, the equiv-

alent lead thickness of these gloves was about 0.1 mm (0.004 in.). Lead-impregnated gloves 1.52 mm (0.060 in.) thick were considered, but were not used because they seriously impaired the manual dexterity of operators.⁽¹⁾

Neutron dose rates are related to the isotopic composition and the mass of the material present. Theoretical equations for calculating neutron dose

rates resulting from spontaneous fission of subcritical spherical masses of plutonium metal isotopes was first presented by Roesch⁽³⁾ and, subsequently, modified by Faust and Endres.⁽²⁾

A graph of the dose rate expected from various concentrations of isotopes Pu^{238} , Pu^{240} , and Pu^{242} is given in Figure 4. The validity of these equations has not been experimentally verified in the laboratory, but Endres and Faust have used them (with excellent results) to calculate neutron dose rates in one facility.

Personnel exposure caused by neutrons has been an infrequent problem in the PFL. Annual whole body exposure from neutrons has been estimated to be about 1/10 the annual exposure from X and gamma rays.* The cultivation of good work habits and housekeeping practices are the two principal reasons for the low exposure received by personnel.

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

FIGURE 4. Calculated Surface Dose Rates from Plutonium Metal Isotopes Due to Spontaneous Fission Neutrons⁽²⁾ (Subcritical Spherical Mass)

For example, uncontrolled accumulations of plutonium are avoided, plutonium is confined as much as possible, and dense shielding is used whenever necessary.

Exposures resulting from alpha-neutron reactions are usually much greater than exposures resulting from spontaneous fission. If combinations of plutonium and low atomic weight materials, such as PuF_4 (Table II), were to be processed in the PFL, neutron dose rates would be minimized by reducing glove box mass limits, employing neutron shielding, and requiring more rigid work procedures during fabrication.

CRITICALITY CONTROL

The basic criteria for establishing rules to prevent an inadvertent criticality at Battelle-Northwest is that at least two control conditions must fail before criticality is imminent. Rules for control and fissile material mass limits are stated in a set of criticality safety specifications. Mass limits are derived by a responsible nuclear physicist, and specifications are approved by experienced safety personnel. Criticality safety specifications for AEC sponsored work do not have to comply with a nuclear materials license.

TABLE II. *Neutron Radiations from Selected Isotopes and Compounds* ⁽²⁾

Isotope	Spontaneous Fission Half-Life, yr	ν n/Fission	Yield, n/g-sec	(α, n) Reaction Rate, n/g-sec
Pu^{236}	3.5×10^9	2.3	3.7×10^4	---
Pu^{238}	3.8×10^{10}	2.33	3.38×10^3	---
$\text{Pu}^{238}\text{F}_4$	---	---	---	2.1×10^6
$\text{Pu}^{238}\text{O}_2$	---	---	---	1.4×10^4
Pu^{239}	5.5×10^{15}	3.0	3×10^{-2}	---
$\text{Pu}^{239}\text{F}_4$	---	---	---	4.3×10^3
$\text{Pu}^{239}\text{O}_2$	---	---	---	4.5×10^1
Pu^{240}	1.22×10^{11}	2.25	1.02×10^3	---
$\text{Pu}^{240}\text{F}_4$	---	---	---	1.6×10^4
$\text{Pu}^{240}\text{O}_2$	---	---	---	1.7×10^2
Pu^{242}	7.1×10^{10}	2.18	1.7×10^3	---
$\text{Pu}^{242}\text{F}_4$	---	---	---	1.7×10^2
$\text{Pu}^{242}\text{O}_2$	---	---	---	2.7
Pu^{244}	2.5×10^{10}	2.3*	5.1×10^3	---

* *Estimated*

Glove boxes containing plutonium are assigned a mass limit according to the form of the plutonium and whether the plutonium is moderated or unmoderated. For example, unmoderated plutonium metal is limited to 2.8 kg per glove box and unmoderated plutonium, in the form of oxide, is limited to 5.0 kg per glove box. The difference in the two mass limits is based on the maximum possible densities of the two forms. Glove boxes containing moderated plutonium are limited to 230 g of plutonium per glove box, whether the plutonium is in metal, oxide, or some other form. All glove

boxes containing plutonium must be posted with a plutonium mass limit sign similar to the one shown in Figure 5.

The distinction between moderated and unmoderated plutonium is made through the hydrogen to plutonium atomic ratio (H/Pu). Plutonium is considered to be moderated if the H/Pu atomic ratio is greater than three, and is considered to be unmoderated if the H/Pu atomic ratio is less than or equal to three. This distinction not only accounts for moderation by water, but it accounts for moderation by plastic, wax, or some other hydroge-

FIGURE 5. Typical Criticality Safety Limit Posting Used in the PFL (Neg. 41503-2)

Whenever plutonium is entered in or removed from a glove box, the transfer must be recorded on a glove box inventory sheet (Figure 6). Up to 2 liters of hydrogenous liquid may be entered in a glove box containing unmoderated plutonium, but the liquid may not be deliberately mixed with the plutonium. The amount of hydrogenous liquid must be recorded on a glove box inventory sheet. Entries, appearing on glove box inventory sheets, must be verified by at least two individuals to assure against calculation errors.

Fuel rods must be clearly labeled as to enrichment, storage array limitations, and the required spacing from other fissile material. Plutonium not in the form of fuel rods must be clearly labeled, and show the total plutonium content and the required spacing from other fissile material.

PLUTONIUM FABRICATION CONTROLS

CONTROL OF FUEL QUALITY

Foreign materials in reactor fuels can be both hazardous and expensive.

* Source and Special Materials

FIGURE 6. Typical PFL Glove Box Inventory Sheet (Neg. 0660331-3)

*FIGURE 7. Slab Array of Fuel Elements
in the PFL (Neg. 41503-1)*

Contaminants can promote cladding corrosion and cause reduced fuel efficiency.

Fuel contamination has best been avoided by controlling the environment to which the fuel is exposed during processing. Only clean dry air or inert gases are used for glove box atmospheres; contaminating materials are eliminated from glove boxes as much as possible. Outgassing the fuel before loading it into cladding is an additional precaution taken with oxide fuel.

CONTROL OF FUEL YIELD

Scrap fuel is not only expensive to recover, but it presents storage problems, criticality control problems and, in some cases, unnecessary plutonium use charges. High fuel yields have been obtained by maintaining good house-keeping practices in glove boxes, and by establishing good fabrication processes. Glove boxes are periodically cleaned of fabrication wastes, and contaminating materials are avoided as much as possible.

CONCLUSIONS

Major problems to be faced by commercial users of plutonium will include:

- Control of personnel exposure
- Cost of scrap recovery
- Loss of contaminated equipment
- Loss of plutonium
- Waste disposal expenses (especially in large metropolitan areas).

The significance of each will primarily depend on the purpose of the facility.

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