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PHOTON AND NEUTRON EMISSION FROM
 PuO_2 PARTICLES AS
A FUNCTION OF PARTICLE SIZE

For

Sandia Corporation

By

Roy S. Denham

March 10, 1967

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INTRODUCTION

The purpose of this memorandum is to present information on photon and neutron emission in $^{238}\text{PuO}_2$ fuel utilized for thermoelectric power systems. The photon and neutron flux at the surface of fuel particles as a function of particle size from 5 μ to 250 μ diameter has been calculated and is presented graphically.

I. FUEL CHARACTERISTICS

Calculations of photon and neutron flux were based on $^{238}\text{Pu O}_2$ fuel as described in Reference 1, supplemented with information from References 2 and 3. The characteristic fuel data from these references and as calculated by the writer and used in this memorandum are listed below:

A.	<u>Chemical Composition</u>	<u>Pu O₂</u>
	Plutonium (total)	88.2 %
	Oxygen	11.8%

B.	<u>Isotopic Composition⁽¹⁾</u>
	^{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 %

C. Specific Activity ⁽¹⁻³⁾ ^{238}Pu 16.8 Ci/gm

D. Density of Pu O₂ ⁽³⁾ 10.0 gm/cm³

Plutonium Fraction	8.82 gm/cm ³	(^{238}Pu 7.15 gm/cm ³)
Oxygen	1.18 gm/cm ³	

E. Activity of Fuel

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

(Contribution of other isotopes to fuel activity is negligible).

F. Spontaneous Fission Rate^(1,3)

²³⁸ Pu	1.14×10^3 fissions/sec-gram
fuel	8.1×10^3 fissions/cm ³ -sec

II. PHOTON FLUX

Photon emission data was based on References 1-5. These references were prepared for different purposes and present their data in varying formats; consequently, they are not always in complete agreement. For example, the 14 Kev L x-ray from conversion of the ²³⁴U daughter of ²³⁸Pu is mentioned only in References 1,2 and 4, and only Reference 2 gives a value for the emission rate; this x-ray is the predominant photon released from the fuel in terms of abundance and total energy, but is a negligible contributor to the radiation from massive or shielded quantities of fuel which are the primary concern under most conditions. The photon emission rates for the fuel described above are listed in Table 1. This tabulation includes the photons resulting from spontaneous fission, decay of fission products and daughter of fission products and daughter products, and accompanying alpha particle interaction with ¹⁸O. (1)

TABLE 1
PHOTON EMISSION RATES FROM $^{238}\text{PuO}_2$ FUEL

Photon Energy (Mev)	Reference Number	Abundance γ/α	Emission Rate	
			$\gamma/\text{cm}^3\text{-sec}$	Mev/ $\text{cm}^3\text{-sec}$
0.017	2	0.13	5.8×10^{11}	1.0×10^{10}
0.0435	1, 5 *	3.8×10^{-4}	1.7×10^9	7.5×10^7
0.0996	1	8×10^{-5}	3.6×10^8	3.6×10^7
0.15	1 **	1.1×10^{-5}	4.9×10^7	7.4×10^6
0.203	1	4×10^{-8}	1.8×10^5	3.6×10^4
(.04 - 0.5) Δ	1		2.1×10^9	$\sim 1.2 \times 10^8$
0.76	1, 3	5×10^{-7}	2.2×10^6	1.7×10^6
0.875	1	2×10^{-7}	9×10^5	7.8×10^5
(0.5 - 1.0) Δ	1		3.3×10^6	$\sim 2.5 \times 10^6$
(1.0 - 2.0)	1		4.7×10^4	$\sim 8 \times 10^4$
(2.0 - 2.0)	1		1.5×10^4	$\sim 4 \times 10^4$
(3.0 - 5.0)	1		1.4×10^3	$\sim 6 \times 10^3$
(5.0 - 7.0)	1		2.3×10^2	$\sim 1.4 \times 10^3$
TOTAL		0.13	5.8×10^{11}	1.0×10^{10}

Δ Not included in total

* Reference 2 indicates a 45 kev photon with an abundance of 3.4×10^{-4}

** Reference 3 indicates an abundance of 8.8×10^7 $\gamma/\text{watt-sec}$ (3.4×10^8 $\gamma/\text{cm}^3\text{-sec}$) for this energy, but does not make reference to the 0.10 or 0.20 Mev photons.

Photon flux calculations at the particle surface were based on the equation for uncollided flux at the surface of a uniform sphere. (6).

$$1) \quad \varphi = \frac{S_V}{2 \mu_s} \left[1 - \frac{1}{\mu_s d} + \frac{e^{-\mu_s d}}{\mu_s d} \right]$$

where S_V is the specific activity of the particle in $\gamma/\text{cm}^3\text{-sec}$,
 d is the particle diameter in cm,
 μ_s is the linear attenuation coefficient, of the material in cm^{-1} , and
 φ is the flux in $\gamma/\text{cm}^2\text{-sec}$

$$2) \quad \mu_s(\text{PuO}_2) = \left[\mu_s/\rho(\text{Pu}) \frac{\text{Pu}}{\text{PuO}_2} + \mu_s/\rho(\text{O}_2) \frac{\text{O}_2}{\text{PuO}_2} \right] \rho$$

where $\mu_s/\rho(\text{Pu})$ and $\mu_s/\rho(\text{O}_2)$ are the mass attenuation coefficients for Pu and O_2 respectively, in cm^2/gm

$\frac{\text{Pu}}{\text{PuO}_2}$ is the mass fraction of Pu in the particle = .882

$\frac{\text{O}_2}{\text{PuO}_2}$ is the mass fraction of O_2 in the particle = .118

ρ is the particle density = 10 gm/cm^3

The calculated attenuation coefficients for photons in the particle material are listed in Table 2.

