NEUTRON SPECTRA
OF PLUTONIUM COMPOUNDS
Part 1: $^3$He and $^6$Li
Spectrometer Measurements

L. W. Brackenbush
L. G. Faust
February 1970

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Part 1: $^3$He and $^6$Li Spectrometer Measurements

L. W. Brackenbush and L. G. Faust
Radiological Sciences Department
Environmental and Life Sciences Division

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The neutron energy spectra of compounds of high-exposure plutonium (23 percent $^{240}$Pu) are presented. The neutron spectrum of plutonium tetrafluoride shows a peak at 1.3 MeV, which agrees with previously published measurements; the neutron spectrum of plutonium dioxide is shown to have a very broad peak at about 2 MeV. A previously unreported spectrum of a plutonium-aluminum alloy (21 percent Pu - 79 percent Al) is presented with a peak at about 1.6 MeV. The average fluence to dose conversion factors calculated from the measured spectra are: Plutonium dioxide - $2.7 \times 10^{-9}$ rad/n/cm², plutonium tetrafluoride - $2.5 \times 10^{-9}$ rad/n/cm², plutonium-aluminum alloy - $2.6 \times 10^{-9}$ rad/n/cm², and plutonium metal (spontaneous fission spectrum) - $2.6 \times 10^{-9}$ rad/n/cm².

ABSTRACT

The neutron energy spectra of compounds of high-exposure plutonium (23 percent $^{240}$Pu) are presented. The neutron spectrum of plutonium tetrafluoride shows a peak at 1.3 MeV, which agrees with previously published measurements; the neutron spectrum of plutonium dioxide is shown to have a very broad peak at about 2 MeV. A previously unreported spectrum of a plutonium-aluminum alloy (21 percent Pu - 79 percent Al) is presented with a peak at about 1.6 MeV. The average fluence to dose conversion factors calculated from the measured spectra are: Plutonium dioxide - $2.7 \times 10^{-9}$ rad/n/cm², plutonium tetrafluoride - $2.5 \times 10^{-9}$ rad/n/cm², plutonium-aluminum alloy - $2.6 \times 10^{-9}$ rad/n/cm², and plutonium metal (spontaneous fission spectrum) - $2.6 \times 10^{-9}$ rad/n/cm².
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INTRODUCTION

To determine neutron dose rates and shielding requirements from plutonium compounds used in the manufacture and processing of reactor fuel elements, it is desirable to know the neutron energy spectra. Knowing the neutron spectrum and the neutron flux, it is possible to calculate neutron dose rates. Pure plutonium metal has a relatively low neutron emission rate arising from spontaneous fission. However, when plutonium comes into contact with material of low atomic number, the neutron emission may be increased by orders of magnitude. The neutron spectra from these $(\alpha,n)$ reactions with elements of low atomic number is quite different from the spontaneous fission spectrum. This paper presents the neutron energy spectra from $\text{Pu-F}(\alpha,n)$, $\text{Pu-O}(\alpha,n)$, and $\text{Pu-Al}(\alpha,n)$ reactions along with a list of factors to convert from neutron flux to neutron dose rate.

Accurate neutron spectra from plutonium compounds are quite difficult to obtain because of the relatively low neutron yields of small amounts of plutonium compounds and because most neutron spectrometers are very insensitive. Helium-3 and lithium-6 semiconductor spectrometers were used for these measurements because (1) they have very good resolution, (2) they are moderately sensitive (they have a sensitivity of $10^{-5}$ to $10^{-6}$ events per neutron/cm$^2$), (3) the electronic equipment required is fairly simple, and (4) the data do not require an elaborate unfolding technique as do proton recoil data. As will be explained, the $^3\text{He}$ spectrometer gives questionable results.
at lower neutron energies (below about 1 MeV) when a strong gamma field is present. A detailed explanation of the theory, operation and analysis of the data is given in the following sections.

