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BIOHAZARDS OF AEROSPACE NUCLEAR SYSTEMS
Quarterly Report
for January 1 - March 31, 1967

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SANDIA LABORATORIES

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ABSTRACT

Analytical studies have been made on the probability of entry of large particles into the nose. A computer program was written to determine various fluid dynamical parameters of particles. Calculations have been made on the maximum range of the alpha from ^{238}Pu in tissue and on dose rates from small $^{238}\text{PuO}_2$ microspheres. Estimates are given of the gamma and neutron dose rates to skin from contact with these sources. Difficulties in handling these microspheres without spreading contamination is discussed.

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SUMMARY

In support of the aerospace nuclear safety program, analytical and experimental studies have been made on the potential hazards in the use of $^{238}\text{PuO}_2$. These studies included calculations which showed that for a sedentary subject no microsphere of PuO_2 larger than 170 microns could be inhaled into the nose. A computer program was written to determine various fluid dynamical properties of particles having densities from 1 to 12 grams/cm³.

Dosimetric studies have included calculations of the alpha, gamma, and neutron dose rates to tissue from the microspheres. Dose rates from the alpha emission are orders of magnitude higher at close distances than the other radiation emitted.

Measurements of the 17 Kev X-ray from ^{239}Pu show greatly decreased transmission through polyvinyl chloride as compared with a plastic material which does not contain chlorine.

In manipulating the $^{238}\text{PuO}_2$ particles it was discovered that the "clean" microspheres contaminated the probes, container and counting equipment with which they came in contact.

LEGEND

A	atomic weight
A_γ	gamma activity, photons/sec-gm
C	concentration of particles inhaled
Ci	curies
C_o	ambient particle concentration
E	energy, Mev
e_s	efficiency of collection
n	neutron
p	number of particles/cm ³
Q	volume flow rate, cm ³ /sec
R	range of alpha ray
Re	Reynolds number
S	cross-sectional area, cm
U	flow velocity, cm/sec
V_s	settling velocity, cm/sec
Z	atomic number
α	maximum range of alphas, microns
μ	microns
μ Ci	microcurie
ρ	density, grams/cm
ϕ	flux density, emissions/sec-cm ²
χ	distance from source
ζ	drag coefficient

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1.0 INTRODUCTION

This report includes a summary of work performed at Oak Ridge National Laboratory during the first quarter of 1967 in support of the program on aerospace nuclear safety of the Radiological Effects Division, SANDIA Corporation. Primary emphasis of this work is on the physical and internal dosimetry of small radioactive particles. During this report period, most of the effort was directed specifically toward radiological health problems associated with $^{238}\text{PuO}_2$ microspheres; however, some dose calculations were performed for a $^{90}\text{SrTiO}_3$ isotope power source assuming three different configurations of the source and shielding materials.

2.0 INHALATION HAZARDS OF PLUTONIUM MICROSPHERES

2.1 Probability of Entry into Nose

Studies on the probabilities of entry of PuO_2 particles into the external nares are important to any analysis of hazards that such material may present to the respiratory system. Using the respiratory air flow curves of Silverman,^{2.1} et al., and an assumed cross-sectional area of intake of 2.5 cm^2 , velocity flow charts have been constructed. As a first approach these flow velocities were compared with the settling velocities of spherical particles of density 10. This gives a relationship (dashed line, Fig. 2.1) showing the maximum size particle that can be inhaled during various time fractions of the inspiratory cycle and is not directly equivalent to the probability of particle intake. A more useful value is the ratio of the concentration of inhaled particles to the ambient concentration as a function of particle size. This was calculated as follows:

$$\text{Let } U_t = Q/S$$

$$\text{where } Q = \text{air flow rate at a time } t, \text{ cm}^3/\text{sec}$$

$$S = \text{cross-sectional area of external nares, cm}^2$$

$$U_t = \text{flow velocity at time } t, \text{ cm/sec.}$$

If the particles are assumed to be uniformly mixed with the exposure volume, the concentration of material entering the nares will be pUS (particles/sec) where p is the number of particles per cm^3 . However, since the particles have a downward velocity

Fig. 2.1 Fraction of particles inhaled (solid line) and theoretical maximum (dashed line) as a function of microsphere diameter.

