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CALCULATION OF GAMMA DOSE RATES
AT THE SURFACE
OF PLUTONIUM OXIDE SOURCES

H. H. Van Tuyl

January 1970

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AT THE SURFACE OF PLUTONIUM OXIDE SOURCES

By

H. H. Van Tuyl

Fuels and Materials Department
Chemistry and Metallurgy Division

January 1970

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ABSTRACT

A computer program, PUSHLD, has been developed for calculating gamma dose rates at the surface of plutonium oxide sources. Radiation attenuation due to self-shielding and external shielding were included, along with secondary radiations from Compton scattering, X-rays from photoelectric absorption, and electrons from photoelectric absorption. Shielding curves are presented which may be used to determine the surface dose rate from a plutonium source of any isotopic composition and age. Calculated dose rates agreed within 20% with measured values for several plutonium isotopic compositions and three shield materials.

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CALCULATION OF GAMMA DOSE RATES
AT THE SURFACE OF PLUTONIUM OXIDE SOURCES

1.0 INTRODUCTION

Plutonium has been processed for many years with only occasional problems due to excessive gamma and neutron dose rates. As larger amounts of high exposure plutonium become available and are fabricated into fuel elements for recycle, the dose rates during plutonium handling become increasingly important. The quantity of plutonium being processed is increasing, and the content of the troublesome plutonium isotopes, particularly ^{238}Pu and ^{241}Pu , is also increasing. Both of these trends increase the dose rate from plutonium. Thus, a better understanding of the dose rate from plutonium sources is necessary for optimum design of process equipment and facilities.

Calculations of gamma dose rates were undertaken to permit intercomparison of experimental, semi-empirical, and theoretical dose rates. Good agreement between experimental and theoretical surface dose rates would improve credibility of advanced calculations for extended source geometries and distances from the source, thereby reducing the need for extensive measurements at all geometries of interest. The calculations are being compared with measurements of surface gamma dose rates, which are being made at BNWL.⁽¹⁾ With a variety of plutonium sources, a semi-empirical equation for surface gamma dose rates was also developed by Roesch⁽²⁾ and has been modified by Faust⁽³⁾.

Most plutonium isotopes emit gamma radiation of less than 100 keV and only small amounts of higher energy radiations⁽⁴⁾. Much of this low energy radiation is absorbed in the source, accompanied by the emission of lower energy X-rays. In the absence of external shielding, most of the dose rate from plutonium sources is due to X-rays from the L and M

shells. X-rays from the N and lower shells are of such low energy as to be of little practical external consequence. Conventional computer programs, such as ISOSHLD-II⁽⁵⁾, do not provide sufficiently accurate treatment of radiations below 100 keV, and thus give only approximate dose rates for plutonium sources. A special computer program PUSHLD, was therefore developed to assure accurate calculations for both low energy and higher energy radiations of importance in plutonium dose rate determinations.

2.0 SUMMARY

A computer program, PUSHLD, has been developed for calculating gamma dose rates at the surface of plutonium oxide sources. For mathematical simplicity, the source was assumed to be a hemisphere, with the dose point at the center of the sphere. Radiation attenuation due to self-shielding and external shielding and production of secondary radiation were included in the program, but geometry effects due to the movement of the dose point away from the surface of the source were not included. Secondary radiations of photons from Compton scattering, X-rays from photoelectric absorption, and electrons from photoelectric absorption were included.

Shielding curves are presented for pure plutonium isotopes and their significant daughters. These curves may be used to determine the dose rate from a plutonium source of any isotopic composition and age. Additional shielding curves are given for a number of plutonium isotopic compositions. The attenuation factor due to a given shield is dependent on the plutonium isotopic composition and age, and on other shielding materials between the source and detector. The effect of time on dose rate cannot be evaluated by simple rules of thumb, since it is dependent on shield type and thickness as well as plutonium isotopic composition and age since separation.

Calculated dose rates were compared with measured values for several plutonium isotopic compositions and three shield materials. Agreement was within 20%, and no significant systematic variations were observed.

3.0 PRINCIPLES OF GAMMA EMISSION AND SHIELDING

The shielding calculations presented in this report represent selected, idealized conditions which are seldom encountered by plutonium processors. Application to real systems requires judicious extrapolation of the idealized data. A general knowledge of the basic gamma emission and attenuation processes is required to understand the derivation of the idealized data, and to apply the data to practical systems.

Principles of gamma emission and shielding are well documented in many places^(6,7,8). The most important fundamentals are summarized here to make the remainder of the report more intelligible. Extra emphasis is placed on factors which affect low energy radiations, since these are usually the major contributors to dose rates from plutonium.

PHOTON EMISSION

A gamma ray is a high energy electromagnetic photon resulting from nuclear rearrangements. The energy range of interest in plutonium shielding is from 0.04 to 2.6 MeV. Gamma rays occur when an excited nuclear state de-excites to a lower energy state of the same nuclide. Two or more gamma photons may be emitted in de-exciting a single excited state of an atom. Most of the plutonium gamma rays arise when alpha decay populates an unstable excited state of the daughter nuclide. This unstable state will then de-excite by emission of one or more gamma rays until the nucleus reaches the ground state (which is the most stable state of the nuclide). Gamma rays also occur when an excited nuclear state is created by other means, such as beta emission.

Characteristic X-rays are different from gamma rays only in their mode of formation and range of energies. While gamma rays originate in the nucleus, X-rays originate in the extra-nuclear electron shells. When a K shell electron is ionized, an electron from a higher shell or from outside the atom will fill the vacancy. This is accompanied by the emission of quantized electromagnetic radiation (called K X-rays when the K shell is being filled) equal in energy to the difference between the initial and final energy states of the electron. The K X-ray group consists of several photon energies, since electrons in several energy levels are available for filling the K shell. For convenience, the $K\alpha_1$ X-ray energy is often taken to represent all of the K X-ray group. Characteristic X-rays have energies below 0.15 MeV.

During emission of a gamma or X-ray photon, internal photoelectric absorption may occur, in which the photon energy is absorbed by an extra-nuclear electron of the same atom. The kinetic energy of the ejected electron is equal to the initial photon energy less the binding energy of the electron. This binding energy is approximately equal to the energy of the X-ray for the shell from which the electron was ejected. The secondary X-ray, emitted when an electron fills the site vacated in the conversion process, may also be internally converted. Thus, the radiation observed from gamma or X-ray transitions in a given nuclide may consist partly of secondary X-rays and mono-energetic electrons, and fewer primary photons than expected.

The fraction of X-rays which escape internal conversion is called the fluorescent yield, ω . Values of the K shell fluorescent yield are given approximately by

for values of Z above 40. The L shell fluorescent yield is lower than the K shell fluorescent yield on an atomic number basis, but about the same on an X-ray energy basis.

The extent of internal conversion of gammas is dependent on both the energy and the type of transition (i.e., spin and parity change). Conversion is very common below 0.2 MeV, and usually negligible above 1 MeV. The extent of internal conversion has been determined experimentally for many nuclides. The result is often given as α , which is the ratio of conversion electrons to escaping primary photons for a specific gamma transition. Another common expression is α_K , which is the ratio of K shell conversion electrons to escaping primary photons. In this latter case, conversion in the L and higher shells should also be included in determining the number of gamma photons which leave the nucleus. Care must be exercised to see whether gamma data have already been corrected for internal conversion before applying the appropriate corrections. Many tabulations of data have part of the gamma data corrected for internal conversion and part of it uncorrected with no clear indication of which is which.

PHOTON ATTENUATION PROCESSES

As photons pass through matter, they are removed or reduced in energy by three principal mechanisms: Compton scattering, photoelectric absorption, and pair production. These processes, together with other less important processes, contribute to the attenuation of photon intensity.

Attenuation Coefficient

The most important quantity characterizing the attenuation of gamma and X-radiation is the attenuation coefficient, μ . This quantity, which depends on the photon energy (E) and the atomic number (Z) of the medium, may be defined as the relative

probability per unit path length that a photon will undergo an interaction. In a thin layer, dt , within a slab shield, a reduction will occur in the intensity (I) of the photon beam due to absorption or scattering. The resulting fractional reduction of the beam intensity, $-dI/I$, is proportional to the shield thickness, dt , with the attenuation coefficient, μ , being the proportionality constant; i.e.,

$$-dI/I = \mu dt.$$

Integrating this equation with an incident beam intensity, I_0 , and constant absorption coefficient throughout the slab (homogeneous medium) gives

$$I(t) = I_0 e^{-\mu t}$$

This is the basic equation for gamma or X-ray attenuation, but modifications must be made for geometry considerations and secondary radiations.