SUMMARY

Neutron spectra and flux measurements are usually required for accurate neutron dose rate measurements. This report presents the results of neutron spectra measurements for compounds of high-exposure plutonium (about 23 percent $^{240}$Pu) produced by power reactors. A high exposure PuF$_4$ source gave a peak neutron energy of 1.3 MeV, which agrees with previous measurements; a PuO$_2$ source gave a very broad peak, which can be resolved into a spontaneous fission spectrum with a peak at about 0.7 MeV and an $^{180}$(α,n) spectrum with a peak at about 2.4 MeV. A 21 percent plutonium - 79 percent aluminum alloy gave a neutron spectrum with a peak at about 1.6 MeV. The average fluence to dose conversion factors for these sources are as follows: PuF$_4$ - $2.5 \times 10^{-9}$ rad/n/cm$^2$, Pu-Al alloy - $2.6 \times 10^{-9}$ rad/n/cm$^2$, PuO$_2$ - $2.7 \times 10^{-9}$ rad/n/cm$^2$, and pure plutonium metal (fission spectrum) - $2.6 \times 10^{-9}$ rad/n/cm$^2$.

THEORY AND OPERATION OF THE SPECTROMETERS

Theory of Operation of the $^3$He Neutron Energy Spectrometer

The $^3$He neutron detector consists of two carefully matched silicon surface barrier detectors separated by 1 mm and sealed into a double walled aluminum can into which $^3$He gas can be admitted.
Each surface barrier detector has a sensitive area of about 2 cm$^2$ and a depletion depth of 250 microns at 50 volts bias. The sensitive volume of the detector head is about 280 mm$^3$. The 1 mm separation results in some particle loss around the edges and some straggling and loss of resolution for high gas pressures (greater than 10 atm).

Fast neutrons interact with the $^3$He gas in the detector head by the following reaction:

$$ ^2\text{He}^3 + \text{n}^1 \rightarrow ^1\text{H}^3 + ^1\text{H}^1 + 0.764 \text{ MeV} $$

Ignoring small energy losses due to straggling, the energy of the incident neutron is equal to the energy deposited in the surface barrier detectors by the triton and proton minus the Q value of the reaction. The relative intensity of the neutron spectrum is determined by correcting for the change in neutron cross section with energy.

The upper limit of usefulness of this device is about 5 to 6 MeV due to (1) competition of $^3$He(n,d)$^3$He reactions for energies above 4.4 MeV, (2) the range of reaction products from higher energy neutrons requires a greater depletion depth in silicon, and (3) large depths of silicon result in competition from neutron induced reactions in the silicon. Fortunately, the neutron spectra of plutonium compounds contain very few neutrons above 5 MeV. Overall resolution of the spectrometer depends upon energy, and the peak width from thermal neutrons is less than 100 keV (FWHM). The efficiency of this device is quite low - about $10^{-5}$ to $10^{-6}$ events per neutron per square centimeter.
A fast coincidence system is used in an attempt to record only the triton and proton from a neutron reaction with $^3$He and to exclude gamma and other neutron interactions. Unfortunately, the surface barrier detectors are also quite sensitive to gamma rays and to neutron induced events in the silicon. To minimize the "background" from these extraneous sources, it is necessary to have a very fast coincidence system.

The fast coincidence system used with the $^3$He detector head is shown in Figure 1. Care must be exercised in the selection of the charge sensitive preamplifiers, for the large input capacitance of the surface barrier detectors can create serious electronic noise problems. The linear amplifiers are operated with 1.8 microsecond time constants for doubly differentiated pulses to satisfy the multichannel analyzer input signal requirements. The single channel analyzer discriminators are set to trigger only on pulses corresponding to protons and tritons from thermal neutron reactions and are operated in the crossover pick-off mode to minimize jitter. The coincidence unit has a 100 nanosecond (2 tau) resolution time. Smaller resolution times would be desirable to reduce the random coincidence from gamma and Si(n,p) reactions, but the slight jitter introduced by electronic noise makes it necessary to have relatively large resolution times.

