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NEUTRON SPECTRA
OF PLUTONIUM COMPOUNDS

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

L. W. Brackenbush
L. G. Faust

February 1970

AEC RESEARCH &
DEVELOPMENT REPORT

L. W. Brackenbush 33314 3701-L MAY 21 1970

BNWL-1262

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BNWL-1262

UC-34, Physics

NEUTRON SPECTRA OF PLUTONIUM COMPOUNDS

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

L. W. Brackenbush and L. G. Faust

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Environmental and Life Sciences Division

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Printed in the United States of America
Available from
Clearinghouse for Federal Scientific and Technical Information
National Bureau of Standards, U.S. Department of Commerce
Springfield, Virginia 22151
Price: Printed Copy \$3.00; Microfiche \$0.65

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 - 2.7×10^{-9} rad/n/cm², plutonium tetrafluoride - 2.5×10^{-9} rad/n/cm², plutonium-aluminum alloy - 2.6×10^{-9} rad/n/cm², and plutonium metal (spontaneous fission spectrum) - 2.6×10^{-9} rad/n/cm².

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

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 (α, 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/cm²), (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 ^3He 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 $^{18}\text{O}(\alpha, n)$ 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 - 2.5×10^{-9} rad/n/cm², Pu-Al alloy - 2.6×10^{-9} rad/n/cm², PuO_2 - 2.7×10^{-9} rad/n/cm², and pure plutonium metal (fission spectrum) - 2.6×10^{-9} rad/n/cm².

THEORY AND OPERATION OF THE SPECTROMETERS

Theory of Operation of the ^3He Neutron Energy Spectrometer

The ^3He 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 ^3He gas can be admitted.

Each surface barrier detector has a sensitive area of about 2 cm² and a depletion depth of 250 microns at 50 volts bias. The sensitive volume of the detector head is about 280 mm³. 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 ³He gas in the detector head by the following reaction:

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 ³He(n,d)D 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 ^3He 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 ^3He 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 (2τ) resolution time. Smaller resolution times would be desirable to reduce the random coincidence from gamma and $\text{Si}(n,p)$ reactions, but the slight jitter introduced by electronic noise makes it necessary to have relatively large resolution times.

Calibration of the ^3He Spectrometer

The spectrometer was calibrated by using nearly monoenergetic neutrons produced by $\text{T}(p,n)$ and $\text{D}(d,n)$ reactions from a 2 MeV

FIGURE 1
Block Diagram of Fast Coincidence System for ^3He Neutron Spectrometer

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 ^3He 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 ^3He 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 ^3He 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.

FIGURE 2
Energy Calibration Curve for ^3He Spectrometer

The neutron peaks sometimes contained only about 100 counts per ten channels, so that the counting statistics are relatively poor.

The ^3He spectrometer also exhibits pulse pile-up problems in strong gamma fields. Exposures made with a $^{12}\text{C}(\text{d},\text{n})$ 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 ^3He Spectrometer

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

FIGURE 3
Neutron Cross Sections for $^3\text{He} (n,p)$ and $^6\text{Li} (n,\alpha)$ Reactions

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 ^3He 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}Pu). The "gross" measurement was made with 100 psia of ^3He 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 $^3\text{He}(n,p)\text{T}$ 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 ^3He Spectrometer Exposed to a PuF_4 Source

FIGURE 5
 Neutron Spectrum from PuF_4

Theory and Operation of the ^6Li Neutron Energy Spectrometer

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

As in the case of the ^6He spectrometer, the neutron energy is determined by measuring the energy of the reaction products. The $^6\text{Li}(n,\alpha)$ 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 ^6Li detector is less than that of the ^3He detector. If the thickness of the ^6LiF layer is increased to increase the neutron sensitivity, the resolution of the detector deteriorates. For a 150 microgram/square centimeter thick layer of ^6LiF , the width of the peak resulting from slow neutrons is typically 300 keV (FWHM). The ^6Li 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 $^6\text{Li}(n,\alpha) \text{ dn}'$ 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 $^6\text{Li}(n,\alpha) \text{ dn}'$ and $^6\text{Li}(n,\alpha)^3\text{H}$ 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 ${}^6\text{Li}$ 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 ${}^6\text{Li}$ spectrometer and because of electronic gain shift problems.

