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Title: **Effect of Compositional Variation in Plutonium on Process Shielding Design**

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Effect of Compositional Variation in Plutonium on Process Shielding Design

Radiation dose rate from plutonium with high ^{239}Pu content varies with initial nuclidic content, radioactive decay time, and impurity elemental content. Large variation in photon dose rate may result from variation in ^{241}Pu and ^{236}Pu initial contents. The high intensity, low-energy γ-emission from ^{241}Am, the first daughter of ^{241}Pu, may cause large doses for unshielded processing of bare plutonium. The high energy γ-emission from ^{208}Tl, a daughter in the ^{236}Pu decay chain, may be the primary photon dose contributor for heavily shielded processes. Variation in neutron dose rate results from variation in impurity (α,n) reactions and spontaneous fission, due to variation in the ^{241}Am and ^{240}Pu contents, respectively. Because dose variation implies shielding variation, knowledge of the composition of the plutonium to be processed should be incorporated in shielding design analysis.

The two idealized states of "old plutonium" and "clean plutonium," whose initial compositions are given in Table I, provide approximate upper and lower bounds on dose rate variation. Old plutonium has not undergone chemical separation of non-plutonium elements since initial separation following reactor production and has aged to produce maximum dose rate. Clean plutonium has undergone enough separation and aging to eliminate neutron dose from impurity (α,n) reactions and photon dose from ^{241}Am and ^{236}Pu progeny. The clean plutonium state can be approached with existing separation technology.[1-4]

Whole-body dose rates were calculated for the two composition states, using unshielded and shielded plutonium spheres of varying density. The dose rates from these variable density spheres are similar to those from expanded plutonium configurations encountered during processing. The dose location of 40 cm from the sphere center is representative of operator standoff for direct handling of plutonium inside a glove box. The radiation sources were

generated by using ORIGEN-S[5] to calculate nuclide decay and photon energy spectra and SOURCES[6] to calculate neutron energy spectra. Radiation transport was performed with MCNP-4A[7], incorporating the ANSI/ANS-1991 fluence-to-dose factors[8].

The results in Table II have shielding implications for glove boxes with only structurally inherent shielding, especially for processing of old plutonium in an expanded configuration. To preclude streaming through the glove and viewing ports, additional photon shielding equivalent to the glove box steel walls must be located between the plutonium and the ports. However, the inconvenience to hands-on operations imposed by the additional shielding may necessitate remote handling. Even with a shielding equivalency design, it is unlikely that the 1-rem annual dose design requirement[9] would be met for exposure times needed for most processes.

Further reduction in total dose rate by using lead to reduce photon dose rate is shown in Table III for two density cases representing compact and expanded plutonium configurations. As lead thickness increases, the relative difference in total dose rate between old and clean plutonium diminishes, due to the rapidly increasing attenuation of ^{241}Am high-intensity, low-energy γ-emission in old Pu. Photon dose rate becomes insignificant after lead thicknesses of 1-2 cm and $\gtrsim$5 cm for clean and old plutonium, respectively. Significant additional dose rate reduction can be achieved only by reducing neutron dose rate with thick hydrogenous shielding. Maintaining shielding equivalency at the glove ports with thick shielding would almost certainly dictate remote handling.

Table I

Initial Nuclidic and Impurity Contents of Plutonium Metal for Calculating Photon and Neutron Sources

Plutonium Isotopes and ^{241}Am			Impurities		
Nuclide	Initial Content (wt % of Pu)		Element	Content (ppm)	
	Old Pu	Clean Pu		Old Pu	Clean Pu
^{236}Pu	2.5e-6 (25 ppb)*	0	Be	3†	0
^{238}Pu	0.05†	0.05†	B	50†	0
^{239}Pu	92.33‡	93.89‡	C	200†	0
^{240}Pu	6.50†	5.96§	Mg	500†	0
^{241}Pu	1.00†	0	Al	130†	0
^{242}Pu	0.10†	0.10†			
^{241}Am	0.02 (200 ppm)†	0			

*Value is based on the isotopic content of plutonium calculated to be produced in blanket region of a Liquid-Metal-Fast-Breeder Reactor[10]. 10 ppb of ^{236}Pu and 0.02 wt % of ^{238}Pu were produced in the blanket region. The 10 ppb of ^{236}Pu was scaled up to 25 ppb to be consistent with the above listed 0.05 wt % ^{238}Pu limit. The rationale for scaling is the expected small variation of ^{236}Pu:^{238}Pu ratio with variation of ^{238}Pu content because both ^{236}Pu and ^{238}Pu are produced from neutron irradiation of ^{237}Np. Other results in the reference suggest a slowly varying ^{236}Pu:^{238}Pu ratio with large increases in ^{238}Pu content.