**Calibration of the $^3$He Spectrometer**

The spectrometer was calibrated by using nearly monoenergetic neutrons produced by T(p,n) and D(d,n) reactions from a 2 MeV
Van de Graaff accelerator. These reactions are comparatively free of strong gamma ray production which might interfere with the energy calibration. Since the efficiency of the detector is very low (about $10^{-5}$ to $10^{-6}$ events per n.cm$^2$), thick targets were used to produce as many neutrons as possible to improve counting statistics. The resulting neutron peaks observed were quite broad, so that the leading edges were used for calibration purposes in some instances. The energy calibration is shown in Figure 2 for the $^3$He detector oriented with the surface barrier detectors parallel to the neutron beam. The non-linear region below about 0.3 MeV is believed to be due to differences in the forward scatter of the triton and proton from fast neutron reactions and due to differences in the efficiency of the surface barrier detectors with a thin layer of gold on their surfaces. A precision pulse generator was used to provide a secondary calibration of the spectrometer to ascertain whether electronic gain shifts occurred. The spectrometer was recalibrated several times, and no significant changes were noted over three-month periods.

The $^3$He detector head exhibits some angular response. For exposures made with 1.0 MeV neutrons, the separation between the thermal neutron peak and 1 MeV neutron peak increased as the $^3$He detector head was rotated from a position with the surface barrier detectors parallel to the neutron beam to a position with the surface barrier detectors perpendicular to the neutron beam. The separation between the peaks increased about 5 percent when the detector was positioned at 45°; and it increased about 12 percent when the detector was placed at 90° "perpendicularly" to the neutron beam.
The neutron peaks sometimes contained only about 100 counts per ten channels, so that the counting statistics are relatively poor.

The $^3$He spectrometer also exhibits pulse pile-up problems in strong gamma fields. Exposures made with a $^{12}$C(d,n) reaction producing 1.48 MeV neutrons gave unusual results -- the neutron peak occurred in a channel corresponding to 1.7 MeV neutrons. The gamma dose rate was not measured while the accelerator was operating, but after the accelerator was shut down, the residual target dose rate was about 100 mR/hr. With these kinds of dose rates, it is quite probable that some summing occurred.

Experimental Procedures for Operation and Data Analysis of the $^3$He Spectrometer

To acquire neutron spectral data, the $^3$He detector head is filled with 80 to 100 psia of $^3$He gas which has been purified to remove tritium (purity of 1 part $^3$H per $10^{11}$ parts $^3$He) and "gross" or "foreground" exposure is made. The $^3$He is then removed and a "background" count is made to compensate for events from gamma, Si(n,p), and other interactions. A fit of the data is made and the "background" subtracted. The resultant data are then corrected for the changes in $^3$He cross section with energy taken from Figure 3.\(^{(1)}\)

To obtain accurate spectra, it was necessary to isolate the spectrometer and to construct a jig to shield and position the $^3$He detector head for accurate "background" counts. The jig consists of a one-inch lead shield to reduce the gamma background with a 1/8-inch tin shield to reduce any lead X-rays present. The $^3$He detector
FIGURE 3

Neutron Cross Sections for $^3$He (n,p) and $^6$Li (n,α) Reactions

Neutron Energy in MeV

Neutron Cross Section in Barns
head fits inside an aluminum cylinder to maintain the same geometry for "background" runs.

Figure 4 shows a semilogarithmic plot of the data obtained by positioning the $^3\text{He}$ spectrometer head in the lead shielded jig and exposing it for a two-week period to a plutonium fluoride sample three inches in diameter by three inches high containing 539 grams of high exposure plutonium (22 percent $^{240}\text{Pu}$). The "gross" measurement was made with 100 psia of $^3\text{He}$ gas filling; the "background" measurement was made with the detector head evacuated. Note the great number of "background" events at lower energies due to random coincidences of gamma rays and $\text{Si}(n,p)$ reactions in the semiconductor detectors. Subtracting the fitted values of the "background" exposure from the fitted values of the "gross" exposure results in very large statistical errors at lower neutron energies. Figure 5 shows the "background" corrected curve obtained by subtracting fitted values. The points with error bars are the results obtained by using the actual "gross" values rather than fitted values. To obtain the neutron energy spectrum, it is necessary to correct for the changes in the $^3\text{He}(n,p)^3\text{T}$ cross section with energy as shown in Figure 3. The solid line in Figure 5 is obtained by multiplying the "background" corrected curve (shown as a dashed line) by the inverted cross section for the $^3\text{He}(n,p)$ reaction normalized to a value of 1.00 for the peak occurring at 1.7 MeV in the $^3\text{He}(n,p)$ cross section.
FIGURE 4
Semilogarithmic Plot of "Foreground" and "Background" Data for the $^3$He Spectrometer Exposed to a PuF$_4$ Source
Theory and Operation of the $^6$Li Neutron Energy Spectrometer