The data analysis of the ${}^6\text{Li}$ spectrometer is similar to that of the ${}^3\text{He}$ spectrometer. The "background" is much lower because of the high Q value of the ${}^3\text{Li}(n,\alpha)$ reaction, so that the ${}^6\text{Li}$ 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.⁽³⁾ Since the ${}^6\text{Li}$ 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 ${}^6\text{Li}$ neutron spectrometer exposed to a large, low exposure (6 percent ${}^{240}\text{Pu}$) 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 ${}^3\text{He}$ neutron spectrometer with a high exposure (22 percent ${}^{240}\text{Pu}$) 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 ${}^{240}\text{Pu}$) 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 ${}^{18}\text{O}(\alpha, n)$ by isotopic enrichment. The spectrum below about 1.5 MeV is uncertain because of competition from

TABLE 1

NEUTRON SOURCE DATA

<u>Source Identification</u>	<u>Mass Plutonium</u>	<u>Isotopic Composition in Weight Percent</u>					<u>Size of Source</u>	<u>Yield Neutrons/sec</u>
		<u>^{238}Pu</u>	<u>^{239}Pu</u>	<u>^{240}Pu</u>	<u>^{241}Pu</u>	<u>^{242}Pu</u>		
A PuF_4	765 g	2×10^{-5}	93	6	0.5		3-1/2" dia. x 4-1/8"	5×10^6
B PuF_4	518 g	0.090	72.8	22.3	3.81	0.99	3" dia. x 3"	6.4×10^6
C PuO_2	725 g	0.072	72.4	22.7	3.89	0.97	3-1/4" dia. x 2-3/4"	2.3×10^5
D Pu-Al	1.25 kg	0.561	65.25	23.68	7.10	3.39	2-1/2" x 2 1/4" x 4"	4×10^5

TABLE 2

SUMMARY OF MEASURED NEUTRON SPECTRA

<u>Source</u>	<u>Source Identification</u>	<u>Spectrometer</u>	<u>Shield</u>	<u>Peak* Energy</u>	<u>Average* Energy</u>
PuF ₄	A 765 gram	⁶ Li	No shield	1.10 MeV	1.29 MeV
PuF ₄	A 765 gram	⁶ Li	1" Lucite	1.10 MeV	1.25 MeV
PuF ₄	B 518 gram	³ He	1" lead and 1/8" tin	1.33 MeV	1.42 MeV
PuO ₂	C 725 gram	³ He	1" lead and 1/8" tin	2.2 MeV	2.0 MeV
Pu-Al alloy	D 1.25 kg	³ He	1" lead and 1/8" tin	1.60 MeV	1.55 MeV

