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AEC Research and Development Report

PHOTON INTENSITIES AND THEIR
IMPORTANCE IN COUNTING
TRANSURANIC MATERIALS

MAY 1972

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PHOTON INTENSITIES AND THEIR IMPORTANCE IN
COUNTING TRANSURANIC MATERIALS

By
K. L. Swinth

Applied Physics and Instrumentation Department

May 1972

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PACIFIC NORTHWEST LABORATORIES
RICHLAND, WASHINGTON 99352

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TABULATION OF TRANSURANIC NUCLIDE PHOTON INTENSITIES

PHOTON INTENSITIES AND THEIR IMPORTANCE IN
COUNTING TRANSURANIC MATERIALS

SUMMARY

This report presents ratios of X-ray to alpha intensities for a number of transuranic materials and illustrates how these ratios can vary with changing isotopic composition. Plutonium is used as an example. Also shown is how the biological hazard as measured by the Maximum Permissible Body Burden (MPBB) can vary with isotopic composition for plutonium.

Plutonium from spent power reactor fuels presents a greater hazard on a unit weight basis than high-grade fissile material. The X-ray intensity is initially smaller for the power reactor material but increases as the ^{241}Am grows into the material. In fact, the X-ray intensity will eventually be greater than that from the high-grade material.

Although the isotopic composition is an important facet in determining the biological hazard and in interpreting the photon intensities from transuranic materials, it is not the only complication in interpretation of counting data. The low penetrating power (absorption) and, because of this, the manner in which the material is deposited will also affect the interpretation of counting data.

BACKGROUND

When one is attempting to determine the amount of a transuranic emitter present and when the alpha particles are attenuated by material surrounding the nuclide, one must base the assay on more penetrating radiations such as photons. Most of the transuranic alpha emitters lack gamma rays of sufficient intensity to be useful for assay at low concentrations; however, X-rays emitted by the daughter nuclide during the decay are of sufficient intensity for an assay. If one knows the ratio of X-ray intensity to alpha intensity, the alpha activity can be found; however, this ratio varies with isotopic composition and composition varies with time.

In most plutonium samples, for example, ^{239}Pu is the largest constituent and has a low specific activity. However ^{241}Am , which grows into the sample as a result of decay of ^{241}Pu , has a high specific activity and a high X-ray yield. Thus, the X-ray emission rate changes with time. In many cases the comparison of X-ray intensity with a calibration standard will give adequate measurements, but an accurate and complete radiation safety analysis requires knowledge of the isotopic composition of the sample.

DISCUSSION

X-RAY INTENSITIES

Measurements of transuranic X-ray intensities have been performed with two different counting systems and are reported elsewhere^(1, 2). Table 1 summarizes these measurements along with other relevant data for various isotopes. The average values were used for the calculations presented on the following pages and are compared in the Appendix with other values taken from the literature. The averages listed in Table 1 include both the measured values and values taken from the literature. Although the experimental precision reported by most of the authors is good, the accuracy of the various measurements is estimated to be $\pm 5\%$ at best. Without analyzing each experiment in considerable detail, a more conservative estimate of the accuracy would be $\pm 7\%$.

The ^{237}U and the ^{241}Am listed in the table result from the decay of ^{241}Pu . Plutonium-241 beta decays to ^{241}Am 99.9+% of the time, but in $4.4 \times 10^{-3}\%$ ⁽²⁵⁾ of the decays, alpha particles are emitted resulting in ^{237}U which is important because of its high specific activity. Although the other daughters are radioactive, they are unimportant because of their low specific activities. A large number of gamma rays are emitted in the decay of the transuranic isotopes; however, the only gamma generally considered important is the 59.6 KeV gamma of ^{241}Am which has an intensity of 35.9%. The Appendix lists many of the other gamma rays from the transuranic materials.