The attenuation coefficient has a dimension of inverse length, and thus varies as the absorber density, ρ . This density dependence is commonly removed by use of the "mass attenuation coefficient", μ/ρ , instead of the linear attenuation coefficient. If μ is in units of cm^{-1} and ρ is in units of g/cm^3 , the mass attenuation coefficient will be expressed in the customary units of cm^2/g . Values of selected mass attenuation coefficients are shown in Figure 3-1. More complete tabulations are given by Hubbell and Berger⁽⁹⁾. More recent values⁽¹⁰⁾ for elements above thorium are appreciably different from these values, but the older values are believed to be good for lower atomic numbers. Values of the principal components of the mass attenuation coefficient for lead are shown in Figure 3-2, along with the total attenuation coefficient.

FIGURE 3-1. Mass Attenuation Coefficients for
Common Shield Material

FIGURE 3-2. Components of Mass Attenuation Coefficient for Lead

Photoelectric Absorption

In photoelectric absorption, also called the atomic photoeffect, a photon disappears and an electron is ejected from the atom. The electron carries away all the energy of the absorbed photon, less the energy with which the electron was bound to the atom. It is thus very similar to internal conversion. The K-shell electrons, which are the most tightly bound, are the most important since the probability of interaction is higher and the energy of secondary X-radiation is higher. However, if the photon energy is below the binding energy of a given shell, an electron from the shell cannot be ejected. Hence, a plot of the absorption coefficient versus photon energy exhibits characteristic "absorption edges" which may be seen as discontinuities in Figures 3-1 and 3-2. As a crude approximation, the photoelectric attenuation coefficient varies as the 3.5 power of the atomic number, and inversely as the cube of the photon energy. The contribution due to L and higher shells increases with atomic number--from zero for hydrogen to about 25% of the total photoelectric attenuation coefficient for uranium.

Compton Scattering

In Compton scattering, a photon collides with an electron, loses some of its energy and is deflected from its original direction of travel. Compton scattering varies with the charge-to-mass ratio of the absorber, Z/A , and is thus roughly independent of atomic number. Hydrogen has a high attenuation coefficient due to Compton scattering because of its unusually high charge to mass ratio.

Compton scattering occurs over the entire energy range with a rather slow decrease in probability at higher energies. It is the predominant mechanism for energy loss in the region

between photoelectric absorption at low energies and pair production at high energies. For low atomic number materials, this covers almost the entire energy range of interest, while for high atomic number materials, Compton scattering is important only in the vicinity of 1 to 4 MeV.

Pair Production

In electron-positron pair production, a photon disappears in the field of a charged particle, and an electron-positron pair is formed. Pair production requires a photon of at least 1.022 MeV (or $2 mc^2$) for production in the field of the nucleus, and 2.044 MeV (or $4 mc^2$) for production in the field of an electron. Pair production occurs chiefly in the field of the nucleus, especially for high atomic number materials. The attenuation coefficient due to pair production varies approximately as the square of the atomic number. As may be seen in Figure 3-2, pair production is relatively unimportant in the energy region of interest for plutonium shielding.

Other Processes.

In Rayleigh scattering, photons are scattered by bound atomic electrons so that the atom is neither ionized nor excited. This effect is usually disregarded in shielding calculations, since little energy loss occurs and scattering is either at a small angle or a small fraction of the total attenuation.

At higher energies (usually above 5 MeV), a photonuclear effect occurs which is similar to the photoelectric effect, but a nucleon (usually a neutron) is emitted instead of an electron. The photonuclear threshold is sufficiently high to make this process of little or no concern in plutonium processing.

Additional attenuation processes have been observed or postulated, but they are completely insignificant with respect to attenuation of photons in the energy range of interest.

BUILDUP OF SECONDARY RADIATION

Each attenuation process results in some secondary radiations. The photoelectric effect produces X-rays and electrons, Compton scattering produces the scattered photons, and pair production results in the 0.511 MeV radiations due to the eventual annihilation of the positron. These secondary radiations result in a higher dose rate than predicted by the simple exponential law. This increase is usually included in calculations by use of one or more "**buildup factors**", which are the ratio of the experimentally measured dose rates to the dose rate calculated by the simple exponential law. (This definition actually applies only to a dose rate buildup factor, which is the one most commonly used in shielding calculations.) The buildup factor is dependent on the photon energy, shield material, and shield and source geometry.

Compton Buildup Factors

Except at low energies where the photoelectric effect predominates, the Compton scattering process is responsible for most of the secondary radiation. The calculation of buildup factors for even simple situations is a formidable task since the source and shield geometries, probable energy distribution, and direction of gamma rays must be considered, and the photons may be scattered many times before finally being removed by photoelectric absorption. Factors for a few simple, idealized cases and a few different elements have been calculated by different workers, but the data currently accepted as best were calculated by Nuclear Development Associates⁽¹¹⁾ using a National Bureau of Standards computer. Although the project

required several man-years of effort and a large amount of machine time, the results of interest to most shielding problems are quite meager in number. Some of these data, i.e., dose rate buildup factors for a point isotropic source in an infinite medium, are most important for plutonium shielding. The range of values covered by Nuclear Development Associates is adequate for most normal shielding problems, but several situations occur in which data outside this range are required. In particular, the shielding of gamma and X-rays from trans-uranic elements requires application of buildup factors for lower energy radiations. The calculations have therefore been extended to lower energies (below 0.5 MeV) by a modified "straight ahead" approximation that is much simpler although less accurate than the moments method used by Nuclear Development Associates. The two methods usually agreed within 20% over the range covered by the moments method. Results for iron and lead, which neglect the lead L-shell edges, are shown in Figures 3-3 and 3-4.

X-Ray Buildup Factors

At energies slightly above the K X-ray absorption edge, the secondary X-ray from photoelectric absorption is much more penetrating than the primary photon. The X-ray buildup factor approaches the exponential of the difference in mean free paths for the primary photon and the X-ray. The importance of the effect is clearly illustrated in Figure 3-4. In spite of its large magnitude, the X-ray buildup factor is less of a problem than the Compton buildup factor. The X-ray buildup factor is important only near the X-ray absorption edge; and the attenuation coefficient is high in this region, even for the secondary X-ray. Thus, a small additional thickness of material will result in a large absorption of secondary X-rays. Furthermore, a single secondary photon is all that

FIGURE 3-3. Buildup Factors for Iron

FIGURE 3-4. Buildup Factors for Lead

usually needs to be considered, whereas with Compton scattering, a great number of successive scatterings may be required to properly account for the buildup of secondary radiation. Secondary X-rays can contribute an appreciable fraction of the total dose rate through thin shields such as leaded gloves. Therefore, this source of radiation must be carefully considered.

Electron Buildup Factors

The electrons produced by the photoelectric effect can also contribute to the dose rate. The range of electrons in matter is usually much less than that of photons. However, electrons produced near the surface of a shield can escape the shield to cause additional ionization. This effect is most pronounced with high atomic number shields such as lead or uranium, since the photoelectric effect is greater with these shields. Electrons lose energy by numerous ionizing collisions and thus have a rather well defined path length in an absorber. Their path is not straight, however, so that the effective penetration through an absorber is about half of the total path length. A small amount of low atomic number absorber placed on the outside of a high atomic number shield would reduce the electron buildup factor considerably. Even the layer of dead skin (about 7 mg/cm^2) reduces the buildup factor due to low energy electrons. However, the contribution of electrons to the dose rate is real, and must be considered in accurate calculations.

DOSE RATE UNITS

The original dose rate unit, the Roentgen, is the amount of gamma or X-radiation which, in 0.001293 g of air, will produce ions carrying one esu of quantity of either sign. A Roentgen corresponds to an energy absorption of about 87.7 ergs/g in air or 96.5 ergs/g in water. In recent years, both of these values have been revised from lower values. The

value in water is higher than that in air simply because water has a higher energy absorption coefficient. The quantity of ionizing radiation incident on a detector is often calculated as a flux, ϕ , usually expressed as photons or particles/cm²-sec. Dose rate units, however, are related to the effect of the radiation on its surroundings, rather than numbers of photons or particles. A photon flux can be converted to a dose rate by simple application of conversion factors to yield the equation

$$D = 6.576 \times 10^{-5} \phi E \mu_E / \rho,$$

where D is the dose rate in R/hr, E is the photon energy in MeV, and μ_E/ρ is the energy absorption coefficient for air in cm²/g. The energy absorption coefficient for air varies substantially with energy, especially below 0.05 MeV. Between 0.06 and 2 MeV, it is constant within about 20%, with a value of 0.0288 cm²/g (or 3.72×10^{-5} cm⁻¹). If the dose rate in a material other than air is desired, the energy absorption coefficient for that material must be used in place of the coefficient for air.