The $^6$Li neutron detector consists of two silicon surface barrier diodes placed face to face with a thin film of $^6$LiF evaporated onto the face of one of the diodes. Neutrons interact with the $^6$Li according to the reaction:

$$^3\text{Li}^6 + \text{n}^1 \rightarrow ^2\text{He}^4 + ^1\text{H}^3 + 4.78 \text{ MeV}$$

As in the case of the $^6$He spectrometer, the neutron energy is determined by measuring the energy of the reaction products. The $^6$Li(n,α) reaction has such a high Q value that there are few "background" events from gamma and neutron induced events in the silicon. Hence, a relatively slow coincidence system can be used. The sensitivity of the $^6$Li detector is less than that of the $^3$He detector. If the thickness of the $^6$LiF layer is increased to increase the neutron sensitivity, the resolution of the detector deteriorates. For a 150 microgram/square centimeter thick layer of $^6$LiF, the width of the peak resulting from slow neutrons is typically 300 keV (FWHM).

The $^6$Li spectrometer has a useful operating range from 0.5 MeV to about 10 MeV due to competing nuclear reactions. The most important competing reaction is the $^6$Li(n,α) d reaction, which has a threshold energy of 1.72 MeV. On the average, the energy of the outgoing neutron is around 1 to 2 MeV, so that the energy difference between the $^6$Li(n,α)d reaction and $^6$Li(n,α)$^3$H reactions is about 8 MeV. For neutron spectra with a large number of neutrons above about 5 MeV, the spectra below 1 or 2 MeV may be in error.\(^2\)
The $^6$Li spectrometer was also calibrated by exposure to monoenergetic neutrons produced by a Van de Graaff accelerator. However, the results are more uncertain because of the lower sensitivity of the $^6$Li spectrometer and because of electronic gain shift problems.

The data analysis of the $^6$Li spectrometer is similar to that of the $^3$He spectrometer. The "background" is much lower because of the high Q value of the $^3$Li(n,a) reaction, so that the $^6$Li spectrometer results are probably more accurate at lower neutron energies close to 1 MeV. The data must be corrected for changes in the cross section with energy as shown in Figure 3. Since the $^6$Li spectrometer has poorer resolution, one must be careful to subtract out the thermal neutron peak before applying cross section corrections to the data. This can be done by plotting the thermal peak on a semilogarithmic graph and linearly extrapolating the edge of the peak to higher neutron energies.

RESULTS

The experimentally measured neutron energy spectra from various plutonium compounds are shown in Figures 6 through 10. The relative neutron intensity (with dimensions of neutrons) is plotted against the neutron energy (with dimensions of MeV). The area under the curves in each of these figures is normalized to a value of 100 neutron-MeV (i.e., the curves are distribution functions which have ordinate values in percent).
The plutonium compounds used for these measurements were in the form of loose powders, which were sealed inside of two thin walled steel cans. A summary of the plutonium sources used is shown in Table 1. The results of these measurements are summarized in Table 2, and a summary of published spectral data is included in Table 3 for comparison.

Figure 6 shows the neutron spectra obtained from a $^6$Li neutron spectrometer exposed to a large, low exposure (6 percent $^{240}$Pu) plutonium tetrafluoride source (source A in Table 1). The area under each curve is the same, so that the effects of the one-inch Lucite moderator are apparent. The moderator increases the peak width, removes a few higher energy neutrons, and creates a few lower energy neutrons. The peak occurs at 1.1 MeV, which is somewhat lower than other measurements indicate (see Table 3). The energy calibration for these measurements is somewhat uncertain.