VARIATION IN X-RAY INTENSITIES

Because of the large variations in X-ray yield and in specific activity as noted in Table 1, the ratio of X-rays per alpha decay will vary considerably depending on the isotopic composition of the material of interest. The isotopic composition of the transuranic materials can vary widely. Plutonium, a man-made element, is produced primarily by the irradiation of ^{238}U within a reactor. Plutonium isotopic composition

varies with the flux level, the total flux, and the neutron energy spectrum. Flux levels and energy spectra vary from reactor to reactor, and thus, the isotopic composition of plutonium exposed to the same fluence will depend upon the reactor.

TABLE 1. Data for Some Heavy Isotopes

Isotope	Specific Activity Curies/g	Maximum Permissible Body Burden		X-rays per Alpha	
		μCi	μg	Measured ^(a)	Average ^(b)
^{238}Pu	16.8	0.04	0.0023	0.114 (0.112)	0.115
^{239}Pu	6.16×10^{-2}	0.04	0.65	0.0460 (0.0476)	0.0465
^{240}Pu	0.227	0.04	0.18	0.115 (0.106)	0.108
^{241}Pu	113	0.90	0.0082	- - -	- - -
^{242}Pu	3.91×10^{-3}	0.05	0.13	- - -	0.100
^{241}Am	3.22	0.05	0.016	- - -	0.376
^{237}U	8.2×10^4	- - -	- - -	- - -	0.63
^{242}Cm	3.21×10^3	0.05	1.55×10^{-5}	0.117 (0.113)	0.106
^{244}Cm	81.1	0.1	0.00123	0.0918 (0.0937)	0.0923
^{237}Np	7.06×10^{-4}	0.06	85	0.597 (0.543)	0.57

a. Values in parenthesis refer to data taken with the proportional-scintillation system (Ref 1) while the other values were taken with the solid-state system (Ref 2).

b. The averages include the measured values and recent values from the literature (see Appendix).

Figure 1 shows the calculated variation in isotopic composition with total exposure for a flux level of $5 \times 10^{13} \text{ n/cm}^2\text{-sec.}$ ⁽³⁾ If the plutonium produced in a reactor is for use as a fissile material the ^{239}Pu content will be greater than 90% by weight, whereas plutonium discharged from power reactors⁽⁴⁾ or recycle systems will be on the order of 60% ^{239}Pu by weight.

FIGURE 1. Variation in Isotopic Composition of Plutonium from Uranium Fuel Irradiated in a Thermal Flux of $5 \times 10^{13} \text{ n/cm}^2\text{-sec}$

Figure 2 shows the variation in X-ray intensities with time for a fissile grade plutonium. This figure illustrates the relative importance of the various isotopes. It is apparent that changes in the X-ray intensity with time are due to the build-up of ^{241}Am . It is important to note the intensity of the ^{238}Pu and ^{241}Am radiations even with their small physical abundances.

FIGURE 2. Variation in X-ray Intensities for a Fissile Grade Plutonium. The initial isotopic composition by weight is: ^{238}Pu -0.0074%, ^{239}Pu -94.36%, ^{240}Pu -5.29%, ^{241}Pu -0.34%, ^{242}Pu -0.015%.

Figure 3 illustrates the change in the number of X-rays per alpha particles for changing isotopic composition and indicates the relative contribution from ^{239}Pu . This was calculated for an isotopic composition changing in the manner illustrated in Figure 1. The large changes expected from the growth of ^{241}Am are not shown on this figure. Plutonium samples will contain ^{241}Am and larger ratios are to be expected.

FIGURE 3. *Change in X-rays Per Alpha Particle and Relative Contribution from ^{239}Pu for changing isotopic composition*

USE OF X/α RATIOS

As mentioned previously the transuranic elements are often monitored by their X-ray emissions. If a lung counter, survey instrument, or other counting unit is extensively calibrated for one isotope, it is not generally practical to perform a new calibration for each isotopic composition encountered. Two general methods can be used to standardize the counted material with the calibration material. In the first method, one would know the isotopic composition and use this to calculate X to alpha ratios. However, the isotopic composition may be difficult to obtain and then one can revert to intercomparison of X to alpha ratios with a counter.