The Roentgen applies only to gamma or X-radiation, but a similar concept is used to define dose rates from beta radiation and electrons. The unit usually used in this report is the rad, which represents 100 ergs absorbed energy per gram of material. The rad and Rep are often used interchangeably (in spite of about 3.5% error), while the Roentgen is reserved specifically for gamma and X-radiation. The rad is used as the dose rate unit in this report since both photons and electrons are contributing to the dose rate.

4.0 NUCLEAR DECAY PROPERTIES

The isotopic composition of plutonium is not constant with time, but changes as the shorter-lived plutonium isotopes decay. Two plutonium isotopes give rise to daughter products which substantially affect dose rates. Shielding requirements are therefore dependent not only on the plutonium isotopic composition, but also on the time since separation (or chemical purification) of plutonium from its daughter products. The amount and energy distribution of penetrating radiation from each nuclide is different, and must be known to permit calculation of the dose rate from that nuclide.

DECAY RELATIONSHIPS

The most important decay relationships among plutonium isotopes and their daughters are shown in Figure 4-1. Most of the plutonium isotopes decay to long-lived uranium daughters (half-life of over 10^5 years). Only small amounts of these uranium isotopes are formed; they and their daughters do not contribute appreciably to the dose rate. Two plutonium isotopes do produce daughters which can be important in shielding considerations. ^{241}Pu decays primarily by beta emission to form ^{241}Am . Since americium has a half-life longer than ^{241}Pu , the americium activity associated with a plutonium source continues to increase for many years. A small amount of the ^{241}Pu decays by alpha emission to form ^{237}U . Since the half-life of ^{237}U is much shorter than that of ^{241}Pu , it reaches an equilibrium activity in about two months.

Decay of ^{236}Pu is much more complicated. No very long-lived nuclides are formed until the decay chain reaches stable ^{208}Pb . Even though the amount of ^{236}Pu is very small (usually less than one part per million), its daughters can have an appreciable effect on dose rates from plutonium sources as measured through heavy shielding.

FIGURE 4-1. Daughters Formed by Decay of Pu Isotopes

Radiation from long-lived nuclides, such as the plutonium isotopes, is best characterized on the basis of the amount of nuclide present. The same is true of long-lived daughters produced by the decay of plutonium isotopes, such as ^{232}U (produced from ^{236}Pu) and ^{241}Am (produced from ^{241}Pu). Nuclides with much shorter half-lives than those of the parents grow in until an equilibrium amount is reached--at which time the rate of formation is equal to the rate of decay. The amount of short-lived daughter is then most easily expressed in relation to this equilibrium activity. The ^{237}U formed by decay of ^{241}Pu and the decay chain formed from ^{232}U are treated in this way. The weight fraction of short-lived nuclides is always small, but the activity comes into equilibrium with the activity of the parent nuclide.

The decay of the plutonium isotopes is shown in Figure 4-2, along with the amount of conversion to long-lived daughters. Figure 4-3 shows the approach to equilibrium in the formation of ^{237}U by the decay of ^{241}Pu . Most of the ^{241}Pu decays to ^{241}Am , but a small fraction decays by alpha emission to form ^{237}U . The ratio of ^{237}U activity to ^{241}Pu activity is given by

$$\frac{A_{\text{U}}}{A_{\text{Pu}}} = \frac{R \lambda_{\text{U}}}{\lambda_{\text{U}} - \lambda_{\text{Pu}}} \left[1 - e^{-(\lambda_{\text{U}} - \lambda_{\text{Pu}}) t} \right],$$

where A denotes the activity, λ is the decay constant ($\log_e 2/\text{half-life}$), t is the time since separation of uranium from the plutonium, and R is the fraction of ^{241}Pu decays which produce ^{237}U . As t becomes large, the ratio approaches the saturation activity,

$$\frac{R \lambda_{\text{U}}}{\lambda_{\text{U}} - \lambda_{\text{Pu}}}$$

FIGURE 4-2. Decay of Pu Isotopes and Growth of Daughters

FIGURE 4-3. Growth of ^{237}U Activity into Equilibrium with ^{241}Pu

The saturation activity of ^{228}Th in equilibrium with ^{232}U is obtained in the same manner as shown in Figure 4-4. More remote daughters of the same decay chain have saturation activities given by

$$S_d = \frac{\lambda_d S_p}{\lambda_d - \lambda_p},$$

where S is the saturation activity ratio and the subscripts p and d refer to parent and daughter respectively.

PHOTON EMISSION

The energy and abundance of penetrating radiation from plutonium isotopes and their daughters can be derived from published decay scheme data. Most of the photon spectra used in this report were derived from the tabulation by Lederer, Hollander and Perlman⁽⁴⁾. Transition abundances were derived from branching ratios shown on the decay schemes in most cases, and photon abundances were derived from these by applying internal conversion coefficients. For many of the isotopes of interest, gamma photon abundances were available in the tabulations either as percent of transitions or as relative photon abundance. X-ray photon abundances were derived from the transition abundances, together with whatever data were available on transition type, electrons per gamma, internal conversion ratios among the various shells, and fluorescence yield. Data on X-ray energies, fluorescence yield, and internal conversion coefficients were obtained from appendices to the decay scheme tabulations⁽⁴⁾.

A complete tabulation of X-rays from each nuclide would involve hundreds of energy values. For simplicity, the K group of X-rays was reduced to only two, the $K\alpha$ and $K\beta$. Similarly, the L1 and L2 groups were reduced to only two groups, while three groups were used for the L3 shell. An even more drastic

FIGURE 4-4. Saturation of Daughters with ^{232}U Parent

simplification was made for the N and M groups by reducing them to a single group each. These simplifications seem to be justified because of the small differences between actual X-ray energies and assumed values, and uncertainties in branching ratios for the production of the various X-ray groups.

The photon abundances used for PUSHLD calculations are shown in Table 4-1, except that the X-ray abundances are shown by shell only instead of being divided into subgroups. For the ^{236}Pu decay chain, very few X-rays are shown because of the low abundance of ^{236}Pu . X-rays from other plutonium isotopes would normally overshadow the contribution from ^{236}Pu .

*TABLE 4-1. Photons From the Decay of
Pu Isotopes and Their Daughters*

^{239}Pu					
Energy, keV	Abundance, ppm	Energy, keV	Abundance, ppm	Energy, keV	Abundance, ppm
U-N-x	3800	144.2	2.0	367.4	0.64
U-M-x	50000	161.4	0.9	368.7	0.56
U-L3-x	28000	171.4	0.7	375.2	10
U-L2-x	32000	179.1	0.5	380.4	2.0
U-L1-x	6500	189.1	0.6	382.9	1.6
U-K-x	45	195.6	0.8	393.5	4.0
13.0	3	203.5	3.6	413.7	10
38.6	60	255.5	0.6	422.6	0.6
46.2	6.4	264.0	0.24	426.7	0.12
51.6	160	297.6	0.36	451.6	1.4
56.9	6.4	311.8	0.20	639.0	0.17
68.6	5.6	321.1	0.32	645.0	0.20
77.6	4.4	323.9	0.36	652.0	0.13
98.0	10	333.0	3.2	659.0	0.16
103.0	1.6	336.3	0.72	718.0	0.04
116.0	7.2	341.7	0.48	758.0	0.06
129.3	40	345.1	3.5	771.0	0.18

TABLE 4-1. (contd)

^{238}Pu					
Energy, keV	Abundance, ppm	Energy, keV	Abundance, ppm	Energy, keV	Abundance, ppm
U-N-x	2000	U-K-x	2.0	743	0.09
U-M-x	27000	43.5	380	767	0.35
U-L3-x	70000	100.0	80	786	0.06
U-L2-x	78000	153.1	10	854	0.05
U-L1-x	3100	203	0.04	1001	0.008
^{240}Pu					
U-N-x	1800	U-L1-x	2700	160.0	12
U-M-x	24000	U-K-x	2.0	642.0	0.14
U-L3-x	59000	45.3	500	688.0	0.04
U-L2-x	70000	103.6	120		
^{241}Pu					
U-N-x	0.03	U-L2-x	0.56	U-K-x	17
U-M-x	0.40	U-L1-x	2.8	145.0	1.6
U-L3-x	0.23				
^{237}U					
Energy, keV	Abundance, %	Energy, keV	Abundance, %	Energy, keV	Abundance, %
Np-N-x	1.3	33.2	0.019	221.8	0.022
Np-M-x	17	43.4	0.074	234.4	0.021
Np-L3-x	16	51.0	0.20	267.5	0.76
Np-L2-x	11	59.5	36	332.4	1.3
Np-L1-x	14	64.8	1.3	335.4	0.10
Np-K-x	56	103.0	0.60	337.7	0.027
13.8	0.001	164.6	2.0	368.6	0.050
26.4	2.4	208.0	23	370.9	0.12