Figure 7 shows the neutron spectrum obtained from a $^3$He neutron spectrometer with a high exposure (22 percent $^{240}$Pu) plutonium tetrafluoride source, designated as source B in Table 1. Note that the energy peak at 1.3 MeV is in excellent agreement with previously published data obtained from nuclear emulsions and stilbene crystals (proton recoil) shown in Table 3.

The spectrum from a high exposure (22.7 percent $^{240}$Pu) plutonium dioxide source, designated as source C, is presented in Figure 8. The oxide contains naturally occurring oxygen; no attempt was made to change the neutron yield from $^{180}$(a,n) by isotopic enrichment. The spectrum below about 1.5 MeV is uncertain because of competition from
### TABLE 1

**NEUTRON SOURCE DATA**

<table>
<thead>
<tr>
<th>Source Identification</th>
<th>Mass Plutonium</th>
<th>Isotopic Composition in Weight Percent</th>
<th>Size of Source</th>
<th>Yield Neutrons/sec</th>
</tr>
</thead>
<tbody>
<tr>
<td>A PuF₄</td>
<td>765 g</td>
<td>2x10⁻⁵ 93 6 0.5</td>
<td>3-1/2&quot; dia. x 4-1/8&quot;</td>
<td>5 x 10⁶</td>
</tr>
<tr>
<td>B PuF₄</td>
<td>518 g</td>
<td>0.090 72.8 22.3 3.81 0.99</td>
<td>3&quot; dia. x 3&quot;</td>
<td>6.4 x 10⁶</td>
</tr>
<tr>
<td>C PuO₂</td>
<td>725 g</td>
<td>0.072 72.4 22.7 3.89 0.97</td>
<td>3-1/4&quot; dia. x 2-3/4&quot;</td>
<td>2.3 x 10⁵</td>
</tr>
<tr>
<td>D Pu-Al</td>
<td>1.25 kg</td>
<td>0.561 65.25 23.68 7.10 3.39</td>
<td>2-1/2&quot; x 24&quot; x 4&quot;</td>
<td>4 x 10⁵</td>
</tr>
<tr>
<td>Source</td>
<td>Identification</td>
<td>Spectrometer</td>
<td>Shield</td>
<td>Peak* Energy</td>
</tr>
<tr>
<td>---------</td>
<td>----------------</td>
<td>--------------</td>
<td>-------------------------</td>
<td>--------------</td>
</tr>
<tr>
<td>PuF₄</td>
<td>A 765 gram</td>
<td>⁶Li</td>
<td>No shield</td>
<td>1.10 MeV</td>
</tr>
<tr>
<td>PuF₄</td>
<td>A 765 gram</td>
<td>⁶Li</td>
<td>1&quot; Lucite</td>
<td>1.10 MeV</td>
</tr>
<tr>
<td>PuF₄</td>
<td>B 518 gram</td>
<td>³He</td>
<td>1&quot; lead and 1/8&quot; tin</td>
<td>1.33 MeV</td>
</tr>
<tr>
<td>PuO₂</td>
<td>C 725 gram</td>
<td>³He</td>
<td>1&quot; lead and 1/8&quot; tin</td>
<td>2.2 MeV</td>
</tr>
<tr>
<td>Pu-Al alloy</td>
<td>D 1.25 kg</td>
<td>³He</td>
<td>1&quot; lead and 1/8&quot; tin</td>
<td>1.60 MeV</td>
</tr>
</tbody>
</table>