†Maximum value from 1985 Rocky Flats specification[11].

§Maximum shipment-average value from 1985 Rocky Flats specification.

‡Set to 100 minus sum of wt %s of the other nuclides.

Table II

Dose Rates from 4.5-kg Homogeneous Plutonium Spheres
Unshielded and Shielded by 0.7-cm Thick 304L Stainless Steel

(0.7-cm thick 304L stainless steel is representative of glove box walls)

Calculational model is 4.5-kg plutonium sphere either unshielded or centered inside 304L stainless-steel spherical shell with thickness of 0.7 cm and inside radius of 15 cm. Dose location is 40 cm from center of plutonium sphere.

Density (g/cm^3)	Dose Rate (mrem/h)*									
	Photon				Neutron+(n,γ)†		Total			
	Unshielded		Shielded		Unshielded		Unshielded		Shielded ‡	
	Old Pu	Clean Pu	Old Pu	Clean Pu	Old Pu	Clean Pu	Old Pu	Clean Pu	Old Pu	Clean Pu
19.86 §	23.40	1.263	2.617	0.7342	4.380	2.982	27.8	4.24	7.00	3.72
2.546	90.14	4.837	8.107	2.758	1.731	1.137	91.9	5.97	9.84	3.90
1.074	158.4	8.347	11.83	4.570	1.632	1.066	160	9.41	13.5	5.64
0.489	266.3	13.28	15.66	6.675	1.597	1.040	268	14.3	17.3	7.71

*Listed values are sample means with relative errors < 1%. (Relative error = one standard deviation of the mean ÷ sample mean)

†Less than 1% (n,γ) contribution to neutron+(n,γ) dose rate.

‡The shielded neutron+(n,γ) contribution is assumed to be equal to unshielded contribution. The neutrons are not significantly attenuated by the 0.7-cm thick 304L stainless steel; the (n,γ) photons may be significantly attenuated, but their dose rate contribution is insignificant.

§Theoretical density of alpha-phase plutonium metal.

Table III

Dose Rates from 4.5-kg Homogeneous Plutonium Spheres Enclosed by Spherical-Shell Lead Shields

Each lead shield has inside radius of 15 cm and is concentric with the plutonium sphere. Dose location is 40 cm from center of plutonium sphere.

Shield Thickness (cm)	Photon Dose Rate (mrem/h)				Total Dose Rate (mrem/h) †			
	Pu Density of 19.86 g/cm^3		Pu Density of 1.074 g/cm^3		Pu Density of 19.86 g/cm^3		Pu Density of 1.074 g/cm^3	
	Old Pu	Clean Pu	Old Pu	Clean Pu	Old Pu	Clean Pu	Old Pu	Clean Pu
0	23.40	1.263	158.4	8.35	27.8	4.24	160	9.41
0.02	9.88	0.940	56.6	6.06	14.3	3.92	58.2	7.13
0.05	4.22	0.837	20.3	5.36	8.60	3.82	21.9	6.43
0.1	2.75	0.732	12.2	4.62	7.13	3.71	13.8	5.69
0.2	2.35	0.583	10.0	3.57	6.73	3.56	11.6	4.64
0.5	1.78	0.306	6.69	1.72	6.16	3.29	8.32	2.79
1.0	1.26	0.115	4.20	0.566	5.64	3.10	5.83	1.63
2.0	0.752	0.0249	2.25	0.101	5.13	3.01	3.88	1.17
3.0	0.484	0.00927	1.38	0.0325	4.86	2.99	3.01	1.10
5.0	0.211	0.00252	0.571	0.00770	4.59	2.98	2.20	1.07
Unshielded Neutron+(n,γ) Dose Rate (mrem/h) (from Table II)					4.380	2.982	1.632	1.066

*Listed values are sample means with relative errors < 1%. (Relative error = one standard deviation of the mean ÷ sample mean)

†The neutron+(n,γ) dose rates are not significantly reduced by lead with listed thicknesses. The unshielded neutron+(n,γ) dose rates have added to the shielded photon dose rates to obtain estimates of total dose rate.

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