TABLE 4-1. (contd)

^{241}Am					
Energy, keV	Abundance, ppm	Energy, keV	Abundance, ppm	Energy, keV	Abundance, ppm
Np-N-x	4400	165	0.4	427	0.3
Np-M-x	59000	169	1.0	570	0.1
Np-L3-x	100000	208	6.0	597	0.1
Np-L2-x	280000	222	0.4	619	0.8
Np-L1-x	160000	268	0.3	641	0.1
Np-K-x	40	293	0.2	653	0.5
26.4	25000	312	0.3	662	5.0
33.2	300	323	1.6	680	0.06
42.6	6.0	332	1.8	689	0.4
43.4	700	335	6.0	695	0.1
55.6	260	338	0.1	710	0.09
59.54	360000	369	3.0	722	2.8
67.5	10	371	1.3	737	0.12
99.0	240	377	1.6	757	0.13
103.0	190	384	0.3	767	0.08
125.0	50.0	420	0.4	771	0.10
146.0	2.0				

^{242}Pu					
U-N-x	1800	U-L3-x	59000	U-L1-x	2700
U-M-x	24000	U-L2-x	70000	44.7	500

TABLE 4-1. (contd)

^{236}Pu					
Energy, keV	Abundance, ppm	Energy, keV	Abundance, ppm	Energy, keV	Abundance, ppm
47.6	31	472.8	0.09	584	0.06
109	120	516	1.7	645	2.4
165	6.6	564	0.9		
^{232}U					
Energy, keV	Abundance, %	Energy, keV	Abundance, %	Energy, keV	Abundance, %
Th-L-x	30.0	129.0	0.082	327.5	0.0034
57.5	0.21	270.0	0.0038		
^{228}Th					
Ra-L-x	25.0	132	0.19	216	0.29
84.5	1.6	167	0.12		
^{224}Ra					
241	4.2				
^{212}Pb					
Bi-K-x	33	177	0.24	300	3.6
115	0.7	238.6	47	415	0.16
^{212}Bi (and ^{208}Tl Daughter)					
40	1.1	453	0.1	893	0.4
41	0.40	486	0.18	953	0.1
211	0.10	511	9.0	1076	0.7
233	0.10	583	31	1094	0.2
253	0.40	727	7.1	1513	0.29
277	3.6	763	0.7	1620	1.6
288	0.28	785	1.0	1800	0.11
328	0.1	860	4.3	2615	36

5.0 METHODS OF CALCULATION

Calculations were made by a point kernel method with secondary radiation included through the use of buildup factors. Shield materials were selected to represent normal structural and shielding materials commonly used in glove boxes. Because of the large amount of low energy radiation, considerable effort was expended in developing and applying adequate low energy buildup factors.

GEOMETRY

One objective of this study was to compare calculations with experimental measurements of surface dose rates. Recent measurements⁽¹⁾ have been made using a cylindrical source with a diameter of about 2.5 in. and a height of about 0.5 in., containing 60 to 80 g of PuO_2 . Calculations were made to represent this geometry, but with simplifying assumptions. The source was considered to be a hemisphere, and the dose was calculated at the center of the sphere. Plutonium is a very effective shielding material for most of the radiations of interest, so the radiation reaching the detector usually comes from only a thin surface layer of the plutonium. From this standpoint, essentially any geometry could be used to obtain a good estimate of surface dose rates. However, there are some penetrating radiations which need to be considered, and the dose rates from such radiations are dependent on the amount of plutonium present in the source. The size of the source used for experimental measurements was limited to about 65 g of PuO_2 with a density of 1.61 g/cm^3 . Unless otherwise specified, the subsequent, calculated surface dose rates relate to this source size.

Hemispherical geometry was chosen because a simple analytical formula can be obtained to represent the dose rate from the entire source, while for cylindrical geometry, a double numerical integration is required.

The error in this approximation is believed to be quite small since the source is sufficiently large to approximate even an infinite slab. The surface flux, ϕ , is given by the equation

$$\phi = \frac{S_v}{2\mu r} [1 - e^{-\mu' r}] E_2(\mu t),$$

where S_v is the source strength per unit volume, μ' is the absorption coefficient for self-shielding, r is the radius of the hemisphere, μ is the absorption coefficient for the external shield, t is the thickness of the external shield, and the function E_2 is the exponential integral. This equation is for primary gamma radiation only, and secondary radiation (Compton scattered gammas, secondary X-rays, and photoelectrons) must be included by the use of buildup factors.

SHIELD COMPOSITIONS

Shield materials considered are Lucite, PVC (polyvinyl chloride), leaded gloves, steel, lead glass, and lead. Table 5-1 lists compositions and densities used. These are listed as the density of each constituent element in each of the shield materials. PVC is commonly used in plastic bags for transferring materials from glove boxes, and is similar in composition to Neoprene, which is used for glove-box gloves. The other materials were chosen because they are common structural or shielding materials used around shielded glove boxes.

Two types of leaded gloves were considered; a 30 mil glove containing four mils of lead equivalent, and a 60 mil glove containing 14 mils of lead equivalent. Both are

manufactured by the Charleston Rubber Company. The gloves comprise a sandwich of leaded Neoprene between two layers of plain Neoprene. The leaded Neoprene thickness is about half of the total glove thickness. For photon attenuation purposes, the gloves were assumed to be homogeneous, but for electron buildup calculations the outer one quarter-thickness of the glove was assumed to be plain Neoprene. Since only two standard glove thicknesses were considered, the dose rate is shown at only two shield thicknesses.

The composition of lead glass is only approximated with respect to elements of intermediate atomic number. For example, mass attenuation coefficients for oxygen are used instead of those for silicon, and iron is used instead of arsenic. This approximation was made to stay within the allowable number of shield materials in the program library and still cover all of the desired shield compositions in a single run. Errors in this approximation are minimal since attenuation by lead dominates in the energy region in which attenuation by the actual components differs markedly from the attenuation by the component used.

BUILDUP FACTORS

Buildup in the source is treated as being exclusively due to X-rays from photoelectric absorption, with the X-rays being treated the same as primary gamma rays from the source. Buildup in the external shielding is treated as a composite of a Compton buildup factor and an X-ray buildup factor. The Compton buildup factor is obtained from tables supplied to the program which uses three point interpolation on the logarithm of the buildup factor and the logarithm of the energy, followed by three point interpolation on the logarithm of the shield thickness in relaxation lengths.

TABLE 5-1. Composition of Shield Materials

Material	Density of Component in Shield Material, g/cm ³						Total
	Hydrogen	Carbon	Oxygen	Chlorine	Iron	Lead	
Lucite	0.0935	0.6958	0.3707	-	-	-	1.16
PVC	0.0629	0.4997	-	0.7374	-	-	1.30
Steel	-	-	-	-	7.86	-	7.86
Light Lead Glove	0.0711	0.6784	-	0.5005	-	1.47	2.72
Heavy Lead Glove	0.0711	0.6784	-	0.5005	-	2.63	3.88
Lead Glass	-	-	1.4700	-	0.07	4.66	6.20
Lead	-	-	-	-	-	11.20	11.20

The X-ray buildup factor is calculated from the formula

$$B = 1 + \sum \rho (\mu - \mu_c) S \omega Y \left(\frac{\mu - \mu_c}{\mu - \mu_x} \right) [e^{(\mu - \mu_x) t} - 1],$$

where: B is the buildup factor;

ρ is the density of the absorber;

μ , μ_c , and μ_x are the attenuation coefficients for the absorber, Compton scattering, and attenuation of the X-ray, respectively;

S is the fraction of the photoelectric cross-section for the particular shell;

Y is the yield of a particular X-ray for each photoelectric absorption in its parent shell;

ω is the fluorescence yield of the X-ray;

t is the absorber thickness;

and the summation is over a group of eleven X-rays (two from the K shell, seven from the three L shells, and a single X-ray group from the M and N shells).