*Note: The numbers given contain only two significant figures.
### TABLE 3

**SUMMARY OF PUBLISHED NEUTRON SPECTRA**

<table>
<thead>
<tr>
<th>Material or a,n Reaction</th>
<th>Peak* Neutron Energy</th>
<th>Average* Neutron Energy</th>
<th>Spectrometer</th>
<th>Comments</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{239}$PuF($\alpha$,n)</td>
<td>0.87 MeV</td>
<td></td>
<td>$^6$Li- 200 keV resolution</td>
<td>1 kg source with some neutron scatter</td>
<td>G.W.R. Endres. Neutron Spectra Measurements at Hanford Work Locations, HW-SA-3525, General Electric Co. 1964.</td>
</tr>
<tr>
<td>$^{238}$PuF($\alpha$,n)</td>
<td>1.2 MeV</td>
<td></td>
<td>$^6$Li and $^3$He</td>
<td>Calibrated by PuBe and $^{252}$Cf</td>
<td>T.R. Herold. Nuclear Instr. &amp; Methods, v. 71, p. 40. 1969.</td>
</tr>
<tr>
<td>$^{238}$PuF($\alpha$,n)</td>
<td>1.35 MeV</td>
<td>1.44 MeV</td>
<td>Stilbene(proton recoil)</td>
<td>Very few neutrons above 3.2 MeV Predicted maximum</td>
<td>M.E. Anderson. Transactions American Nuclear Soc., v. 9, p. 600. 1960.</td>
</tr>
<tr>
<td>$^{238}$PuO($\alpha$,n)</td>
<td>2.6 MeV</td>
<td>2.4 MeV</td>
<td>Stilbene(proton recoil)</td>
<td>Low energy tail below 1.5 MeV</td>
<td>Anderson, ibid.</td>
</tr>
<tr>
<td>$^{238}$PuO$_2$</td>
<td>2.3 MeV</td>
<td></td>
<td>$^6$Li - 275 keV resolution</td>
<td>Calibrated by changing gain, PuBe, and $^{252}$Cf</td>
<td>T.R. Herold. Nuclear Applications v. 4, p. 19. Feb. 1968.</td>
</tr>
</tbody>
</table>

*Note: Numbers listed are accurate to two significant figures only.*
FIGURE 6
Neutron Spectrum from $^6$Li Spectrometer Exposed to PuF$_4$
FIGURE 7
Neutron Spectrum from PuF₄
FIGURE 8
Neutron Spectrum from PuO$_2$
$^3$He(n,d)D and other reactions and from large statistical errors from "background" events. A spectrum presented by Anderson$^4$ for 238PuO$_2$ shows the peak occurring at 2.6 MeV, and data published by Herold$^5$ shows the peak at 2.3 MeV. The results of the $^3$He spectrometer measurement show a very broad peak at about 2.2 MeV for the high exposure plutonium spectrum in Figure 8.

It is assumed that the spontaneous fission spectrum of 240Pu and 238Pu is very similar to the Watt fission spectrum for 235U fission induced by thermal neutrons. Terrell$^6$ concludes that all experimentally measured fission spectra (235U + n, 239Pu + n, and 252Cf) are Maxwellian distributions, and that the average energy of the distribution is related to $\nu$, the average number of neutrons emitted per fission, by the formula $\bar{E} = 0.74 + 0.653 (\nu + 1)^{1/2}$ where the energy is in MeV. Data compiled by Faust$^7$ show that the values of $\nu$, and hence the average energy of the fission spectrum, are quite similar for the thermal neutron fission of 235U ($\nu = 2.46$, $E = 1.95$ MeV), and spontaneous fission of 238Pu ($\nu = 2.33$, $E = 1.93$ MeV), and spontaneous fission of 240Pu ($\nu = 2.26$, $E = 1.92$ MeV). Spontaneous fission of 238Pu and 240Pu account for the majority of the neutrons generated by pure plutonium metal sources. Table 4 shows Faust's data for the neutron yields from spontaneous fission and (a,n) reactions for various plutonium isotopes and compounds. These data represent the best estimates available several years ago and may be revised by more recent measurements.