Knowledge of the isotopic composition is ideal since parameters of interest such as the X-ray emission rate per Maximum Permissible Body

Burden or per unit dose can then be calculated. The biological hazard of plutonium material will depend on the isotopic composition as illustrated in Figure 4.

FIGURE 4. *Change in the Relative Biological Hazard of a Plutonium Sample with Changing Isotopic Composition*

In the case of plutonium, intercomparison of sources by counting is valid over a small range since the ratio of X-rays per unit time to dose rate or MPBB changes slowly and primarily reflects the growth of ²⁴¹Am into the mixture. Figure 5 shows the calculated ratio of X-ray emission rate to MPBB versus the X/α ratio. This is calculated for materials exposed in a reactor at the same nominal flux level used for the previous figures and before ²⁴¹Am has grown into the mixture.⁽³⁾ Families of these curves will exist for different flux levels and for the growth of

FIGURE 5. Variation of ^{239}Pu Isotopic Content and in X-rays Per Maximum Permissible Body Burden for Changing X-ray Per Alpha Ratios

^{241}Am into the mixture as illustrated by the dotted curves which were calculated for irradiation at a different flux. For high ^{239}Pu content the number of X-rays per MPBB will change primarily due to the growth of ^{241}Am ; therefore, a simple calculation can be used to roughly predict the number of X-rays/sec per MPBB.

$$X/\text{MPBB} = 73 \frac{(X/\alpha)_T}{(X/\alpha)_T - BK(\gamma/\alpha)_T}, (X/\text{sec}/\text{MPBB})$$

where $(X/\alpha)_T$ = Measured X/α ratio for material

$(\gamma/\alpha)_T$ = Measured ratio of 60 keV gammas to alphas

K = Constant relating counter X-ray efficiency to gamma efficiency.

B = Ratio of ^{241}Am X-ray abundance to gamma abundance, 1.05.

It should be noted that simple photon counting can be used to determine the X/MPBB ratio. The major reason for using the X/ α ratio is the background reduction and accuracy achieved with the coincidence technique. The above formula gives the X/MPBB based on bone as the critical organ.

If one has a figure similar to Figure 5, he can predict the X/MPBB ratio for all compositions if he knows the X/ α ratio, the γ/α ratio and the alpha activities at 5.5 and 5.1 MeV⁽¹⁾ for his specific sample.

$$(X/\alpha)' = [(X/\alpha)_T - K(\gamma/\alpha)_T] \frac{\alpha_T}{\alpha_{5.1}}$$

$$(X/\alpha)' = X/\alpha \text{ ratio corrected for } {}^{241}\text{Am}$$

$$(X/\alpha)_T, K, (\gamma/\alpha)_T = \text{see previous equation}$$

$$\alpha_T = \text{total alpha activity } (\alpha_T = \alpha_{5.5} + \alpha_{5.1})$$

$$\alpha_{5.1} = {}^{239}\text{Pu} + {}^{240}\text{Pu} \text{ alpha activity}$$

$$\alpha_{5.5} = {}^{238}\text{Pu} + {}^{241}\text{Am} \text{ alpha activity}$$

Next one finds the X/MPBB ratio from his figure using the corrected X/ α ratio, $(X/\alpha)'$, and then calculates the total X/MPBB ratio from:

$$(X/\text{MPBB})_T = X/\text{MPBB} \frac{(X/\alpha)_T}{(X/\alpha)'} \alpha_T/\alpha_{5.1}$$

With a solid-state system,⁽²⁾ the above steps are reduced to reading X/MPBB from the figure and multiplying by the ratio of the total to ${}^{241}\text{Am}$ X-ray activities since this system can be gated on the individual alpha peaks.

Similar techniques can be used to correct the alpha dose rate for isotopic composition. The dose rate does not vary as fast with decreasing ${}^{239}\text{Pu}$ content as the number of MPBB/ μg , but it too increases.

Similar techniques are applicable to other transuranic alpha emitters; however, the techniques require some knowledge of the composition of the material. By using a solid state system the alpha energy groups of ^{237}Np , ^{242}Cm , and ^{244}Cm can be identified and used to determine the activities from these isotopes when they are found in the presence of other transuranic elements.