The attenuation of electrons is much less straightforward than the attenuation of gammas because of the more irregular path of electrons and their greater number of collisions

As a first approximation, the ionization due to incident electrons was assumed to be linearly related to the thickness of absorber, decreasing to zero as the thickness of absorber approaches one-half the total path length of the electrons:

$$I = I_0 \left(1 - \frac{2x}{t}\right),$$

where I and I_0 are the attenuated and initial ionizations, x is the thickness of absorber, and t is the total path length (or range) of the electron.

The production of electrons in a thin layer of absorber is proportional to the photoelectric absorption coefficient $(\mu - \mu_c)$, the weight fraction (w) of the photoelectric absorber in the total absorber, the layer thickness (dx), and the photon flux (ϕ_γ):

$$\phi_\beta = (\mu - \mu_c) w \phi_\gamma dx$$

The electron intensity dI from a thin layer dx at distance x from the detector is

$$dI = k \left(1 - \frac{2x}{t}\right) dx,$$

which integrates to

$$I = kx \left(1 - \frac{x}{t}\right).$$

If $x = t/2$ (the maximum effective range of electrons), the electron intensity is $kt/4$. For a thick homogeneous absorber, the ratio of electron to gamma dose rate is

$$\frac{D_e}{D_\gamma} = \frac{(\mu - \mu_c) t w (-\frac{dE}{dx})}{4 E_\gamma (\mu/\rho)},$$

where $(-\frac{dE}{dx})$ is the electron energy lost per unit path length, E_γ is the gamma energy, and (μ/ρ) is the energy absorption coefficient for gamma rays of energy E_γ in air.

Gammas below about 80 keV produce conversion electrons which are totally absorbed by the 7 mg/cm² layer of dead skin, so that the only buildup from these gammas is due to photoelectric absorption and a small amount of Compton scattering.

For leaded gloves, a layer of about 7.5 mils of neoprene is outside the leaded neoprene layer, so that conversion electrons must have an energy of over 250 keV to penetrate from the leaded layer to the detector.

PROGRAM DESCRIPTION

PLJSHLD operates on a library of basic data and photon yields supplied by input cards, and calculates the surface dose rate from pure nuclides through external absorbers. The pure nuclide output is the dose rate from each photon energy transmitted through each shield, and total dose rate through each shield. Total dose rates for the pure nuclides are used to compute dose rates for mixtures of isotopes. The mixture output is the total dose rate for the mixture and the dose rate due to each component of the mixture. Output is given for up to 15 decay times to permit evaluation of the effects of parent decay and daughter growth.

Shielding data for up to seven elements are part of the input library. This includes mass attenuation coefficients at 25 energies, the energy of six X-ray absorption edges, energies of 11 X-rays, and fluorescent yield values for each X-ray. Densities of each element are specified for each shield material being considered. The shield material may be a single element or may contain up to seven components. A total of 100 shield materials can be used. The composition of the source is similarly specified.

For each shield material, a Compton buildup factor and an electron buildup factor need to be specified. Often the same buildup factor must be used for more than one shield composition since buildup factor data are rather limited. Only seven sets of data for Compton buildup and eight for electron buildup are allowed. Compton buildup factors consist of values at 10 energies and seven relaxation lengths for each material. Electron buildup factors consist of values for 25 energies.

Photon libraries are read for each nuclide being considered (up to 20), with up to 100 photons for each nuclide. The data are supplied as photon energy and photons per 100 disintegrations (i.e., corrected as necessary for internal conversion). Usually the X-rays from internal conversion are important and are included in the photon library in 11 X-ray groups, corrected for fluorescent yield. These X-rays arise in the daughter atom; alpha decay of plutonium results in uranium X-rays being formed.

Much of the radiation from plutonium isotopes occurs at low energies where photoelectric absorption is important and mass attenuation coefficients change rapidly with energy. Accurate values for the mass attenuation coefficients are required for good shielding calculations. Therefore, interpolation of mass attenuation coefficients was done by three point interpolation on the logarithms of both energy and mass attenuation coefficient. This results in much greater accuracy in the region of interest than does two-point linear or logarithmic interpolation.

Similar interpolation is performed for Compton buildup factors, where the interpolation is first on relaxation lengths and then on energy. Interpolation for electron buildup factors is by linear interpolation since values are less certain and vary only slowly with energy.

The program calculates a dose rate for the pure nuclide in a nonattenuating source with no external shielding, and then applies a self-shielding correction. X-rays generated by self-shielding are treated the same as photons from the source and are explicitly included in the list of photons from the pure nuclide. No other buildup of secondary radiation is considered within the source.

External shielding is included by the simple exponential attenuation law and corrected for Compton, X-ray and electron buildup. The first shield material is retained in place, and other shield materials are placed outside of this shield. Subsequent shields are removed, however, before inserting the next shield into its place. The first shield represents a small plastic window covering a detector, or the dead layer of skin on a person. Other shields are homogeneous, and no provisions are made for handling laminated shielding. Usually, four to eight compositions of shielding material (at 10 to 20 thicknesses each) are included in a single run.

Mixtures are accommodated by reading in abundance percentages of each isotope in the mixture. The total dose for a pure nuclide is weighted by the fractional abundance of the nuclide, and the weighted sum for all nuclides is the total dose rate for the mixture. Changes in composition with time are included by a decay matrix supplied as input. This matrix identifies the parent which results in the desired daughter at each time being considered. In addition to the initial composition, up to 14 periods of time can be included.

Typical running time is 3.5 min for 13 nuclides with about 30 photons each, 7 shield materials with a total of 70 different thicknesses, and 14 isotopic mixtures with 14 different aging times each.

6.0 RESULTS AND DISCUSSION

Surface gamma dose rates from plutonium oxide sources were calculated with a variety of shield materials and thickness. Photon attenuation by the shields and buildup of secondary radiations were included in the calculations, but the geometry effect due to thickness of the shield was not included. Dose rates were first calculated for pure nuclides in a plutonium oxide matrix, and these data for pure nuclides were used to calculate dose rates from plutonium with various isotopic compositions. The surface dose rate from plutonium

varies not only with plutonium isotopic composition, but also with time since separation from its daughter products, source size, chemical composition, and density. The effect of each of these variables on dose rate is discussed below.

The attenuation curves for pure nuclides (Figures 6-1 through 6-13) were derived on the basis of surface dose rates from a 65 g source of pure PuO_2 . External shielding comprised 7 mg/cm^2 of tissue, equivalent to the dead skin layer, along with variable amounts of other shielding. The curves are not directly applicable to other shielding situations (such as laminated shields), but can be used to derive semiquantitative data. For example, Figure 6-1 indicates a dose rate reduction of about a factor of 350 for a lightweight leaded glove. This attenuation factor is applicable only if other shielding is comparable to that used for the calculations. If the initial dose rate was determined through 10 mils of steel, the effect of adding a leaded glove would be much less than a factor of 350. The dose rate inside the glove would be only 3.8 rads through the steel, instead of 670 rads for an unshielded source. The dose rate outside the glove would be about 1.9 rads in either case. The overall attenuation is still a factor of 350, but only a factor of two is attributable to the leaded glove in the composite shield. If the shielding materials were reversed (steel placed outside the leaded glove), the dose rate through the glove would be 1.9 rads. Attenuation to this dose rate would require 72 mils of steel. An additional 10 mils of steel, or 82 mils effective total, would reduce the dose rate to only 1.8 rads. Thus, the attenuation due to 10 mils of steel can be as much as a factor of 180, or less than 10%, depending on other shielding between the source and the steel.

PURE NUCLIDES

Surface dose rates from pure nuclides of interest are plotted in Figures 6-1 through 6-13 for up to 350 mils shield

FIGURE 6-1. Attenuation of Dose Rate from Pure ^{238}Pu

FIGURE 6-2. Attenuation of Dose Rate from Pure ^{239}Pu

FIGURE 6-3. Attenuation of Dose Rate from Pure ^{240}Pu

FIGURE 6-4. Attenuation of Dose Rate from Pure ^{241}Pu

FIGURE 6-5. Attenuation of Dose Rate from Pure ^{242}Pu

FIGURE 6-6. Attenuation of Dose Rate from ^{237}U in Equilibrium with ^{241}Pu

FIGURE 6-7. Attenuation of Dose Rate from Pure ^{241}Am

FIGURE 6-8. Attenuation of Dose Rate from Pure ^{236}Pu

FIGURE 6-9. Attenuation of Dose Rate from Pure ^{232}U

FIGURE 6-10. Attenuation of Dose Rate from ^{228}Th in Equilibrium with ^{232}U

FIGURE 6-11. Attenuation of Dose Rate from ^{224}Ra in Equilibrium with ^{232}U

FIGURE 6-12. Attenuation of Dose Rate from ^{212}Pb in Equilibrium with ^{232}U

FIGURE 6-13. Attenuation of Dose Rate from ^{212}Bi in Equilibrium with ^{232}U

thickness. All of the data are presented as four-cycle semi-log plots to facilitate evaluation of the relative attenuation of radiations from each nuclide. The abscissa is the same for all graphs, but the ordinate is changed as appropriate. In accordance with the exponential attenuation law, a mono-energetic photon would be expected to yield a straight line attenuation curve on a semilog plot. Compton and X-ray buildup factors would cause a slight deviation from a straight line but no appreciable curvature. Electron buildup can cause an appreciable deviation from a straight line in the first few mils of a shield, but when the electrons escaping from the shield are characteristic of the outer shield material, they will no longer cause appreciable curvature. Curvature is caused by attenuation of the more easily shielded components; the transmitted photon spectrum becomes increasingly difficult to attenuate as shield thicknesses increase.

