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METHODS OF ASSESSING THE LUNG DOSE FROM INHALED $^{238}\text{PuO}_2$

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CFSTI REICES

Abstract

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Plutonium-238 is a prime fuel for space electric power having been used in the SNAP-9A system and is the candidate fuel for SNAP-19 and SNAP-27 in the oxide form. The fuel is made in the form of microspheres (50-250 μ) designed to eliminate inhalation hazards. However, there are thermo and aerodynamic forces present in launch and re-entry situations that may breakup or vaporize this fuel to inhalable sizes. This study is concerned with the methods of assessing lung doses from inhaled plutonium-238 dioxide. Methods of approach are examined and a discussion of these doses is made in relation to the biological effects observed. A model is given which considers the alpha particle energy loss with increasing size and the dose rate to a tissue sphere corresponding to the attenuated alpha range. The present ICRP uniform dose concept is also shown and some comparisons made between the two methods.

Introduction

After a high-altitude release of $^{238}\text{PuO}_2$ and the production of inhalable particles you can show through a probability analysis that large numbers of people can inhale a few particles. Particle sizes are expected to be in the range 10-100 μ , as seen from the SNAP-9A debris. This is a special lung exposure case that is not covered by the ICRP lung dose

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approach or the ICRP Lung Committee deposition predictions. Their models pertain to some known particle distribution uniformly deposited in the lung and there is no definite guidance for an unknown distribution of a few particles. The pulmonary compartment with its long retention times for deposited insoluble material was the area of interest in these studies. This region was considered to consist of moist nonciliated epithelial tissue of varying thickness. Some investigators estimate the depth of this tissue and end the alpha range at that point, however, there is evidence of cell damage below the basement membrane and therefore the doses were calculated throughout the alpha range. The density of this epithelial tissue was taken to be 1.0 gm/cc.

Fuel Properties

The alpha and gamma emissions from $^{238}\text{PuO}_2$ are given in Figure 1.^{1, 2} The alpha energy is considered to be 5.49 Mev throughout this study. The neutron abundance as a function of energy is given in Figure 2. for neutrons resulting from spontaneous fission and (α, n) reactions with oxygen.³ Isotopic fuel characteristics are shown in Table 1.

Efficiency of Inhalation and Deposition of $^{238}\text{PuO}_2$ Particulates

To predict the efficiency of inhalation and deposition in the lower lung, the ICRP Lung Dynamics Committee's recommendations were used with modification to allow for the probability of particles entering the nose and the deposition of submicron particulates. In Figure 3, the predicted

deposition in the nasopharynx is shown as a function of particle size for particles of density 1 and 10. The abscissa is in terms of mass median diameter but we feel these curves can be used on single particles as well. These curves also represent the efficiency of particles entering the nose. In Figure 4, percent deposition in the pulmonary compartment is shown for unit and density 10 particles. Since there is little experimental data on the inhalation efficiency of particles less than .1 micron, the extension of these curves should be considered tentative until verified.

ICRP Method of Calculating Lung Dose

Everyone is quite familiar with the ICRP approach that considers the particulate matter uniformly deposited in the critical organ. Figure 5 shows the dose rates from the uniform irradiation of tissue from $^{238}\text{PuO}_2$ particulates as a function of particle size. The large particle sizes shown probably cannot be deposited in the lower lung but are included for academic reasons and for those biologists engaged in lung implantations of large plutonium particles. This method suffers from the fact that in actual exposures, deposition is nonuniform and in the case of an alpha emitter, the energy only affects a small lung mass. In addition, there is appreciable alpha loss and degradation in the PuO_2 particle itself; however, this can be corrected as is shown in Figure 6. Please note that these are alpha dose rates and the X-ray doses from $^{238}\text{PuO}_2$ will not be discussed at this time.

Local Tissue Dose

In Figure 7, the alpha $\frac{dE}{dx}$ dose rate is shown for a one-micron diameter $^{238}\text{PuO}_2$ particle as a function of tissue depth. The curve is a composite of the work of Dousset and LeGrand.⁷ The range is about 40.5 microns. We assume that the particle is surrounded by a tissue sphere, then the volumes of tissue irradiated as a function of particle size are shown in Figure 8. More important is the mass of tissue irradiated in Figure 9. If we consider that the alpha energy is deposited in a tissue mass equivalent to the alpha particle range then the local dose rates are quite high as shown in Figure 10. The same relationships with correction for alpha degradation are shown in Figure 11.