Figure 9 shows a calculated plutonium dioxide spectrum generated from a Watt fission spectrum$^8$ and an $^8$O(a,n) spectrum.$^9$ The $^16$O(a,n) spectrum shown is from a $^{210}$Po-a-0 source with a peak neutron
<table>
<thead>
<tr>
<th>Isotope</th>
<th>Spontaneous Fission</th>
<th>Reaction Rate</th>
<th>Yield</th>
<th>Nuclide</th>
<th>(α,n) Reaction Rate</th>
</tr>
</thead>
<tbody>
<tr>
<td>236Pu</td>
<td>3.5 × 10^3 y</td>
<td>2.3</td>
<td>3.7 x 10^4</td>
<td></td>
<td></td>
</tr>
<tr>
<td>238Pu</td>
<td>4.9 × 10^10 y</td>
<td>2.33</td>
<td>2.62 x 10^3</td>
<td></td>
<td></td>
</tr>
<tr>
<td>238PuF_4</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>2.1 x 10^6</td>
</tr>
<tr>
<td>238PuO_2</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>1.4 x 10^4</td>
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<tr>
<td>239Pu</td>
<td>5.5 × 10^15 y</td>
<td>3.0</td>
<td>3 x 10^{-2}</td>
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<tr>
<td>239PuO_2</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>4.5 x 10^1</td>
</tr>
<tr>
<td>240Pu</td>
<td>1.22 × 10^{11} y</td>
<td>2.25</td>
<td>1.02 x 10^3</td>
<td></td>
<td></td>
</tr>
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<td></td>
<td></td>
<td></td>
<td>1.6 x 10^4</td>
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<td>1.7 x 10^2</td>
</tr>
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<td>2.18</td>
<td>1.7 x 10^3</td>
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<tr>
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<td>2.3*</td>
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<td>2.6 x 10^3</td>
</tr>
</tbody>
</table>

*Estimated

---

Figure 9
Calculated Neutron Spectrum from PuO₂
energy of 2.4 MeV. Neutron yield calculations, which are based upon
the isotopic composition of the sample (source C in Table 1) and the
neutron yield data in Table 4, show that about one-fourth of the
neutron yield is generated by $^{18}$O(a,n) reactions, and about three-
fourths of the neutron yield comes from spontaneous fission of the
plutonium. The area under each curve in Figure 9 is proportional to
the neutron yield. Curve C is composed of the sum of one-fourth of
the distribution function for the $^{18}$O(a,n) spectrum and three-fourths
of the distribution function for the spontaneous fission spectrum.
Note that this calculated spectrum is quite similar to the experi-
mentally measured spectrum in Figure 8.

Figure 10 shows the neutron spectrum from a 21 percent plutonium-
79 percent aluminum alloy. The isotopic composition of the plutonium
(designated as source D) is given in Table 1. Neutron yield measure-
ments show the neutron yield from this sample to be approximately 13
times higher than that arising from spontaneous fission alone. Pres-
umably, most of the neutrons are generated from Al(a,n) reactions
since the sample contains very small amounts of other light elements.
The spectrum below about 1 MeV is somewhat uncertain because of
large statistical errors, and the energy calibration may be slightly
in error. The Pu-Al source used is quite large, and the angular
response characteristics of the $^3$He detector may cause small errors
in the energy calibration for neutrons entering the detector nearly
perpendicular to the surface barrier detectors.
Neutron Spectrum from Pu-Al Alloy

Neutron Energy in MeV

Relative Intensity
CONCLUSIONS

The results obtained with the $^3$He spectrometer from high exposure plutonium compounds (as shown in Table 2) are in general agreement with the few previously published spectra from a variety of different sources (as shown in Table 3). The $^3$He spectrometer results have sharper neutron peaks, probably because of the better energy resolution of about 100 keV. The $^3$He spectrometer results are not very accurate below about 1 MeV due to large statistical errors arising from neutron and gamma ray interactions with the silicon in the surface barrier detectors. These low energy spectra will be measured at a later date with proton recoil or $^3$He proportional counter spectrometers. The relative accuracy of the spectral measurements is indicated by the error bars shown in Figure 5. Notice that these errors are only statistical counting errors and do not include errors in the cross section corrections. The plutonium dioxide spectrum has slightly larger errors because of the lower neutron yield.

Plutonium dioxide spectra vary from one sample to another depending upon the isotopic composition of the sample. Probably the most accurate plutonium dioxide spectra can be calculated from the sum of a spontaneous fission spectrum and an $^{180}$O(a,n) spectrum (10) weighted by the appropriate amounts determined from neutron yield calculations from Table 4 for spontaneous fission and (a,n) reactions from plutonium isotopes. Note that the neutron spectra presented in Figure 8 and 9 are typical of high exposure plutonium containing large amounts (about 23 percent) $^{241}$Pu and relatively
small amounts (about 0.07 percent) $^{238}\text{Pu}$; these spectra differ from $^{238}\text{PuO}_2$ spectra because of the much higher $0(a,\text{n})$ yield for $^{238}\text{Pu}$.