All of the nuclides of interest emit more than one photon energy of interest in shielding. Details of the photon abundances are discussed in a previous section. The principal characteristics of the individual nuclides are discussed below, along with some interpretations of the attenuation curves for the pure nuclides.

All of the curves show relatively little attenuation by Lucite. PVC is usually a much better shield than Lucite because photoelectric absorption by the chlorine in PVC is sufficient to attenuate low energy radiations such as L X-rays (about 17 keV for uranium and plutonium). Materials containing heavier elements are more effective for attenuating low energy radiations because of the increased photoelectric absorption. Their greater density also contributes to a better attenuation on a linear thickness basis. The high photoelectric cross-section of lead can cause a greater dependence on atomic number than on density. Lead glass is less dense than steel, but is usually a better attenuator. Even the

relatively low density lead glove is usually a better attenuator than steel, but there are exceptions.

^{238}Pu

The principal gamma transition in the decay of ^{238}Pu is about 44 keV, but because of internal conversion, few photons of this energy occur. Most of the photons are the uranium L X-rays resulting from this internal conversion. Another important photon occurs at 100 keV. Although low in abundance, it is relatively penetrating in the plutonium oxide source, since its energy is just below the K absorption edge where the mass attenuation coefficient is low. Higher energy radiations, in the neighborhood of 800 keV, are very low in abundance, but become the principal contributors to the dose rate from ^{238}Pu when the lower energy radiations have been shielded out.

The shielding curves in Figure 6-1 show the effects of the various shield materials on the ^{238}Pu radiations. The principal radiation through Lucite shielding in the thicknesses considered is the L X-ray group. The slight curvature is caused by this not being a monoenergetic photon but a group of photons. The attenuation of the L X-rays by PVC is shown by its curve for up to about 150 mils of shielding. Above this energy, the more penetrating 100 keV component becomes increasingly important. At 350 mils, the radiation comprises the 100 keV and 800 keV groups, with the 100 keV group still predominating. The L X-ray group is attenuated by about 10 mils of steel, and the 100 keV group predominates up to about 200 mils. At 350 mils, the radiation transmitted through steel is mostly the 800 keV group.

With lead-containing shields, the L X-rays are attenuated very rapidly. Only about 50 mils of lead or lead glass shielding is required to eliminate almost all of the low energy radiation, leaving only the 800 keV group.

^{239}Pu

The unshielded dose rate from pure ^{239}Pu (Figure 6-2) is about a factor of 500 less than that from ^{238}Pu . For each nuclide, the principal contributor to dose rate is the L X-ray group. Most of the difference is explained by a difference in half-life of a factor of 300, and decay scheme differences account for the rest. The Lucite shielding curve is quite similar for ^{238}Pu and ^{239}Pu , since in each nuclide it represents attenuation of the L X-ray group. The other curves are similar at low shield thicknesses, but deviate from the ^{238}Pu pattern after a reduction factor of about 10 in dose rate. The penetrating components from ^{239}Pu are at about 400 keV, as illustrated by the steeper attenuation curve for lead shielding over 50 mils thick. With a shielding thickness between 5 and 20 mils, steel is more effective than lead glass or even lead itself. Both the density and the mass attenuation coefficient of lead are higher than those for steel, and more photon attenuation is obtained with lead; however, the difference is in the electron buildup factor. In lead, photoelectric absorption accounts for most of the attenuation of 400 keV photons, as shown in Figure 3-2. With iron, however, the photoelectric effect is not large above 200 keV. Thus, the greater photon attenuation in lead is more than offset at low shield thicknesses by the increased electron buildup factor. Lead gloves are more effective shields than lead glass because the photoelectrons generated in lead gloves are attenuated by the Neoprene layer outside the leaded Neoprene.

 ^{240}Pu

The principal characteristics of the decay of ^{238}Pu and ^{240}Pu are very similar, as would be expected since they are both even numbered isotopes. The difference factor of 100 in unshielded dose rates (Figure 6-3) is almost all due to the

difference in half-life. Another difference is in the characteristics of the penetrating component. The penetrating component from ^{240}Pu (about 650 keV) is both less energetic and less abundant than that from ^{238}Pu . The unshielded dose rate from this component can be estimated by extrapolating the portion of the lead curve above 100 mils back to zero shield thickness. Also, the slope of the shielding curve for lead is characteristic of the energy of penetrating radiations.

^{241}Pu

The unshielded dose rate from ^{241}Pu (Figure 6-4) is similar to that from ^{239}Pu in spite of the difference in half-life of a factor of over 1000. Most of the ^{241}Pu decays are by beta emission to the ground state of ^{241}Am , so that no photon emission accompanies these decays. A small fraction (0.00235) of the ^{241}Pu decays are by alpha emission. The abundance of photons accompanying ^{241}Pu decay has not been established with any reasonable accuracy. For this study, the decay was assumed to occur only through 145 keV photons and the X-rays which are formed by internal conversion of this gamma. Most of the internal conversion takes place in the K shell, so that the X-rays have energies on the order of 100 keV. The shielding curves for Lucite and PVC are very similar, showing that chlorine does not have a sufficiently high atomic number to cause appreciable attenuation of photons of this energy by photoelectric absorption. The other curves do not show the abrupt decrease in dose rate with thin shields, but they show the absence of a penetrating component since they do not become relatively flat at high shield thicknesses.

^{242}Pu

The decay of ^{242}Pu is, as expected, similar to that of ^{238}Pu and ^{240}Pu . However, no penetrating components of its decay have been observed. The unshielded dose rate (Figure 6-5) is very much less than the lower plutonium isotopes because of the long half-life of ^{242}Pu . The curve for PVC shows the attenuation of the L X-ray group below about 150 mils, followed by a transition to the attenuation of the 45 keV gamma photon. The steep attenuation curves for steel, lead glass and lead reflect the large mass attenuation coefficient in these materials below 45 keV.

 ^{237}U

Thus far, we have been concerned with the principal plutonium isotopes. Two additional nuclides, ^{237}U and ^{241}Am , arise as decay products of ^{241}Pu . The half-life of ^{241}Pu is sufficiently short (13.2 yr) for appreciable decay to occur in times of interest in plutonium reprocessing. However, the ^{237}U becomes appreciable in much shorter times. Its 6.7 day half-life permits it to come to rapid equilibrium with ^{241}Pu . Since this equilibrium is rapidly achieved, the dose rate is presented for ^{237}U in equilibrium with pure ^{241}Pu (Figure 6-6) rather than the dose rate from pure ^{237}U . Adjustments were previously described for times before establishment of equilibrium.

All of the radiations from ^{237}U are rather penetrating in the range of 100 to 400 keV. Essentially no attenuation is obtained with either Lucite or PVC, which shows that the principal radiation is more penetrating than the L X-rays. The continuous curvature of shielding curves for the other materials shows the complex nature of the decay scheme. At deep penetrations (350 mils), the dose rate through steel arises

mostly from 200 keV gammas, while for lead glass or lead most of the dose rate is from 330 keV gammas.

^{241}Am

The predominant feature of ^{241}Am decay (Figure 6-7) is its 60 keV photon, although the dose rate from L X-rays is appreciable with light shielding. Lucite does not attenuate any of the radiation very effectively, while PVC is effective against the L X-rays only. Radiation more penetrating than the 60 keV gamma represents less than 1% of the unshielded dose rate. Thin lead shielding (about 40 mils) is sufficient to attenuate ^{241}Am radiation by over two orders of magnitude. The remaining radiation is largely over 0.5 MeV, and is attenuated slowly with increasing shield thickness.