TABLE 2
ATTENUATION COEFFICIENTS FOR PuO_2

Photon Energy (Mev)	Mass Attenuation Coefficients		Linear Attenuation Coefficient PuO_2 (cm^{-1})
	$\text{Pu}^{(4)}$ (cm^2/gm)	$\text{O}_2^{(6)}$ (cm^2/gm)	
.017	42.4	1.48	376
.044	13.3	0.23	117
.099	1.4	0.15	12.5
.150	2.65	0.14	23.4
.203	1.26	0.12	11.2
.760	0.113	0.070	1.08
.875	0.096	0.068	0.93

If the mean free path of the photon in the particle is long in comparison to the particle size (i.e. $\frac{1}{\mu} \gg d$), self-shielding in the particles can be neglected and the equation for the flux becomes

$$3) \quad \varphi = 0.25 d S_v$$

For particles of $d \leq 250 \mu$, self-shielding can be neglected for all photon energies greater than 0.5 Mev.

The gamma flux for the photon energies and energy groups listed in Table 1 has been calculated for the surface of 5, 25, 50, 100, 150 and 250 μ particles and is presented in Figures 1-5.

As shown in Figures 3 and 4, the abundance of higher energy photons increases with time, primarily as a result of the ingrowth of the ^{212}Bi ^{208}Tl daughters of ^{236}Pu . The time indicated on the dashed lines in these figures refers to time elapsed since purification of the plutonium isotopes.

It is apparent that for particles of the size considered, the 17 Kev x-ray overshadows all other photons. With spherical particles 1 cm in diameter, the energy emission from higher energy photons would approximately equal that of the L x-ray.

III. NEUTRON FLUX

Neutrons in PuO_2 result primarily from the α, n reaction with oxygen. The calculated yield of α, n neutrons is 1.9×10^4 neutrons/gm-sec, ⁽¹⁾ or 1.9×10^5 n/cm³-sec.

Spontaneous fission of ^{238}Pu yields 2.33 ± 0.08 n/f; ⁽¹⁾ other plutonium isotopes in the fuel do not contribute significantly to the neutron spectrum. The spontaneous fission neutron flux then is 2.33 n/f $\times 8.1 \times 10^3$ fission/cm³-sec = 1.9×10^4 n/cm³-sec. (References 2 and 3 indicate a fission neutron yield approximately 25% higher).

The total neutron yield is approximately 2.1×10^5 n/cm³-sec. Approximately 90% of the neutrons have an energy between 1.0 and 5.0 Mev. ⁽¹⁾

The total neutron cross-section for ^{238}Pu and ^{239}Pu in the 1-10 Mev range is less than 10 barns. The mean free path is then greater than λ , where

$$\lambda = \frac{1}{\mu} = \frac{1}{\sigma N}$$

$$\lambda = \frac{1}{\mu} = \frac{1}{10^{-23} \text{ cm}^2/\text{atom} \times \frac{6 \times 10^{23} \text{ atoms}}{238 \text{ grams}} \times \frac{8.82 \text{ gms Pu}}{\text{cm}^3}}$$

$$\lambda = 4.5 \text{ cm}$$

Particles up to 250 μ diameter can therefore be considered non-selfshielding. A correction must be made however, for the escape of alpha particles from the PuO_2 , reducing the neutron generation in small particles. The neutron

flux at the surface of a particle can be expressed by

$$\varphi = 0.25 d \left[S_{v_f} + S_{v_\alpha} (1 - f) \right]$$

where S_{v_f} and S_{v_α} are the specific neutron activity resulting from fission and α , n neutrons, respectively and f is the probability of an alpha particle escaping from the fuel sphere. For fuel spheres with a diameter less than the range of the alpha (16.8μ)⁽⁸⁾ all the alpha particles will escape and the sole neutron source is spontaneous fission. For diameters greater than the alpha range the escape fraction is:

$$f = 1.5 \frac{R}{d} \left(1 - \frac{1}{3} \frac{R^2}{d^2} \right)$$

where R is the alpha range in PuO_2 , 16.8μ

$$\text{Then } \varphi = 0.25 d \left[S_{v_f} + S_{v_\alpha} \left(1 - 1.5 \frac{R}{d} + 0.5 \frac{R^3}{d^3} \right) \right]$$

The calculated neutron flux at the surface of 5μ to 250μ particles is shown in Figure 6.

FIGURE 1

PHOTON FLUX FROM $^{238}\text{PuO}_2$ FUEL PARTICLES

KE LOGARITHMIC 46 7403
 3 X 3 CYCLES
 NEUFFEL ESSER Energy Flux, MeV/cm²-sec
 MADE IN U.S.A.

Particle Size, μ

FIGURE 2

PHOTON FLUX FROM $^{238}\text{PuO}_2$ FUEL PARTICLES

FIGURE 3

PHOTON FLUX FROM $^{238}\text{PuO}_2$ FUEL PARTICLES

FIGURE 4

PHOTON FLUX FROM $^{238}\text{PuO}_2$ FUEL PARTICLES

KE LOGARITHMIC 46 7403
3 X 3 CYCLES
KEUFFEL & ESSER CO.
MADE IN U.S.A.

Particle Size, μ

FIGURE 5

PHOTON FLUX FROM $^{238}\text{PuO}_2$ FUEL PARTICLES

FIGURE 6

NEUTRON FLUX FROM $^{238}\text{PuO}_2$ FUEL PARTICLES

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