Discussion

Both dose methods fall short in evaluating the biological effects expected or observed. The effects observed show that effects are related to the number and size of particles in the lung. For example, at Battelle-Northwest Laboratories an exposure of Pu^{239} to dogs of 5-10 μc in the size range .1 - .5 CMD was sufficient to cause death. Now 5 μc of .1 micron size particles contain about 5.3×10^7 particles, and 5 μc of 1 micron size particles contain about 5.3×10^4 particles.

$$5 \mu\text{c of 10 micron} = 5.3 \times 10^1 \text{ particles}$$

$$5 \mu\text{c of 100 micron} = < 1 \text{ particle}$$

Now the uniform dose concept would say that 5 μc would be fatal regardless of size. Spheres greater than 100 microns have been implanted in dog lungs

at Los Alamos and the dogs have not died yet. What you see is a lesion of dead and inflamed tissue around the particle site. I think this points out that effects are caused by the number of damage sites as well as activity deposited.

The local tissue dose probably describes the actual doses received but no one knows how to handle doses of this magnitude in terms of effect. It would be desirable to convert this dose back to a uniform dose since maximum allowable ICRP doses use these values. One can do this by multiplying these local tissue dose rates by the ratio of local tissue mass irradiated and the total mass of the lung.

It is immaterial what dose values are used as long as they describe effect. Right now both approaches are unsatisfactory.

Hazard Comparison

The relative hazard of inhaled insoluble alpha emitters can be obtained in terms of the number of cells irradiated over the range of the alpha particle. The approach is geometric and one must assume that all cells have equal probability of becoming cancerous, and that all cancer induction is direct and not a result of circulating toxins beyond the alpha range. The volume of tissue irradiated per particle is given by:

$$\frac{\pi}{6} \left[(d+2R)^3 - d^3 \right] \quad (1)$$

and the number of cells irradiated per particle given by:

$$N_c = \frac{\text{Volume of tissue per particle}}{\text{Volume of a cell}} \quad (2)$$

or

$$N_c = \frac{\pi}{6V_c} \left[(d + 2R)^3 - d^3 \right] \quad (3)$$

These relationships are shown in Figure 12. The number of particles per curie is given by

$$N_p = \frac{6}{\pi S d^3} \quad (4)$$

and the total number of cells irradiated per curie as a function of "d" is:

$$N_t(d) = \frac{1}{SVc} \left[\left(\frac{d + 2R}{d^3} \right)^3 - 1 \right] \quad (5)$$

we can now define a relative hazard factor defined by

$$H(d) = \frac{N_t(d)}{N_t(d_{ref})} \quad (6)$$

using a 250 micron particle as the reference, we obtain the relative hazard as a function of size. See Figure 13.

Conclusions

The previous discussion probably raised more questions than answers. There is a definite need for additional study into the relation between dose and effect for insoluble alpha emitters. The wide use of these fuels in aerospace applications may well depend upon the answers to the lung dose problem. Dose values can be calculated in various ways but have little value unless they describe effect.

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Fig 1

Properties of $^{238}\text{PuO}_2$

Disintegration Energy and Scheme¹

Alpha (1)
(MEV)

Gamma (1,2)
(MEV)

Intensity (1,2,3)
(Per Alpha Disintegration)

5.495 (72%)

0.017

0.13

5.452 (28%)

0.0438

3.8×10^{-4}

5.352 (0.09%)

0.099

8.0×10^{-5}

5.208 (0.005%)

0.150

1.0×10^{-5}

0.203

4.0×10^{-8}

0.760

5.0×10^{-7}

0.875

2.0×10^{-7}

Fig. 2

TABLE 1

$^{238}\text{PuO}_2$ Fuel Characteristics

Specific Activity $(\frac{1}{3})$ ^{238}Pu 16.8 Ci/gm

Chemical Composition of PuO_2

Plutonium 88.2%

Oxygen $(\frac{4}{3})$ 11.8%

Density of PuO_2 $(\frac{10}{3})$ 10.0 gm/cm³

Plutonium Fraction 8.82 gm/cm³ (^{238}Pu 7.15 gm/cm³)

Oxygen 1.18 gm/cm³

Activity of Fuel

$$\begin{aligned} 7.15 \text{ gm/cm}^3 \times 16.8 \text{ Ci/gm} &= 1.2 \times 10^2 \text{ Ci/cm}^3 \\ &= 4.5 \times 10^{12} \alpha/\text{cm}^3\text{-sec} \\ &= 3.92 \text{ watts/cm}^3 \end{aligned}$$

$^{238}\text{PuO}_2$ Fuel Isotopic Composition $(\frac{3}{2})$

^{236}Pu $1.2 \times 10^{-4}\%$

^{238}Pu 81%

^{239}Pu 15%

^{240}Pu 2.9%

^{241}Pu 0.8%

^{242}Pu 0.1%

Particle Diameter vs Percent Deposition in the Nasopharynx Considering the Probability of the Particle Entering the Nose
(Breathing Rate = 20 L/min.)