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

TABLE 3

SUMMARY OF PUBLISHED NEUTRON SPECTRA

<u>Material or α, n Reaction</u>	<u>Peak* Neutron Energy</u>	<u>Average* Neutron Energy</u>	<u>Spectrometer</u>	<u>Comments</u>	<u>Reference</u>
$^{239}\text{PuF}(\alpha, n)$	0.87 MeV		^6Li - 200 keV resolution	1 kg source with some neutron scatter	G.W.R. Endres. <u>Neutron Spectra Measurements at Hanford Work Locations, HW-SA-3525</u> , General Electric Co. 1964.
$\text{Po-F}(\alpha, n)$	1.3 MeV		Nuclear track emulsions		A. Azilvasi, et al. J. <u>Nuclear Energy</u> , v. 11, p. 131. 1960.
$^{238}\text{PuF}(\alpha, n)$	1.2 MeV		^6Li and ^3He	Calibrated by PuBe and ^{252}Cf	T.R. Herold. <u>Nuclear Instr. & Methods</u> , v. 71, p. 40. 1969.
$^{238}\text{PuF}(\alpha, n)$	1.35 MeV	1.44 MeV	Stillbene(pro- ton recoil)	Very few neutrons above 3.2 MeV Predicted maximum	M.E. Anderson. <u>Transactions American Nuclear Soc.</u> , v. 9, p. 600. 1960.
$^{238}\text{PuO}(\alpha, n)$	2.6 MeV	2.4 MeV	Stillbene(pro- ton recoil)	Low energy tail below 1.5 MeV	Anderson, <i>ibid.</i>
$^{238}\text{PuO}_2$	2.3 MeV		^6Li - 275 keV resolution	Calibrated by changing gain, PuBe , and ^{252}Cf	T.R. Herold. <u>Nuclear Applications</u> v. 4, p. 19. Feb. 1968.
$\text{Po-O}(\alpha, n)$	2.35 MeV	2.29 MeV	Stillbene - 100% resolution for 4.85 MeV peak in 3 MeV neutrons	Po-Be calibrated by nuclear track emulsions	A.G. Khabkhpashev. <u>Soviet J. Atomic Energy</u> , v. 7, p. 591. 1960.

*Note: Numbers listed are accurate to two significant figures only.

FIGURE 6
Neutron Spectrum from ^6Li Spectrometer Exposed to PuF_4

FIGURE 7
Neutron Spectrum from PuF_4

FIGURE 8
Neutron Spectrum from PuO_2

$^3\text{He}(n,d)\text{D}$ and other reactions and from large statistical errors from "background" events. A spectrum presented by Anderson⁽⁴⁾ for $^{238}\text{PuO}_2$ 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 ^3He 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 ^{240}Pu and ^{238}Pu is very similar to the Watt fission spectrum for ^{235}U fission induced by thermal neutrons. Terrell⁽⁶⁾ concludes that all experimentally measured fission spectra ($^{235}\text{U} + n$, $^{239}\text{Pu} + n$, and ^{252}Cf) are Maxwellian distributions, and that the average energy of the distribution is related to ν , 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 ν , and hence the average energy of the fission spectrum, are quite similar for the thermal neutron fission of ^{235}U ($\nu = 2.46$, $E = 1.95$ MeV), and spontaneous fission of ^{238}Pu ($\nu = 2.33$, $E = 1.93$ MeV), and spontaneous fission of ^{240}Pu ($\nu = 2.26$, $E = 1.92$ MeV). Spontaneous fission of ^{238}Pu and ^{240}Pu 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 (α, 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 $^{18}\text{O}(\alpha, n)$ spectrum.⁽⁹⁾ The $^{18}\text{O}(\alpha, n)$ spectrum shown is from a $^{210}\text{Po}-\alpha-0$ source with a peak neutron

TABLE 4

NEUTRON YIELDS FROM PLUTONIUM ISOTOPES AND COMPOUNDS ^(a)

Isotope	Spontaneous Fission Half Life	ν n/fission	Yield n/sec-g	(α, n) Reaction Rate n/sec-g Isotope
^{236}Pu	$3.5 \times 10^9 \text{ y}$	2.3	3.7×10^4	
^{238}Pu	$4.9 \times 10^{10} \text{ y}$	2.33	2.62×10^3	
$^{238}\text{PuF}_4$				2.1×10^6
$^{238}\text{PuO}_2$				1.4×10^4
^{239}Pu	$5.5 \times 10^{15} \text{ y}$	3.0	3×10^{-2}	
$^{239}\text{PuF}_4$				4.3×10^3
$^{239}\text{PuO}_2$				4.5×10^1
^{240}Pu	$1.22 \times 10^{11} \text{ y}$	2.25	1.02×10^3	
$^{240}\text{PuF}_4$				1.6×10^4
$^{240}\text{PuO}_2$				1.7×10^2
^{242}Pu	$7.1 \times 10^{10} \text{ y}$	2.18	1.7×10^3	
$^{242}\text{PuF}_4$				1.7×10^2
$^{242}\text{PuO}_2$				2.7
^{244}Pu	$2.5 \times 10^{10} \text{ y}$	2.3*	5.1×10^3	
$^{241}\text{AmO}_2$				$2.6 \times 10^3^*$

*Estimated

(a) Data taken from L. G. Faust. Neutron and Photon Spectra, Dose Rate and Its Changes with Time Since Chemical Separation for High Exposure Plutonium, BNWL-SA-1644, Battelle Memorial Institute, Pacific Northwest Laboratories, March, 1968.