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APPENDIX

TABULATION OF TRANSURANIC NUCLIDE PHOTON INTENSITIES

A P P E N D I X

TABULATION OF TRANSURANIC NUCLIDE PHOTON INTENSITIES

This appendix includes photon intensities and other general information for a number of transuranic materials. Since the decay schemes for these materials are complex (90 transitions identified in ^{239}Pu),⁽⁸⁾ I have not included decay scheme data per se and I have limited the data presented. In general, I have limited the alpha particle intensities to those greater than 1% and the photon intensities to greater than one per 10^5 decays. Photons above 150 keV were not included nor were the K X-rays emitted by the daughters. The K X-rays are usually more intense than neighboring gamma rays; however, they are not of interest in assigning decay schemes and are not widely tabulated. This is also the case for the L X-rays; however, a number of values have been tabulated.

Only current measurements have been used and in most cases the data was taken from the original published measurements. In general, the errors shown represent the experimental precision not the accuracy. I feel that most of the values are accurate within $\pm 10\%$.

Corrections for possible angular correlations were not made for the data presented. Experiments I have performed and that others have performed^(4, 22) show that angular correlations are not important within stated accuracies. An exception may be ^{237}Np ;⁽²⁴⁾ however, my preliminary experiments have been inconclusive.

PLUTONIUM - 238   ALPHA DECAY

$$T_{1/2} = 89.6 \text{ yr}$$

$$\text{Specific Activity} = 1.68 \times 10^1 \text{ Curies/gram}$$

ALPHAS

<u>E_{α}, MeV</u>	<u>Abundance</u>
5.495	72%
5.452	28%
5.352	0.13%

PHOTONS

<u>Energy, keV</u>	<u>Intensity, Number/Decay</u>												
<i>L X-rays</i>	$0.112 \pm 0.004^{(1)}$ $0.114 \pm 0.003^{(1)}$ $0.106 \pm 0.003^{(4)}$ $0.1288 \pm 0.0014^{(22)}$												
<i>K X-rays</i>	<u>Intensity, 10^{-5} Number/Decay</u>												
94.66 UK α_2	0.104												
111 UK β_1	0.078												
114.5 UK β_2	0.024												
<i>Gamma</i>	<u>Intensity, 10^{-5} Number/Decay</u>												
	<table><tr><th><u>(7)</u></th><th><u>(16)</u></th><th><u>(17)</u></th></tr><tr><td>34 ± 0.4</td><td>34</td><td></td></tr><tr><td>9.4 ± 0.9</td><td>9.2 ± 0.6</td><td>7.4</td></tr><tr><td>1.02 ± 0.12</td><td>1.27 ± 0.09</td><td>0.68</td></tr></table>	<u>(7)</u>	<u>(16)</u>	<u>(17)</u>	34 ± 0.4	34		9.4 ± 0.9	9.2 ± 0.6	7.4	1.02 ± 0.12	1.27 ± 0.09	0.68
<u>(7)</u>	<u>(16)</u>	<u>(17)</u>											
34 ± 0.4	34												
9.4 ± 0.9	9.2 ± 0.6	7.4											
1.02 ± 0.12	1.27 ± 0.09	0.68											
43.50 ± 0.04													
99.84 ± 0.04													
152.71 ± 0.05													

PLUTONIUM - 239   ALPHA DECAY

$$T_{1/2} = 2.44 \pm 0.05 \times 10^4 \text{ yr}$$

$$\text{Specific Activity} = 6.16 \times 10^{-2} \text{ Curies/g}$$

ALPHAS

<u>E_{α}, MeV</u>	<u>Abundance</u>
5.147	73.3%
5.134	15.1%
5.095	11.5%

PHOTONS

<u>Energy, ⁽⁸⁾ keV</u>	<u>Intensity, Number/Decay</u>

L X-rays

L α_1	11.618	}	
L α_2	13.615		
L α_2	13.439		$0.0460 \pm 0.0010^{(1)}$
L β_1	17.218		$0.0476 \pm 0.0012^{(1)}$
L β_2	16.429		$0.0459 \pm 0.0014^{(6)}$
L γ_1	20.167		$0.067 \pm 0.007^{(15)}$
L γ_2	20.483		$0.029 \pm 0.015^{(28)}$
L γ_4	21.496		$0.04 \pm 0.01^{(29)}$