$-V_s$ then only $p(U_t - V_s)S$ particles/sec can enter. An efficiency factor e_s can then be calculated,

$$e_s = \frac{p(U_t - V_s)S}{pU_t S}$$

which gives the fraction of particles entering compared with the number that would have entered had their settling velocity been zero. This factor, e_s , is then adjusted for the fraction of time during inspiration that U_t occurs. Calculations have been made for ten particle diameters between 26 and 160 microns at ten points on the inspiratory cycle. The results are shown as the solid line on Fig. 2.1 which gives the fraction of particles inhaled ($\frac{C}{C_0}$) versus particle diameter. To apply these values in a given situation the concentration of particles directly under the nasal openings would have to be known (or assumed).

The data used in preparing Fig. 2.1 were calculated from pneumotachograms of sedentary subjects having an average tidal volume of ~830 milliliters. A maximum intake velocity of 367 cm/sec was calculated, based on an estimated peak flow rate of 55 liters/minute through an orifice (nares) having a cross-sectional area of 2.5 cm². This velocity corresponds to the settling velocity of a PuO₂ particle of 180 micron diameter ($\rho = 10 \text{ gms/cm}^3$).

2.2 Particle Settling Velocities

Settling velocities, Reynolds numbers, drag coefficients, relaxation times, and stop distances have been calculated for spherical particles falling freely in air at 20° C and 760 mm Hg. Tables of these data are being prepared for particle densities from 1 to 12 g/cm³ and for diameters 1 to 1000 microns. The Reynolds number is calculated using an empirical formula suggested by Davies^{2,2} which was developed by fitting a polynomial in the dimensionless quantity ($\pm Re^2$) to carefully selected data. From considerations of particle dynamics the quantity ($\pm Re^2$) is obtained for the condition of a spherical particle falling at its terminal velocity. Settling velocities are calculated using the definition of Reynolds number. Table 2.1 lists the settling velocities for spherical particles having a density of 10 g/cm³.

Table 2.1

Terminal Velocities of $^{238}\text{PuO}_2$ Spherical Particles ($\rho = 10 \text{ g/cm}^3$) Falling through Air

Diameter (microns)	Velocity* (cm/sec)	Diameter (microns)	Velocity* (cm/sec)
1	2.98 -2	220	4.07 +2
2	1.19 -1	240	4.46 +2
3	2.68 -1	260	4.84 +2
4	4.76 -1	280	5.22 +2
5	7.44 -1	300	5.58 +2
6	1.07 0	320	5.94 +2
7	1.46 0	340	6.30 +2
8	1.91 0	360	6.65 +2
9	2.41 0	380	6.99 +2
10	2.97 0	400	7.32 +2
20	1.17 +1	420	7.65 +2
30	2.51 +1	440	7.98 +2
40	4.15 +1	460	8.29 +2
50	5.98 +1	480	8.60 +2
60	8.10 +1	500	8.91 +2
70	1.01 +2	520	9.21 +2
80	1.21 +2	540	9.51 +2
90	1.41 +2	560	9.80 +2
100	1.62 +2	580	1.01 +3
120	2.04 +2	600	1.04 +3
140	2.45 +2	700	1.17 +3
160	2.86 +2	800	1.30 +3
180	3.27 +2	900	1.41 +3
200	3.67 +2	1000	1.52 +3

* Expressed as a number times 10 raised to the power shown.

3.0 DOSIMETRY OF $^{238}\text{PuO}_2$ MICROSPHERES

3.1 Alpha Particle Range and Dose Rate in Tissue

In calculating the dose delivered to a small volume of tissue from energetic alpha particles, the most difficult problem is determining the maximum range as a function of alpha particle energy. Ideally, an experimental value of α -range should be used; however, in the absence of empirical data one might use the equation given by Kinsman^{3.1}:

$$R_{\alpha} = 1.24E - 2.62, \quad (4 < E < 8 \text{ Mev});$$

where R_{α} = range in cm of air at 1 atm and 15°C ($\rho = 0.00123 \text{ g/cm}^3$),

and E = energy in Mev.