Fig. 3

Mass Median Diameter (μ)

(Breathing Rate = 20 L/min.)

Mass Median Dia. (μ)

Ref: Health Physic Vol 12
 RAY, R. W. II, 1966

Mass Median Dia. (μ)

Percent Deposition

Alpha Dose Rate, ~~over~~ IN Lung tissue (1000gms.) vs $^{226}\text{RaO}_2$ Microsphere Diameter (5.49 Mev Alpha)

Fig. 5

EUGENE DIETZGEN CO.
MADE IN U. S. A.

NO. 340R-LS10 DIETZGEN GRAPH PAPER
SEMI-LOGARITHMIC
5 CYCLES X 10 DIVISIONS PER INCH

Dose Rate (rem/hr)

Dose Rate

$$D.R. = A_v V E_\alpha D_c / M$$

A_v - α emission rate/unit vol.

V - microsphere vol.

E_α - alpha particle energy

D_c - dose conversion factors

M - lung mass

~~That's all
the
only~~

$^{239}\text{PuO}_2$

Alpha Dose Rate over Lung vs Particle Diameter

FIG. 6

EUGENE DIETZGEN CO.
MADE IN U. S. A.

NO. 340R-LS10 DIETZGEN GRAPH PAPER
SEMI-LOGARITHMIC
5 CYCLES X 10 DIVISIONS PER INCH

(rem/hr)

Dose Rate

$^{238}\text{PuO}_2$ Alpha Dose Rate vs Tissue Depth

(μm Dia. Particle)

$R_{90} = 40.5 \mu\text{m}$ in Tissue

$$\left(\frac{dE}{dx}\right)_1 = \frac{1}{11.86 - 0.17x} \quad 0 \leq x \leq 22.5 \mu$$

$$\left(\frac{dE}{dx}\right)_2 = 0.0005x^2 - 0.0203x + 0.328 \quad 22.5 \mu \leq x \leq 35.5 \mu$$

$$\left(\frac{dE}{dx}\right)_3 = -0.007x^2 + 0.516x - 9.236 \quad 35.5 \mu \leq x \leq 40.5 \mu$$

$$D.R. = \left(\frac{dE}{dx}\right)_m \frac{A_v V D_c}{4\pi x^2 \rho_t}$$

A_v - α emission rate / unit vol.

V - microsphere vol.

ρ_t - tissue density

D_c - dose conversion factors

EUGENE DIETZGEN CO.

MADE IN U. S. A.

(rad/hr)

NO. 340K-L510 DIETZGEN GRAPH PAPER

SEMI-LOGARITHMIC

10 DIVISIONS PER INCH

$\frac{dE}{dx}$ Dose Rate

Volume of Tissue Irradiated (μ^3)

Mass of Tissue Irradiated (10⁻⁷ gms)

Mass of Tissue Irradiated by ²³⁸PuO₂ Alpha Particle as a Function of Microsphere Diameter

$$M = \frac{\pi}{6} \rho_t [(d + 2R_t)^3 - d^3]$$

ρ_t - tissue density

d - microsphere diameter

R_t - alpha particle range in tissue

Particle Diameter (μ)

Dose Rate (Rem/hr)

Dose Rate to Lung Tissue in PuO_2 Alpha Particle Range vs Microsphere Diameter

RCE = 10

$$D.R. = \frac{A_v V E_{\alpha} D_c}{\frac{4}{3} \rho_t [(d + 2R_t)^3 - d^3]}$$

A_v - α emission rate / unit vol.

V - microsphere vol.

E_{α} - alpha particle energy

ρ_t - tissue density

d - microsphere diameter

R_t - alpha particle range in tissue

D_c - dose conversion factor

Dose Rate (rem/hr)

EUGENE DIETZGEN CO.
MADE IN U. S. A.

NO. 340-L310 DIETZGEN GRAPH PAPER
SEMI-LOGARITHMIC
3 CYCLES X 10 DIVISIONS PER INCH

No. of Cells Irradiated/Particle

$$N_c = \frac{V_t}{V_c}$$

N_c - Number of cells

V_t - volume of tissue / particle

V_c - Volume of cell

Ref: W D Bennett, SC

Relative Hazard Factor $H(d)$ vs Particle Diameter

Reference Dia. = 250 μ

MODEL

GRAPH

$R_0 = 40.5 \mu$ in tissue

$$H(d) = \frac{\text{No. of Irradiated cells per Curie of Particles of Diameter, } d}{\text{No. of Irradiated cells per Curie of Particles of Diameter} = 250 \mu}$$

Fig. 16

Particle Diameter (μ)

Relative Hazard Factor

H_{250}