PLUTONIUM DIOXIDE GAMMA
ATTENUATION MEASUREMENTS
THROUGH STEEL

L. W. Brackenbush

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L.W. Brackenbush

Battelle Memorial Institute
Pacific Northwest Laboratory
Richland, Washington

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ABSTRACT

Photon dose-rates and energy spectra of two plutonium dioxide samples were measured, and attenuation measurements made with thin stainless steel absorbers. Relatively low exposure ($\sim 10\%$ ^{240}Pu) and high exposure ($\sim 23\%$ ^{240}Pu) plutonium samples were used. The surface dose-rates from the two samples measured through plastic bags were 1.7 rad/hr and 4.6 rad/hr; the dose-rates were reduced to 0.55 rad/hr and 1.55 rad/hr respectively by 0.020 inches of steel. Spectral measurements show 0.020 inches of steel eliminates most of the activity below 30 keV and appreciably attenuates the activity below 60 keV. Transmission ratios for a pure ^{241}Am source through steel absorbers are also presented.

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INTRODUCTION

A study is being conducted on the radiation characteristics of high-exposure plutonium, which includes measurements of both the neutron and electromagnetic radiations from plutonium and plutonium compounds. This is a preliminary report on dose-rate measurements and the attenuation of x-rays and gamma rays from plutonium dioxide provided by thin stainless steel absorbers. Additional measurements with thicker shielding and a variety of shielding materials will be forthcoming.

The dose-rates were measured with an extrapolation chamber containing a tissue-like plastic used to simulate the backscattering properties of tissue. Due to this backscattering material, the dose-rates will be somewhat higher than those measured with ordinary ionization chambers. The dose-rates are recorded in units of rad/hr and are absorbed dose-rates in air. They are a factor of 0.87 times smaller than exposure dose-rates in air, which are normally expressed in units of Roentgen/hr (1 rad = 1.15 R).

SUMMARY

The dose-rates and the energy spectra from the gamma and x-rays of two plutonium dioxide samples were measured with and without thin stainless steel absorbers. Relatively low exposure ($\sim 10\%$ ^{240}Pu) plutonium and high exposure ($\sim 23\%$ ^{240}Pu) plutonium samples were used. The surface dose-rates of the two oxide samples contained in plastic bags 0.033 inches thick were 1.7 rad/hr and 4.6 rad/hr; 0.020 inches of steel reduced the dose-rates to 0.55 rad/hr and 1.55 rad/hr respectively. The dose-rates through 5, 10, 15, and 20 mils of stainless steel absorbers were measured with an extrapolation chamber on the surface of the bagged plutonium dioxide and at one and two inches away from the surface.

Spectral measurements show 0.020 inches of steel eliminates most of the activity below 30 keV and appreciably attenuates the activity up to approximately 60 keV. Since most of the activity comes from low-energy photons, including those from ^{241}Am , transmission ratio measurements were made for a pure ^{241}Am source with 0.005 to 0.020 inch thick stainless steel absorbers.

DISCUSSION1. Description of PuO₂ Sources Used

Two samples of plutonium dioxide were used in this experiment. Sample A was obtained from irradiated Shippingport reactor fuel and was separated on February 25, 1967. The separation date for Sample B is not precisely known, but it is believed that this sample is 43 ± 3 months old based on the amount of ^{241}Am present. For the gamma-ray energy spectrum measurements, a small amount of plutonium was fixed on a stainless steel planchet; for the dose-rate measurements, about 60 grams of plutonium oxide were triple sealed inside three bags of polyvinyl chloride plastic, each 11 mils thick. The bag containing Sample A was about four and one-fourth inches square; the bag containing Sample B was about three inches by six inches. The measurements were made about 485 days after Sample A was separated. The isotopic compositions of the plutonium samples are listed in Table 1 below:

Table 1Isotopic Analysis of Plutonium by Weight Per Cent

	<u>^{238}Pu</u>	<u>^{239}Pu</u>	<u>^{240}Pu</u>	<u>^{241}Pu</u>	<u>^{242}Pu</u>
Sample A	.531	65.79	22.6	7.78	3.26
Sample B	.024	88.2	10.4	1.22	.097

2. Dose-Rate Measurements

The absorbed dose in air on the surface of a tissue-like plastic was measured with an extrapolation chamber. Due to backscattering, this

measured dose-rate is somewhat higher than dose-rate measurements made with free air ion chambers or "Cutie Pies." The extrapolation chamber used consists of a lucite disc four inches in diameter and about one inch thick used to simulate the backscattering properties of tissue. A threaded aluminum cylinder fits around the disc, and a thin membrane is stretched over the end. The 7 mg/cm^2 cellulose acetate membrane provides electron equilibrium for photons with energies up to about 70 keV, so the Bragg-Gray conditions are satisfied, and the extrapolated surface dose values obtained are valid for plutonium dioxide with its abundance of low-energy photons.