The approximate range in tissue may be computed using the Bragg-Kleeman rule relating range, density and atomic weight as follows^{3.2}:

$$R_{\alpha} \frac{\rho_1}{\sqrt{A_1}} = \frac{R_1 \rho_2}{\sqrt{A_2}}$$

where R_1 = range in material denoted by subscript

ρ = density of material

A = atomic weight

For air, A is taken to be 14.5, $\rho = 0.00123 \text{ g/cm}^3$ and $R_{\alpha} = 4.18$ centimeters. If, for tissue, the effective atomic weight is 9, and $\rho = 1$, then

$$R_T = \frac{R_{\alpha} \rho_{\alpha} \sqrt{A_T}}{\sqrt{A_{\alpha}} \rho_T}$$

Based on this equation the alpha particle from $^{238}\text{PuO}_2$ (5.488 Mev) has a calculated range of 40.5 microns in tissue.

Knowing the range, one may calculate the range average dose to a 40 micron radius spherical shell of tissue surrounding a one micron diameter particle of $^{238}\text{PuO}_2$. The activity level of the source is

$$C = \frac{4\pi}{3} (5 \times 10^{-5})^3 \text{ cm}^3 \times 10 \frac{\text{grams}}{\text{cm}^3} \times 13.8 \frac{\text{Ci}}{\text{gm}} = 7.25 \times 10^{-5} \text{ microcuries}$$

To determine the range average dose over the irradiated volume of tissue set:

$$\text{Dose rate} = \frac{\frac{7.25 \times 10^{-5} \mu\text{Ci}}{1 \mu \text{ particle}} \times \frac{3.7 \times 10^4 \alpha/\text{sec}}{\mu\text{Ci}} \times \frac{5.488 \text{ Mev}}{\alpha} \times \frac{1.6 \times 10^{-6} \text{ erg}}{\text{Mev}}}{(2.78 \times 10^{-7} \text{ cm}^3) \times \frac{1.1 \text{ gms}}{\text{cm}^3} \times \frac{100 \text{ erg/gm}}{\text{rad}}}$$

$$\text{Dose rate} = 0.770 \text{ rads/sec}$$

where $2.78 \times 10^{-7} \text{ cm}^3$ = volume of a 40 micron radius spherical shell around a one micron core. Thus, the range-average alpha particle dose is 0.77 rad/sec (2770 rad/hour). Assuming a quality factor of 20, this corresponds to $5.54 \times 10^4 \text{ rem/hour}$ in tissue surrounding a one micron diameter $^{238}\text{PuO}_2$ microsphere.

The following equations, given by Dousset and LeGrande,^{3.3} were used to calculate energy loss in tissue as a function of distance χ from a $^{238}\text{PuO}_2$ source, where α is the maximum range of the alpha radiation in microns = 40.

For the range $0 \leq \chi \leq (\alpha - 18)$,

$$\frac{dE_2}{d\chi} = f_2(\chi) = \frac{1}{23. - 0.17 (\chi + 106 - \alpha)} ;$$

thus for $0 \leq \chi \leq 22$ microns

$$\frac{dE_2}{d\chi} = f_2(\chi) = \frac{1}{11.78 - 0.17\chi} .$$

For the range $(\alpha - 18) \leq \chi \leq (\alpha - 5)$,

$$\frac{dE_3}{d\chi} = f_3(\chi) = 0.0005 (\chi + 106 - \alpha)^2 - 0.0858 (\chi + 106 - \alpha) + 3.803,$$

and for $22 \leq \chi \leq 35$ microns

$$\frac{dE_3}{d\chi} = f_3(\chi) = 0.0005\chi^2 - 0.0198\chi + 0.3182.$$

Finally, for $(\alpha - 5) \leq \chi \leq \alpha$,

$$\frac{dE_4}{d\chi} = f_4(\chi) = -0.007 (\chi + 106 - \alpha)^2 + 1.433 (\chi + 106 - \alpha) - 73.035,$$