The average fluence to dose conversion factors for each neutron spectrum are presented in Table 5. The doses calculated from these factors are the so-called "first collision doses" (actually KERMA) for a small piece of tissue exposed to a unit fluence of one neutron/cm$^2$. These conversion factors are calculated by using 200 keV increments and the data contained in National Bureau of Standards Handbook 63\cite{11} according to the formula:

$$\bar{R} = \frac{\Sigma R(E) N(E)}{\Sigma N(E)}$$

where $R(E)$ is the rad-cm$^2$/neutron conversion factor and $N(E)$ is the relative neutron intensity. Thus, the neutron dose rates from plutonium compounds can be determined by measuring the neutron flux (dimensions of neutron-cm$^2$/sec), or by calculating the flux for nearly point sources, and multiplying by the fluence to dose conversion factors (dimensions of rad-cm$^2$/neutron) listed in Table 5.

The fluence to dose conversion factors for plutonium dioxide with different isotopic compositions can be calculated by the following equation:

$$\bar{R} = x R_{sf} + (1-x) R_{a,n}$$

where $\bar{R}$ is the average fluence to dose (or dose equivalent) conversion factor.

$x$ is the fraction of the neutron yield from spontaneous fission,
<table>
<thead>
<tr>
<th>Source</th>
<th>Source Identification</th>
<th>Average Energy (MeV)</th>
<th>Fluence to Dose (rad/n/cm²)</th>
<th>Fluence to Dose Equivalent (rem/n/cm²)</th>
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</thead>
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<tr>
<td>PuF₄</td>
<td>A</td>
<td>1.3</td>
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<tr>
<td>PuF₄</td>
<td>B</td>
<td>1.4</td>
<td>$2.53 \times 10^{-9}$</td>
<td>$2.5 \times 10^{-8}$</td>
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<td>PuO₂</td>
<td>C</td>
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<td>$2.74 \times 10^{-9}$</td>
<td>$2.5 \times 10^{-8}$</td>
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<td>PuAl alloy</td>
<td>D</td>
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<td>$2.63 \times 10^{-9}$</td>
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<tr>
<td>Pu metal</td>
<td>(fission spectrum)</td>
<td>1.9</td>
<td>$2.60 \times 10^{-9}$</td>
<td>$2.5 \times 10^{-8}$</td>
</tr>
</tbody>
</table>

(a) See Table 2 for detailed description of the sources.

(b) First collision doses calculated from data on page 7 of National Bureau of Standards Handbook 63.
l-x is the fraction of the neutron yield from $0(\alpha,n)$ reactions,

$R_{sf}$ is the average fluence to dose (or dose equivalent) conversion factor for spontaneous fission ($2.60 \times 10^{-9} \text{ rad-cm}^2/\text{n}
\text{ or } 2.46 \times 10^{-8} \text{ rem-cm}^2/\text{n}$),

$R_{\alpha,n}$ is the average fluence to dose (or dose equivalent) conversion factor for $0(\alpha,n)$ reactions for a $^{210}\text{Po}$ alpha source ($3.14 \times 10^{-9} \text{ rad-cm}^2/\text{n} \text{ or } 2.67 \times 10^{-8} \text{ rem-cm}^2/\text{n}$).

For small masses of material (small in the sense that the neutron energy spectrum is not degraded by a significant number of scattered neutrons and the source neutron multiplication is small), it is possible to calculate the neutron spectra from $(\alpha,n)$ reactions.\textsuperscript{(12,13,14)}

At the present time, neutron spectrum calculations have been only moderately successful. This lack of success can be attributed to rather inaccurate $(\alpha,n)$ and $(\alpha,ny)$ cross sections. Most calculations assumed that the emission of a neutron leaves the daughter nuclei in the ground state and that the emission is isotropic; this is not true in many instances. Accurate $(\alpha,n)$ cross sections are virtually non-existent for many of the light elements. Until accurate cross section data become available, experimentally measured neutron spectra are superior to calculated neutron spectra.
REFERENCES


^{238}\text{PuF},


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