FIGURE 9
Calculated Neutron Spectrum from PuO_2

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 $^{18}\text{O}(\alpha, n)$ 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 $^{18}\text{O}(\alpha, n)$ spectrum and three-fourths of the distribution function for the spontaneous fission spectrum. Note that this calculated spectrum is quite similar to the experimentally 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 measurements show the neutron yield from this sample to be approximately 13 times higher than that arising from spontaneous fission alone. Presumably, most of the neutrons are generated from $\text{Al}(\alpha, n)$ 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 ^3He detector may cause small errors in the energy calibration for neutrons entering the detector nearly perpendicular to the surface barrier detectors.

FIGURE 10
Neutron Spectrum from Pu-Al Alloy

CONCLUSIONS

The results obtained with the ^3He 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 ^3He spectrometer results have sharper neutron peaks, probably because of the better energy resolution of about 100 keV. The ^3He 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 ^3He 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 $^{18}\text{O}(\alpha, n)$ spectrum⁽¹⁰⁾ weighted by the appropriate amounts determined from neutron yield calculations from Table 4 for spontaneous fission and (α, 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) ^{240}Pu and relatively

small amounts (about 0.07 percent) ^{238}Pu ; these spectra differ from $^{238}\text{PuO}_2$ spectra because of the much higher $O(\alpha, n)$ yield for ^{238}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/cm². These conversion factors are calculated by using 200 keV increments and the data contained in National Bureau of Standards Handbook 63⁽¹¹⁾ according to the formula:

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

where $R(E)$ is the rad-cm²/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-cm²/sec), or by calculating the flux for nearly point sources, and multiplying by the fluence to dose conversion factors (dimensions of rad-cm²/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:

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

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 5

FLUENCE TO DOSE CONVERSION FACTORS				
Source	Source (a) Identification	Average Energy (MeV)	Fluence to Dose Conversion Factor (b) (rad/n/cm ²)	Fluence to Dose Equivalent Conversion Factor (b) (rem/n/cm ²)
PuF ₄	A	1.3	2.46 x 10 ⁻⁹	2.4 x 10 ⁻⁸
PuF ₄	B	1.4	2.53 x 10 ⁻⁹	2.5 x 10 ⁻⁸
PuO ₂	C	2.0	2.74 x 10 ⁻⁹	2.5 x 10 ⁻⁸
PuAl alloy	D	1.6	2.63 x 10 ⁻⁹	2.6 x 10 ⁻⁸
Pu metal (fission spectrum)		1.9	2.60 x 10 ⁻⁹	2.5 x 10 ⁻⁸

(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.

- . $1-x$ is the fraction of the neutron yield from $O(\alpha, n)$ reactions,
- . R_{sf} is the average fluence to dose (or dose equivalent) conversion factor for spontaneous fission (2.60×10^{-9} rad-cm²/n or 2.46×10^{-8} rem-cm²/n),
- . $R_{\alpha, n}$ is the average fluence to dose (or dose equivalent) conversion factor for $O(\alpha, n)$ reactions for a ^{210}Po alpha source (3.14×10^{-9} rad-cm²/n or 2.67×10^{-8} rem-cm²/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 (α, n) reactions.^(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 (α, n) and $(\alpha, n\gamma)$ 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 (α, n) cross sections are virtually nonexistent for many of the light elements. Until accurate cross section data become available, experimentally measured neutron spectra are superior to calculated neutron spectra.

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