The unshielded surface dose rate from ^{241}Am is quite high, even somewhat higher than from ^{238}Pu . The contribution of ^{241}Am to the total dose rate can be kept quite low, however, by processing plutonium soon after separation from americium.

^{236}Pu and Daughters

The ^{236}Pu daughters were treated as groups, and very short-lived nuclides were included with their parents (Figures 6-8 through 6-13). Thus ^{224}Ra actually represents a group consisting of ^{224}Ra , ^{220}Rn , and ^{216}Po . Likewise, ^{212}Bi includes ^{212}Po and ^{208}Tl also. Since the daughter with the longest half-life is ^{232}U , all nuclides below ^{232}U were calculated for amounts in equilibrium with ^{232}U . Since ^{236}Pu is normally present in no more than a few parts per million, very high dose rates from pure nuclides are required to yield an appreciable dose rate in actual plutonium. However, ^{208}Tl does have a high enough dose rate to be of significance.

Most of the dose rate ascribed to ^{212}Bi in Figure 6-13 is actually due to 3.1-min ^{208}Tl in equilibrium with it. A 2.61 MeV gamma accounts for most of the dose rate.

An unusual feature of the shielding curves for ^{212}Bi and some other daughters of ^{236}Pu is that Lucite can be a better shield than lead in some situations. This occurs because Lucite attenuates the electrons from the PuO_2 source without generating secondary electrons from the gammas passing through the shield. Lead will generate almost as many secondary electrons as the PuO_2 source, and thus attenuate the dose rate more slowly.

Growth of ^{208}Tl as a daughter of ^{236}Pu is rather slow because of the long half-lives of the first three chain members. After aging for a few years, the effect of ^{236}Pu daughters can be appreciable, especially through heavy shielding.

AS-PRODUCED PLUTONIUM

As long as the volume, density, and composition of the matrix remain constant, the dose rate from a nuclide is directly proportional to the amount present. Thus, a mixture of plutonium isotopes will yield a dose rate equal to the weighted sum of the dose rates from the individual isotopes, where the weighting factor is the fractional amount of each isotope in the mixture. Dose rates for various mixtures of plutonium isotopes were calculated in this manner.

Figures 6-14 to 6-19 show that dose rate increases with exposure for a given reactor and that different reactors yield different dose rates for the same exposure. For example, Shippingport yields a high relative dose rate in lightly-shielded situations, but is comparable to other reactors when large amounts of lead shielding are used. Isotopic compositions of plutonium samples used in this study are shown in Table 6-1.

FIGURE 6-14. Effect of Plutonium Composition on Dose Rate Through Lucite Shielding, Plutonium Aged 100 Days

FIGURE 6-15. Effect of Plutonium Composition on Dose Rate Through PVC Shielding, Plutonium Aged 100 Days

FIGURE 6-16. Effect of Plutonium Composition on Dose Rate Through Steel Shielding, Plutonium Aged 100 Days

FIGURE 6-17. Effect of Plutonium Composition on Dose Rate Through Lead Glove Shielding, Plutonium Aged 100 Days

FIGURE 6-18. Effect of Plutonium Composition on Surface Dose Rate Through Lead Glass Shielding, Plutonium Aged 100 Days

FIGURE 6-19. Effect of Plutonium Composition on Dose Rate Through Lead Shielding, Plutonium Aged 100 Days

*TABLE 6-1. Isotopic Composition of Plutonium Samples**

Sample Description	Isotopic Content, at. %					
	^{236}Pu	^{238}Pu	^{239}Pu	^{240}Pu	^{241}Pu	^{242}Pu
Yankee, 8,000 MWd/MTU	4.0×10^{-7}	0.144	87.0	9.56	3.06	0.208
Yankee, 13,000 MWd/MTU	1.0×10^{-6}	0.34	80.7	12.7	5.59	0.669
Yankee, 24,000 MWd/MTU	3.0×10^{-6}	1.00	67.7	18.9	10.0	2.51
Yankee, 38,000 MWd/MTU	8.0×10^{-6}	2.15	56.4	21.9	13.8	5.77
Dresden, 20,000 MWd/MTU	1.4×10^{-6}	0.403	69.55	21.46	6.862	1.719
Shippingport, 20,000 MWd/MTU	1.4×10^{-6}	1.06	51.2	28.9	11.0	7.78
Shippingport Sample S-61	9.2×10^{-7}	0.568	65.08	23.35	7.662	3.338
Composite Sample B-1	3.5×10^{-7}	0.0255	88.05	10.42	1.407	0.0968
Dresden Sample D-63	1.4×10^{-6}	0.403	69.55	21.46	6.862	1.719
Yankee Sample F-1	4.5×10^{-6}	1.92	63.3	19.2	11.68	3.88

* *The first six samples were used for calculations only, while the last four were used to compare calculations with experiments.*

The photon spectrum from plutonium is a function of both reactor type and total exposure. There is less variation in spectrum from exposure than there is from reactor type. Thus, shielding curves for Yankee plutonium at 8,000 and 38,000 MWd/MTU are roughly parallel over the entire range of shield compositions and thicknesses. Both Dresden and Shippingport show relatively softer spectra, especially with lead shielding. The spectrum of photons is not to be confused with total dose rate. The spectrum, which is a measure of the relative amounts of soft and hard components, determines how much dose rate reduction will be obtained with a given shield. The total dose rate is determined by the number of photons emitted as well as their spectrum or energy distribution.

Just as the shielding curves for a pure nuclide can be analyzed in terms of the component photon energies, so also the shielding curves for as-produced plutonium can be analyzed in terms of its component nuclides. An analysis of 38,000 MWd/MTU plutonium from Yankee reactor, aged 100 days, is shown in Figures 6-20 through 6-25 for various shield materials. Initially, the dose rate is due mostly to ^{238}Pu , but shields other than Lucite soon reduce ^{238}Pu to a minor contributor. Next, ^{241}Pu and its daughters become appreciable, but with heavy lead shielding these become less important than the more penetrating radiation from ^{239}Pu and ^{238}Pu .

The isotopic composition selected for these figures represents a "worst case" rather than an expected composition. The exposure is higher than normal, the reactor parameters favor production of unusually large amounts of ^{238}Pu and ^{236}Pu , and the samples were picked from specific portions of the reactor fuel core; i.e., the samples are not necessarily representative of what may be produced in a current Yankee Reactor.

EFFECT OF TIME

The dose rate contribution from all plutonium isotopes is virtually unchanging with time, but daughters of ^{241}Pu and ^{236}Pu cause an increase of dose rate with time. The discussion of as-produced plutonium was limited to material aged 100 days since chemical purification. At that age, the amount of ^{237}U is essentially at equilibrium, while the amounts of ^{241}Am and daughters of ^{236}Pu are increasing rapidly with time.

Figures 6-26 through 6-31 show the components of dose rate from plutonium aged two years since separation. When compared with Figures 6-20 through 6-25, these figures show the amount of ^{241}Am growth between 100 days and two years, which is about a tenfold increase. As expected, the ^{237}U contribution is essentially constant over this time period,

FIGURE 6-20. Components of Dose Rate Through Lucite Shielding, 38,000 MWd/MTU Yankee Plutonium, Aged 100 Days

FIGURE: 6-21. Components of Dose Rate Through PVC Shielding, 38,000 MWd/MTU Yankee Plutonium, Aged 100 Days

FIGURE 6-22. Components of Dose Rate Through Steel Shielding, 38,000 MWd/MTU Yankee Plutonium, Aged 100 Days

FIGURE 6-23. Components of Dose Rate Through Lead Glove Shielding, 38,000 MWd/MTU Yankee Plutonium, Aged 100 Days

FIGURE 6-24. Components of Dose Rate Through Lead Glass Shielding, 38,000 MWd/MTU Yankee Plutonium, Aged 100 Days

FIGURE 6-25. Components of Dose Rate Through Lead Shielding, 38,000 MWd/MTU Yankee Plutonium, Aged 100 Days

but is starting to decrease slowly. The curves for all plutonium isotopes in Figures 6-26 through 6-31 represent the minimum dose rate from plutonium of that isotopic composition--a situation that holds only immediately after chemical processing to remove uranium and americium. The small contribution from daughters of ^{236}Pu is also shown. When most of the radiation is shielded out (such as with 0.5 in. of lead), the effect of ^{236}Pu daughters is much more pronounced because of the high gamma energy.