hence for $35 \leq \chi \leq 40$ microns

$$\frac{dE_4}{d\chi} = f_4(\chi) = -0.007\chi^2 + 0.5085\chi - 8.982.$$

Some of the results of calculations using these equations are included in Table 3.1 along with estimates of α -particle dose rates based on the given rates of energy deposition in tissue. More complete results are given in Fig. 3.1 and Fig. 3.2, where $\frac{dE}{d\chi}$ and dose rate respectively are plotted as continuous functions of distance from the center of a one micron diameter $^{238}\text{PuO}_2$ microsphere embedded in tissue.

Table 3.1
Dose Rate as a Function of Tissue Depth
from a $^{238}\text{PuO}_2$ Particle 1 μ Diameter

$x(\text{microns})$	$\frac{dE}{dx} \left(\frac{\text{Mev}}{\text{micron}} \right)$	Dose Rate* rad/sec
0.5	0.0855	1170
1.0	0.0861	294
2.0	0.0874	74.6
3.0	0.0887	33.7
4.0	0.0901	19.2
5.0	0.0915	12.5
6.0	0.0929	8.81
7.0	0.0944	6.58
10.0	0.0992	3.39
20.0	0.1193	1.02
25.0	0.1357	0.74
30.0	0.1742	0.66
35.0	0.2391	0.66
39.0	0.2025	0.45

* Dose rate as a function of distance is equal to $\frac{dE}{dx} \times \frac{1}{x^2} \times 3.42 \times 10^3$ rad/sec.

Fig. 3.1 Energy deposition in tissue from ^{238}Pu alpha radiation.

Fig. 3.2 Dose rate from a 1 micron diameter $^{238}\text{PuO}_2$ source in tissue.

3.2 Scintillation Spectrometry of Alpha Radiation

Calculation of dose rate from small $^{238}\text{PuO}_2$ particles requires a knowledge of the energy spectrum of the emitted radiation. The apparatus used for spectrum analysis consists of an Amperex type XP-1010 phototube viewing an NE 102 plastic scintillator disk mounted on a lucite light pipe (Fig. 3.3). The enclosing chamber may be evacuated for calibration purposes or whenever the counting situation requires it. A series of NE 102 scintillator assemblies is available with diameters ranging from 1 to 11 mm, and thicknesses ranging from 32 to 500 microns.

To measure the emission spectrum of the relatively "hot" particles used in this study, it is necessary to separate the particle and the scintillator surface enough to reduce the counting rate to a tolerable level. A 130 μ diameter microsphere (Group 3 #2), containing about 130 microcuries of ^{238}Pu was affixed onto adhesive tape (3 M Co. #180) which was attached to a 1 inch diameter aluminum ring fitted into the source holder of the spectrometer. It was not known if the particle would fracture due to internal voids when subjected to a vacuum. Therefore, to minimize the potential hazard of dispersion of the ^{238}Pu in the event of explosive fracture, the source ring with the particle attached was placed in a container having a small opening in the lid. This container was placed in the vacuum system and the pressure lowered slowly by a pump which was provided with an exhaust filter. The chamber pressure was held at less than 5×10^{-2} torr for an hour and any stresses that might be applied to the particle due to internal pressures were essentially maximum. Air was admitted into the chamber and the filter and chamber were checked for contamination. None was found and the particle appeared to be intact when viewed with a binocular microscope.

The spectrometer was calibrated with an ^{241}Am standard and the source ring was placed in the chamber for measurement using a 3 mm diameter by 50 μ thick NE-102 scintillator. At a source-detector separation of 12.3 cm (geometry factor $\sim 3.72 \times 10^{-5}$) the counting rate was 2803 ± 6 c/m. An estimate of the total emission was 34.0 μCi or 26% of the total activity. This figure is somewhat in error due to scattered alpha particles striking the scintillator and because of analyzer errors associated with the first two channels. The absorbed dose delivered to the scintillator volume may be calculated from the pulse-height spectrum and this will be done with the aid of a computer program.