K X-rays

98.46	UK α_1		
94.64	UK α_2		
110.38	UK β	}	
111.29			
114.55			
			$\approx 12 \pm 1^{(9)}$

Intensity, 10^{-5} Number/Decay*Gamma*

	<u>Intensity, 10^{-5} Number/Decay</u>			
	(15)	(9)	(8)	(6)
38.70 ± 0.03	8.3 ± 1.0	23 ± 4	1.63 ± 0.16	
51.61 ± 0.03	23 ± 2	36 ± 3	13.5 ± 1.4	30.4 ± 5.7
129.27 ± 0.03		5.7 ± 0.5	5.6 ± 0.7	
375.04 ± 0.03		1.3 ± 0.3	1.5 ± 0.1	

PLUTONIUM - 240   ALPHA DECAY

$$T_{1/2} = 6.6 \pm 0.1 \times 10^3 \text{ yr}$$

$$\text{Specific Activity} - 2.27 \times 10^{-1} \text{ Curies/g}$$

ALPHASE_α, MeVAbundance

5.159

75%

5.115

25%

PHOTONS, keVIntensity, Number/Decay*L X-rays*

$$0.115 \pm 0.003^{(1)}$$

$$0.106 \pm 0.004^{(1)}$$

$$0.1047 \pm 0.005^{(22)}$$

Gamma

43.49

$$\approx 1 \times 10^{-4} \text{ (25)}$$

PLUTONIUM - 241   BETA, ALPHA $T_{1/2} = 13.2 \text{ yr}$

Specific Activity - 113 Curies/g

BETAS $E_{\max} = 21 \text{ keV}$

Abundance = 99+%

ALPHAS ⁽¹⁴⁾ E_{α} , MeVAbundance

5.055	}	
5.042		1.5%
4.972		1.3%
4.896		83.2%
4.853		12.1%
4.797		1.4%

PHOTONS ⁽¹⁴⁾Energy, keVIntensity, 10^{-3} Number/Alpha Decay*Gamma*

	44.7 $\pm$ 0.3	1.7 $\pm$ 0.3
	56.6 $\pm$ 0.2	1.1 $\pm$ 0.3
	76.9 $\pm$ 0.2	7.3 $\pm$ 0.7
UK α	94.6 $\pm$ 0.2	127 $\pm$ 12
	98.4 $\pm$ 0.2	208 $\pm$ 20
	103.5 $\pm$ 0.2	45 $\pm$ 5
UK β_1	111.0 $\pm$ 0.2	77 $\pm$ 7
	114.0 $\pm$ 0.2	2.5 $\pm$ 0.5
UK β_2	114.5 $\pm$ 0.2	33 $\pm$ 3
	120.7 $\pm$ 0.5	0.5 $\pm$ 0.2
	148.5 $\pm$ 0.2	90 $\pm$ 9
	160.0 $\pm$ 0.2	3.2 $\pm$ 0.3

AMERICIUM - 241   ALPHA DECAY $T_{1/2} = 458 \text{ yr}$

Specific Activity = 3.22 Curies/g

ALPHAS

<u>E_{α}, MeV</u>	<u>Abundance</u>
5.477	85%
5.435	12.6%
5.378	1.7%

PHOTONS

<u>Energy, keV⁽¹¹⁾</u>	<u>Intensity, Number/Decay</u>

*L X-rays*L_ℓ 11.89L_{α₁} 13.961L_{α₂} 13.776L_{β₁} 17.764L_{β₂} 16.857L_{γ₁} 20.796L_{γ₁} 21.108L_{γ₄} 22.200.376±0.007⁽²⁾Intensity, Number/Decay*Gamma*26.345±0.001⁽³⁾