The surface dose-rate was measured by varying the chamber volume and determining the ionization current per unit volume of air for an infinitesimally thin volume on the surface of the source. Data were collected for six different plate separations, and a least squares linear fit of the data was made to determine the ionization current per cubic centimeter of air at standard temperature and pressure. This value was multiplied by the factor $1.04 \times 10^{-13} \text{ rad/hr per amp/cm}^3$ to obtain the surface dose-rate in air. The dose-rates for both triple bagged plutonium dioxide samples were measured through various thicknesses of stainless steel inserted between the sources and the chamber. The results of these measurements are presented in Table 2. The dose-rates at one inch and two inches from the surface of the plutonium dioxide are also included. Since the surface of the plutonium dioxide powder was relatively rough, the distances may be in error by as much as one-eighth inch.

To check the consistency of these results, the changes in dose-rates were checked by using 100 mil plate separation in the extrapolation chamber

Table 2

Dose-Rates from Plutonium Dioxide Samples with Various Steel Absorber Thicknesses

	PVC Plastic	Stainless Steel Absorber Thickness			
	<u>33 mil</u>	<u>5 mil</u>	<u>10.4 mil</u>	<u>15.1 mil</u>	<u>19.9 mil</u>
<u>Sample A</u>					
Surface Dose-Rate (rad/hr)	4.6	2.4	2.0	1.75	1.55
Photon Dose-Rate One Inch from Surface (rad/hr)	3.2	1.5	1.3	1.2	1.1
Photon Dose-Rate Two Inches from Surface (rad/hr)	1.9	0.82	0.70	0.65	0.56
<u>Sample B</u>					
Surface Dose-Rate (rad/hr)	1.7	0.87	0.70	0.62	0.55
Photon Dose-Rate One Inch from Surface (rad/hr)	1.3	0.58	0.49	0.44	0.39
Photon Dose-Rate Two Inches from Surface (rad/hr)	0.80	0.34	0.29	0.26	0.23

and inserting various steel absorber thicknesses between the plutonium dioxide bag and the extrapolation chamber. The results are shown in Table 3. The dose-rates in this table were determined by multiplying the surface dose-rate by the attenuation factor with various steel absorber thicknesses.

Table 3

Changes in Sample B PuO_2 Dose-Rates
with Stainless Steel Absorbers

	Stainless Steel Absorber Thickness						
	33 mil <u>PVC</u>	5 <u>mil</u>	10.4 <u>mil</u>	15.1 <u>mil</u>	19.9 <u>mil</u>	30 <u>mil</u>	50 <u>mil</u>
Dose-Rate (rad/hr)	1.7	.87	.71	.62	.55	.43	.29
Ratio relative to dose-rate with 33 mil PVC	1.00	.501	.410	.358	.314	.246	.164

Note that these values for Sample B are in very close agreement with the values listed in Table 2, which were obtained for Sample B by a least squares fit of several data points.

Using a linear extrapolation, the attenuation due to the plastic bags can be determined from a plot of dose-rate versus thickness of plastic. With a linear extrapolation, the surface dose-rates for the bare plutonium dioxide surface should be increased by a factor of 1.85 for Sample 1 (6.1 rad/hr) and 1.32 for Sample 2 (2.4 rad/hr).

The absolute accuracy of the extrapolation chamber measurements was checked by surface dose-rate measurements on a natural uranium standard. The value obtained (233 mrad/hr) was 3.8% higher than the accepted value of 225 mrad/hr, so it is assumed that the measured values for the dose-rates are within about 10% of the actual values.

Comparison checks were made with TLD-700 thermoluminescent dosimeters exposed with various thicknesses of stainless steel absorbers. A correction factor of 25% was used for the over-response of the TLD's relative to the radium gamma-ray calibration. This over-response correction factor becomes smaller with increased absorber thickness since the energy spectrum of the photons reaching the TLD's becomes harder. This is indicated by the increased difference between the dose-rates measured by the extrapolation chamber and the TLD's shown in Table 4.