SCHEMATIC OF APPARATUS FOR DOSIMETRY

Fig. 3.3 Schematic of apparatus used for $^{238}\text{PuO}_2$ dosimetry.

3.3 Absorption of 17 Kev X-rays

A $^{238}\text{PuO}_2$ microsphere was sent to the Whole Body Counting Facility at ORNL for studies on the mass absorption by low Z material of the low energy X- and gamma radiation emitted by the source. Particle 3 #1 was used in this dosimetry study. It was spherical, having a diameter of 139 microns, weighed 7 μg and contained about 82 μCi of ^{238}Pu .

For safety in handling, the microsphere was taken from its shipping vial and placed on a strip of transparent adhesive tape (3M Co. #810). It was then covered with a 13 micron thick layer of vinylidene chloride (Saran) which, together with the beryllium window, provided the minimum absorber thickness. A sample of the plastic was examined and found to have a density of 1.64 g/cm^3 (Handbook of Chemistry and Physics lists 1.65 to 1.72).

A thin crystal "wound probe" assembly was used to obtain spectral information. This consists of a 0.1 cm thick sodium iodide scintillator, 2.5 cm diameter that is covered by a 100 micron thick Be light shield. The phototube output is recorded on a 512-channel analyzer (Nuclear Data Model 130).

The microsphere was placed about 0.6 cm above the face of the crystal and a series of absorbers ranging in thickness from 340 to 2380 microns was interposed between source and detector. Vinyl sheets having a measured density of 1.3 g/cm^3 were used as the absorbing material. Because of the high counting rate very short counting times were adequate. A correction is made automatically for coincidence losses; panel meter readings indicated dead time at about 40% for the highest count rate. Background for the energy interval used is less than one count per minute. The analyzer was calibrated to give a channel width of 1.0 Kev; channels 11 through 20 were summed, so that the output was in the form of counts per minute for all energies from 11 through 20 Kev. It was noted that the calculated "half-thickness" for vinyl (.8 mm) was within 10% of that found for the 17 Kev emission from a ^{239}Pu plated source of 9.5 mm diameter. Table 3.2 gives the values found for particle 3 #1.

After these measurements were taken it was observed that absorbers of methyl-methacrylate (Lucite) gave significantly different results. Figure 3.4 shows the preliminary values taken from channels 11 through 20 (primarily the 17 Kev X-rays) using the ^{239}Pu plated source. It is suggested that the chlorine atoms in the vinyl are responsible for the seven-fold increase in the absorption coefficient of this plastic. New absorbers of polystyrene (a hydro-carbon) and biological materials are being acquired to replace the vinyl.

Table 3.2
Count Rate from 139 micron diameter $^{238}\text{PuO}_2$ Microsphere as a Function of
Absorber Thickness

Vinyl Thickness (mm)	Total mg/cm^2	Counts per Minute* (11 - 20 Kev)
0	18.8	934,082
0.34	63.0	729,929
0.68	107.	467,143
1.02	151.	360,195
1.36	196.	274,686
1.70	240.	206,243
2.04	284.	150,830
2.38	328.	122,920

* Using 0.1 cm thick x 2.54 cm diameter NaI(Tl) Crystal, 512-channel analyzer,
Inherent absorber = 13 micron Saran + 100 micron Be.

Fig. 3.4 Transmission of 17 Kev X-rays from ^{238}Pu through two types of plastic absorbers.

3.4 Gamma Dose Rates

Data used in calculations:

Density of $\text{PuO}_2 = 10 \text{ gm/cm}^3$ (Ref. 3.4),

Gamma activity, $A_\gamma = 2.1 \times 10^8$ photons/sec-gm (Ref. 3.5),

Gamma energy = 0.0435 Mev/photon (Ref. 3.5),

Absorbed gamma dose rate = $1.7 \times 10^{-7} \frac{\text{rads/hour}}{\text{photons/cm}^2\text{-sec}}$ (Ref. 3.6).