Another representation of the effects of time is shown in Figures 6-32 through 6-37 for the selected shield materials. Changes below 100 days are largely due to ^{237}U growth, while beyond 100 days the increases are due to ^{241}Am and daughters of ^{236}Pu . The dose rate at 20 yr (relative to that for fresh material) is highly dependent on the type and amount of shielding. The increase is a factor of 6 for unshielded material where L X-rays predominate, a factor of 36 through polyvinyl chloride where 40 to 100 keV gammas predominate, and only a factor of 4 through steel shielding where somewhat harder gammas predominate. Through rather thick lead shielding, the dose rate increases greatly due to the hard components of both ^{241}Am and the ^{236}Pu daughters. It is evident that the effect of time on dose rate cannot be evaluated by simple rules of thumb, since it is dependent on shield types and thicknesses as well as plutonium isotopic composition and age since separation.

EFFECT OF SOURCE SIZE AND DENSITY

As previously mentioned, the dose rate from a hemisphere closely approximates the dose rate from a cylinder, or even an infinite slab, for the low-energy radiations. Since most of the radiation of concern in plutonium shielding is below 100 keV, this approximation is usually valid. As the low-energy radiations are attenuated, however, the remaining

FIGURE 6-26. Components of Dose Rate Through Lucite Shielding, 38,000 MWd/MTU Yankee Plutonium, Aged 2 Years

FIGURE 6-27. Components of Dose Rate Through PVC Shielding, 38,000 MWd/MTU Yankee Plutonium, Aged 2 Years

FIGURE 6-28. Components of Dose Rate Through Steel Shielding, 38,000 MWd/MTU Yankee Plutonium, Aged 2 Years

FIGURE 6-29. Components of Dose Rate Through Lead Glove Shielding, 38,000 MWd/MTU Yankee Plutonium, Aged 2 Years

FIGURE 6-30. Components of Dose Rate Through Lead Glass Shielding, 38,000 MWd/MTU Yankee Plutonium, Aged 2 Years

FIGURE 6-31. Components of Dose Rate Through Lead Shielding, 38,000 MWd/MTU Yankee Plutonium, Aged 2 Years

FIGURE 6-32. Effect of Aging on Dose Rate Through Lucite Shielding, 38,000 MWd/MTU Yankee Plutonium

FIGURE 6-33. Effect of Aging on Dose Rate Through PVC Shielding, 38,000 MWd/MTU Yankee Plutonium

FIGURE 6-34. Effect of Aging on Dose Rate Through Steel Shielding, 38,000 MWd/MTU Yankee Plutonium

FIGURE 6-35. Effect of Aging on Dose Rate Through Lead Glove Shielding, 38,000 MWd/MTU Yankee Plutonium

FIGURE 6-36. Effect of Aging on Dose Rate Through Lead Glass Shielding, 38,000 MWd/MTU Yankee Plutonium

FIGURE 6-37. Effect of Aging on Dose Rate Through Lead Shielding, 38,000 MWd/MTU Yankee Plutonium

radiations are sufficiently energetic to make both source weight and density important, as shown in Figure 6-38. For the selected geometry of this study, the dose rate is related to source size and density as follows:

$$D = k (1 - e^{-\mu \rho r}),$$

where μ is the mass attenuation coefficient, ρ is the source density, and r is the hemisphere radius. For soft radiations, this approaches $D = k$, while for highly penetrating radiations it is $D = K\mu\rho r$. The maximum increase expected with increased weight at constant density is the cube root of the weight, or a dose rate increase factor of 2.15 for a tenfold weight increase. If the weight is held constant and density increased, the dose rate increases with the two-thirds power of the density, or a factor of 3.38 increase on changing from 1.61 to 10.0 g/cm³.

The calculated increases are substantially less than these maximum values since self-shielding is appreciable for the energies which determine dose rates, even through 350 mils of lead shielding. As the dose rates are reduced by increasing shield thickness or efficiency, the changes in dose rates with changes in weight or density would more closely approach the theoretical limiting values.

The effect of weight and density on surface dose rate is determined largely by the inverse square law. As material is added to the outer surface of the hemisphere, the radiation has to travel appreciably farther to reach the dose point than does radiation emitted near the dose point. This increased distance results in a relatively smaller contribution to the dose rate per unit weight of added material. If the dose point were an appreciable distance from the source (perhaps one ft), the dose rate increase would be directly proportional to the weight increase if self-shielding were negligible. Under

FIGURE 6-38. Effect of Source Weight and Density on Dose Rate, 38,000 MWd/MTU Yankee Plutonium, Aged 2 Years

these conditions, the dose rate would no longer be dependent on the density. Thus, both weight and density affect the dose rate at the surface, while weight alone will affect the dose rate at a large distance from the source.

COMPARISON WITH EXPERIMENTS

Measurements of dose rates have been made at BNW⁽¹⁾ with several samples of plutonium and several different absorbers. The plutonium samples were enclosed in three polyvinyl chloride plastic bags, which totalled 34 mils in thickness. Two typical sets of these measurements are compared with calculations as shown in Figures 6-39 and 6-40. Agreement is generally better than 20%, with the main exception being at about 10 mils of lead shielding. From Figure 6-31, it appears that the discrepancy is due to either ^{241}Am or ^{237}U . One possibility is that the mass attenuation coefficient for the 60 keV gamma of ^{241}Am could be lower than the value used in the calculations. However, recent measurements of the mass attenuation coefficient of lead do not support this possibility.⁽¹⁰⁾ Other possible explanations include errors in the decay schemes of either ^{241}Am or ^{237}U . A more likely explanation is that the electron buildup factors are not correct in the energy region of 0.1 to 0.3 MeV. In this region, the electron buildup factors change from being characteristic of the counter window (7 mg/cm², equivalent to the dead skin layer) to being characteristic of the shield material. An error factor of two in the electron buildup factor at 0.2 MeV is possible, and this is the region of principal interest in the case of ^{237}U and 10 mils of lead shielding. Although this explanation is quite possible, it is not sufficiently proven to warrant changes in the calculations.

Agreement between measured and calculated values is only fair for steel with sample S-61 but good with sample B-1. The discrepancy with S-61 is usually 20 to 30%. In the region of

FIGURE 6-39. Comparison of Measured and Calculated Dose Rates for Sample S-61, Aged 180 Days

FIGURE 6-40. Comparison of Measured and Calculated Dose Rates for Sample B-1, Aged 3 Years

the discrepancy, ^{237}U is a major contributor to the dose rate for both S-61 and B-1, but the contribution of plutonium isotopes and ^{241}Am is more important for B-1 because of isotopic composition and age. An error in photon yields from ^{237}U or in the alpha branching ratio for ^{241}Pu might help to explain the difference. However, the difference could also result from a systematic error in that particular set of experiments. Since agreement with B-1 is good, no changes in the program input data appear warranted at this time.

Agreement for all materials is excellent in view of uncertainties in shield material composition, density and thickness, plutonium isotopic composition, and dose rate measurements.

The effect of age on surface dose rate has been measured for several plutonium samples, also enclosed in 34 mils of polyvinyl chloride. Comparisons with calculations are shown in Figures 6-41, 42 and 43 for typical samples. The initial set of measurements, shown in Figures 6-41 and 6-42, did not include enough short-aged measurements to show the growth of ^{237}U , although the measurements do show ^{241}Am growth. The samples were placed in new bags about 600 days after separation so that measurement geometry would be more uniform. Initial calculations with PUSHLD gave dose rates about 30% higher than the measurements. This disagreement led to a literature search for improved mass attenuation coefficients for plutonium. Indeed, better data⁽¹⁰⁾ were found, and these improved data resulted in the calculated values plotted in these figures.

Measurements shown in Figure 6-43 cover the time range over which ^{237}U comes into equilibrium with ^{241}Pu . All of the measurements are about 10% higher than the calculated values.

FIGURE 6-41. Surface Dose Rate from Shippingport Plutonium Oxide,
Sample S-61

FIGURE 6-42. Surface Dose Rate from Dresden Plutonium Oxide, Sample D-63

FIGURE 4-43 Surface Dose Rate from Yankes Plutoni Oxidized Sample F-1

The measurements are consistent, within limits of scatter, with a curve 10% higher than the calculated curve. However, they can also be consistent with a straight line or with other curves similar to the calculated curve: the scatter is great enough to make interpretation quite uncertain.

Good agreement between calculated and measured values was obtained for shielding curves and an "unshielded" surface dose rate at various times. This demonstrates that the basic features of plutonium dose rate and shielding calculations are understood well enough to permit a moderate reliance on calculations. More comparisons with good experimental data are required to determine the true accuracy and limitations of the calculations.

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