Table 4

Comparison of TLD and Extrapolation Chamber
Measurements for Sample B

	<u>33 mil Plastic</u>	<u>5 mil Steel</u>	<u>10.4 mil Steel</u>	<u>15.1 mil Steel</u>	<u>19.9 mil Steel</u>
TLD Dose-Rates (rad/hr)	1.6	.66	.53	.47	.41
Extrapolation Chamber (rad/hr)	1.7	.87	.71	.62	.55

3. Photon Energy Spectrum Measurements

The gamma- and x-ray energy spectra were measured by lithium drifted germanium semiconductor detectors maintained at liquid nitrogen temperatures. The sources were placed on a shelf two inches above the detector and two and one-half inches square stainless steel absorbers of various thicknesses were inserted between the source and the detector. The data were stored in a 4096 multichannel analyzer, read out onto paper tape, and analyzed by the computer code DOT, which graphs the data and determines the photopeak area.

The energy spectra for plutonium oxide Samples B and A are shown in Figures 1 and 2 respectively. These data have not been corrected for the energy response of the detector. The relative efficiency graphs for detectors Ge-1 and Ge-2 (with the 59.5 keV photopeak of ^{241}Am normalized to unity) are shown in Figure 5. Most of the activity from both samples comes from the 59.5, 14.0, 17.8, and 20.8 keV photopeaks from ^{241}Am which has built in from the decay of ^{241}Pu . Very little activity is detected above the 208.4 keV photopeak from ^{237}U , except for a small peak at 662 keV which is thought to be due to ^{137}Cs contamination present in the counting room.

Figure 3 shows the spectral changes the absorbers produce. In Figure 3, the 33 mils of PVC bagging material reduces the 14.0, 17.8, and 20.8 keV photopeak heights, but has no discernible effect on the rest of the spectrum. The addition of 20 mils of stainless steel virtually eliminates all photopeaks below 30 keV and greatly reduces those below 60 keV.

Since much of the reduction in dose-rates with thin steel absorbers is a result of the attenuation of low energy photons from ^{241}Am , a series of attenuation measurements were made with a pure ^{241}Am source supplied by the International Atomic Energy Agency. This source was placed about two inches from a lithium drifted silicon detector (which has much better resolution than the germanium detectors previously used) and six-inch square stainless steel absorbers were placed in front of the source to assure broad beam geometry. The data obtained are believed to be accurate, and the reduction in the various photopeak heights (with the background subtracted) are given in Table 5. The transmission ratios obtained for the 59.5 keV photopeak agree within one or two percent of the values calculated by using a linear attenuation coefficient of 9.36 cm^{-1} for very thin stainless steel absorbers.

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Table 5

Relative Activity Through Various Stainless Steel Absorber Thicknesses with a ^{241}Am Source*

	Photopeak Energy					
	<u>11.2 keV</u>	<u>14.0 keV</u>	<u>17.8 keV</u>	<u>20.8 keV</u>	<u>26.4 keV</u>	<u>59.55 KeV</u>
No Absorber	1.00	1.00	1.00	1.00	1.00	1.00
33 mil PVC Plastic	.342 $\pm$ 7.9%	.385 $\pm$ 1.5%	.639 $\pm$ 1.8%	.761 $\pm$ 1.5%	.918 $\pm$ 1.8%	.984 $\pm$ 1%
5 mil Steel			.0315 $\pm$ 2.9%	.113 $\pm$ 3.2%	.324 $\pm$ 2.4%	.880 $\pm$ 1%
10.4 mil Steel				.0053 $\pm$ 22%	.103 $\pm$ 4.5%	.785 $\pm$ 1%
15.1 mil Steel					.042 $\pm$ 7.5%	.691 $\pm$ 1%
19.9 mil Steel						.613 $\pm$ 1%
50.4 mil Steel						.345 $\pm$ 1%
100.8 mil Steel						.105 $\pm$ 1%

* Calculated from photopeak heights corrected for background. The background was determined by a linear extrapolation between the background levels on either side of the photopeaks. The errors quoted are statistical counting errors and do not include errors which may arise from unresolved photopeaks.

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FIGURE 1
Photon Energy Spectrum from Sample B PuO₂

FIGURE 2
Photon Energy Spectrum from Shippingport PuO₂

FIGURE 3
Photon Energy Spectra from PuO_2 with Steel Absorbers

FIGURE 4
Relative Detector Efficiency

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