3.4.1 Skin Dose Calculations

Table 3.4 gives calculated dose rates to 1 cm^2 of skin from $^{238}\text{PuO}_2$ microspheres ranging from 25 to 250 microns in diameter. Certain simplifying assumptions have been made which result in final figures that represent an upper limit estimate of expected dose rate. These assumptions are (1) There is no self-absorption of the photon energy within the particle; (2) There is no backscattering of the gamma rays from the tissue; (3) Fifty per cent geometry is assumed, that is, half of the total emitted photons enter the skin; and (4) These gamma rays intersect the skin within an area of 1 cm^2 . This is chosen to relate to the early reference in ICRP Publication 6 (Rev. 1962) to averaging the dose over 1 cm^2 area of skin. These assumptions permit the calculation of the dose rate from a representative size particle and extrapolation to other sizes using the ratio of activity levels.

For a 25 micron diameter particle, using the above data these values are derived:

$$\text{volume} = 8.181 \times 10^{-9} \text{ cm}^3,$$

$$\text{mass} = 8.181 \times 10^{-8} \text{ grams},$$

$$A_\gamma = 17.18 \text{ photons/sec-gm}$$

$$\phi (50\%) = 8.59 \text{ photons/sec-cm}^2$$

Thus, the dose rate = 1.46×10^{-3} mrad/hour to 1 cm^2 area of skin.

It should be noted that this figure does not include the contribution from the low energy X-rays. Spectroscopy, using thin crystals, is in progress to determine the magnitude of this contribution (Section 3.3).

3.4.2 Correction Factors for Geometry

Values given in Table 3.4 are surface dose rates and, ignoring electronic non-equilibrium, these would decrease with depth in tissue. This decrease would be due to energy absorption and the inverse square law effect. A geometric factor is given in Table 3.3 to correct for the fraction of energy passing through a 1 cm^2 area at various distances from the source.

Table 3.3

Fraction of Emitted Photons from Point Source on Skin that Pass
through 1 cm² at Various Depths*

Depth (cm)	F
0	0.5000**
.011	.49
.023	.48
.034	.47
.045	.46
.057	.45
.068	.44
.080	.43
.091	.42
.10	.41
.15	.37
.20	.33
.25	.30
.30	.27
.35	.24
.40	.21
.45	.19
.50	.17
.55	.15
.60	.14
.65	.12
.70	.11
.75	.10
.80	.091
.85	.083
.90	.076
.95	.070
1.00	.065

* Assumptions: No photon-tissue interaction occurs.

** 50% of rays enter skin through area of 1 cm².

$$F = \frac{r \csc \phi - d}{2r \csc \phi};$$

where

$r = 0.5642$ cm, radius of 1 cm² circle,

d = depth in tissue, cm,

$$\cot \phi = \frac{d}{r}.$$

Table 3.4

Estimated Maximum Dose Rate to Surface of 1 cm² of Skin from ²³⁸PuO₂

Particle Diameter (microns)	Neutron Dose (mrem/hour)	Gamma Dose (mrads/hour)
25	1.48×10^{-4}	1.29×10^{-3}
50	1.18×10^{-3}	1.03×10^{-3}
75	4.00×10^{-3}	3.48×10^{-2}
100	9.47×10^{-3}	8.26×10^{-2}
125	1.85×10^{-2}	.161
150	3.20×10^{-2}	.279
175	5.08×10^{-2}	.442
200	7.58×10^{-2}	.660
225	1.08×10^{-1}	.940
250	.148	1.29

3.5 Neutron Dose Rates

3.5.1 Skin Dose Calculations

The simplifying assumptions used in previous sections have been applied to obtain a first approximation to the neutron dose. For a 25 micron diameter particle the neutron flux density through the area of concern is assumed to be ϕ (50%) = 1.554×10^{-3} n/sec-cm² and, from section 3.5.2, the neutron dose rate = 1.48×10^{-4} mrem/hour to 1 cm² area of skin. Table 3.4 gives the estimated maximum dose rates to the surface of 1 cm² of skin as a function of ²³⁸PuO₂ particle size. The maximum theoretical neutron emission rate is taken as 3.8×10^4 n/sec-gm (Ref. 3.4) while the absorbed neutron dose is equal to

$$.0954 \frac{\text{mrem/hour}}{\text{n/cm}^2\text{-sec}} \text{ (Section 3.5.2)}$$

3.5.2 Flux to Dose Rate Conversion Factor

A factor was calculated for converting neutrons per square centimeter to equivalent dose based on the approximate neutron spectrum for ²³⁸PuO₂ as shown in Fig. 3.5. Conversion factors, taken from NBS Handbook 63 (Fig. 3.6), were weighted by the relative number of neutrons at each energy, Table 3.5. The average of these factors is 2.65×10^{-8} rems/neutrons-cm². The corresponding flux density producing 100 mrem for a 40-hour work week is 26.2 neutrons/cm²-sec.

4.0 SOLUBILITY STUDIES

A literature search was made for recent work on the solubility of plutonium oxides. The rate of translocation of Pu is apparently quite low although no studies have been found correlating particle size and/or residence time with solubility in lung fluids. Preparations are being made to make measurements on the in vitro solubility of ²³⁸PuO₂ in simulated body fluids.

Fig. 3.5 Neutron spectrum of $^{238}\text{PuO}_2$ (Ref. 3.7).

Fig. 3.6 First collision dose as a function of neutron energy.

Table 3.5
Conversion of Neutron/cm² from ²³⁸PuO₂ to Rems

E ₀ Mev	Relative Number of Neutrons	rem/n-cm ² * x 10 ⁸	Relative Per Cent	Fractional rem/n-cm ² x 10 ¹⁰
.3	1.0	1.5	2.747	4.12
1.0	2.8	2.6	7.692	20.00
1.5	4.3	2.5	11.812	29.53
2.0	7.2	2.6	19.778	51.42
2.4	8.1	2.7	22.251	60.08
2.8	5.5	2.7	15.108	40.79
3.0	4.2	2.8	11.537	32.30
3.5	2.3	2.9	6.318	18.32
4.0	1.0	3.0	2.747	8.24
4.8	0		SUM	2.65 x 10 ⁻⁸

* Ref: NBS Handbook 63, p. 7 (1957)

5.0 TRANSFERABLE RADIOACTIVITY FROM MICROSPHERES

A shipment of $^{238}\text{PuO}_2$ particles was received from the Sandia Corporation in December 1966, and placed in a radiochemical hood for unpacking. The iron cask was probed and smeared and the exterior of each vial was smeared as it was unpacked. No contamination was found on the smears. All particles appeared intact when viewed through the glass vials using a binocular microscope. The vial containing the 155 micron diameter particle (Group 2 #2) was transferred to a different laboratory for dosimetry measurements.

In order to provide containment of the $^{238}\text{PuO}_2$ in the event of loss or fracture, a glove bag was installed around the counting chamber. A $^{235}\text{UC}_2$ particle of 145 microns diameter was placed inside the glove bag and the techniques for handling the plutonium particle were checked with the similar but less hazardous material. An ^{241}Am alpha source was inserted in the chamber and a calibration was obtained for a 3 mm diameter scintillator by adjusting the gain until the peak appeared in the proper channel of the analyzer. A vacuum of <0.1 torr is necessary for this method of calibration and the source-to-scintillator distance was 10 cm. The ^{241}Am was removed and with health physics surveillance, the unopened vial containing particle (Group 2 #2) was placed inside the glove bag, and the bag was sealed. The vial was then opened and a sharpened wooden stick, moistened with water, was used to pick up the particle. Whereas, this was a simple and direct technique with the UC_2 bead, some difficulty was experienced in getting the PuO_2 bead to stick to the end of the wooden probe. The tip of the probe was in prolonged contact with the bead and the bottom of the vial. Part of the difficulty may be attributable to the strong electrostatic attractions observed to be present between the PuO_2 particle and various spots in the vial. When the bead was finally attached to the probe it was brought into position over the plastic scintillator. Before contact was made the PuO_2 bead was forced off the probe by electrostatic forces and landed 3 cm away on an aluminum plate. Subsequent pickup from the plate was accomplished with no difficulty. When an attempt was made to position the bead directly on the scintillator face, difficulty was experienced in separating the bead from the wooden stick. It was necessary to moisten the probe several times during this operation. For this purpose, a drop of water had been

placed on the apparatus nearby. After positioning, the chamber was sealed, but the measurement could not be made because of the excessively high counting rate (~100% dead time). At no time was the PuO_2 bead subjected to a vacuum. The particle was replaced in the vial with no difficulty, the chamber was again sealed and the background examined. The scintillator was found to be grossly contaminated (several thousand counts per minute). The probe and drop of water were found to be contaminated also. The apparatus was sealed and placed in a radiochemical hood where it was decontaminated with ethyl alcohol. There were three possible sources of the observed transferable contamination: the $^{235}\text{UC}_2$ bead, the $^{238}\text{PuO}_2$ bead, or the ^{241}Am source. Because of the low specific activity of uranium the intact UC_2 bead produced a negligible count rate compared with the level of contamination. Also, the effective energy of the uranium alpha group, 4.35 Mev, is much lower than the energy of the alpha contaminant as observed from the swab used to decontaminate the scintillator. While the energy of the ^{241}Am alpha group is approximately the same as the ^{238}Pu alpha group, this possibility is highly improbable in view of the fact that no contamination has been removed from this source in hundreds of identical calibrations before and after the contamination incident. Thus, it is concluded tentatively that the contaminant is plutonium; however, this will be confirmed by radiochemical analysis. The remaining vials were wiped inside with alcohol moistened swabs which were subsequently analyzed for α -activity. Table 5.1.

Because of the extensive cleaning received by these particles before leaving SANDIA and because of the observed minimal contamination of all the other vials, it seems likely, although not certain, that no loose contamination existed on the particles prior to our handling of them. The fact that contact with water is the only obvious difference in handling of the particle over previous treatment suggests that water may have some unexpected deleterious effect on PuO_2 microspheres. Tests will be made on the remaining microspheres to determine whether contact with water will result in spread of contamination.

6.0 OTHER

Shielding calculations were made on three configurations of a 250 watt (electrical) SNAP isotopic power source. Two candidate fuel materials, $^{90}\text{SrTiO}_3$ and $^{238}\text{PuO}_2$, were considered for each unit. Much of this information is of a classified nature and has been delivered to Sandia Corporation under separate cover.

Table 5.1

Results of Smears Taken from Vials used in Shipping of Microspheres

Group Number	Alpha (dis/min)	Beta (dis/min)
Group 1 #1	15	120
2 #1	216	60
3 #1	0	0
4 #1	0	0
3 #2	0	80
4 #2	0	0
2 #3	18	0

7.0 REFERENCES

- 2.1 L. Silverman, et al., AMA Arch. Ind. Hyg. Occupational Med. 3:471 (1951).
- 2.2 C. N. Davies, Proceedings of the Physical Society, 57, Pt. 4, No. 322 (July 1945).
- 3.1 U. S. Department of Health, Education and Welfare, Public Health Service, Radiological Health Handbook, S. Kinsman (ed), September 1960, p. 63.
- 3.2 W. H. Bragg and R. Kleeman, "On the α -Particles of Radium and their Loss of Range in Passing through Various Atoms and Molecules," Phil. Mag. 10, 318-340 (September 1905).
- 3.3 M. Dousset and J. LeGrand. Etude Théorique de la Dose Délivrée A un Tissue Superficiellement Contaminé Par un Emetteur α . Health Physics 11:171-178 (March 1965).
- 3.4 C. A. Rohrmann, "Radioisotopic Heat Sources" HW-76323 (Rev. 1) October 1963.
- 3.5 D. H. Stoddard and E. L. Albenesius, "Radiation Properties of ^{238}Pu Produced for Isotopic Power Generators," DP-984 (July 1965).
- 3.6 B. J. Henderson, "Conversion of Neutron or Gamma Ray Flux to Absorbed Dose Rate," ORSORT Report XDC-59-8-179 (October 1963).
- 3.7 R. J. Everett. (Personal communication).

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