33.211±0.021

43.437±0.020

55.562±0.022

59.537±0.001⁽³⁾

67.520±0.056

98.997±0.04

103

122

125

150

(12)

(2)

(16)

(10)

0.025±0.002

0.011

7×10⁻⁴

0.359±0.007

0.38

0.353±0.006

(2.4±0.3)×10⁻⁴2.3×10⁻⁴2.0×10⁻⁴1.9×10⁻⁴1.6×10⁻⁴≤2×10⁻⁵1.1×10⁻⁵(5±2)×10⁻⁵2.8×10⁻⁵≤2.4×10⁻⁷4.8×10⁻⁶

113.8±1.5 (Np Kβ)

(7±4)×10⁻⁶

URANIUM - 237   BETA

$$T_{1/2} = 6.75 \text{ day}$$

$$\text{Specific Activity} = 8.2 \times 10^4 \text{ Curies/g}$$

BETASE_{max}

≈ 250 keV

Abundance

≈ 96%

PHOTONS⁽¹⁹⁾Energy, keV

26.348 ± 0.010

51.01 ± 0.03

59.543 ± 0.015

164.61 ± 0.02

208.005 ± 0.023

Intensity, 10⁻² Number/Beta Decay

2.41 ± 0.24

0.22 ± 0.10

36.0 ± 3.6

1.97 ± 0.06

23.3

Np X-raysL_ℓ 11.9

L 14.0

L_β 17.8L_γ 20.8K_{α2} 97.0K_{α1} 101.0K_{β1} 113.5K_{β2} 117.5Relative to 208 keV gamma

6.5 ± 0.7

115 ± 12

122 ± 12

33 ± 3

0.177 ± 0.020

0.247 ± 0.025

0.107 ± 0.010

0.034 ± 0.003

} 0.629 ± 0.040

CURIUM - 242   ALPHA $T_{1/2} = 163 \text{ day}$ Specific Activity = 3.21×10^3 Curies/gALPHAS

<u>Energy, MeV</u>	<u>Abundance</u>
6.115	73.5%
6.071	26.5%

PHOTONS

<u>Energy, keV</u>	<u>Intensity, Photons/Alpha Decay</u>
<i>L X-rays</i>	$0.117 \pm 0.003^{(1)}$ $0.113 \pm 0.005^{(1)}$ $0.0991 \pm 0.001^{(21)}$ $0.0975 \pm 0.0014^{(20)}$
	} *
<i>Gamma</i>	<u>(23)</u>
44.0	3.9×10^{-4}
101.8	3.5×10^{-5}
157.6	2.3×10^{-5}

---

*Due to lack of knowledge of the ^{242}Cm isotopic composition, these values are probably high.

CURIUM - 244   ALPHA $T_{1/2} = 17.6 \text{ yr}$

Specific Activity = 81.1 Curies/g

ALPHAS

<u>Energy, MeV</u>	<u>Abundance</u>
5.808	76.7%
5.766	23.3%

PHOTONS

<u>Energy, keV</u>	<u>Intensity, Photons/Alpha Decay</u>
<i>L X-rays</i>	$0.0918 \pm 0.0013^{(1)}$
	$0.0937 \pm 0.0037^{(1)}$
	$0.09484 \pm 0.0014^{(22)}$
	$0.0890 \pm 0.021^{(4)}$
<i>Gamma</i>	<u>(23)</u>
42.88	2.1×10^{-4}
100	1.5×10^{-5}
150	1.3×10^{-5}

NEPTUNIUM - 237   ALPHA

$$T_{1/2} = 2.14 \times 10^6 \text{ yr}$$

$$\text{Specific Activity} = 7.06 \times 10^{-4} \text{ Curies/g}$$

ALPHA

<u>Energy, MeV</u>	<u>Abundance</u>
4.871	3%
4.876	1%
4.815	1.4%
4.803	1.5%
4.786	42%
4.769	28%
4.764	5%
4.708	2%
4.692	1.6%
4.661	5.5%
4.636	6%

PHOTONS⁽¹³⁾

<u>Energy, keV</u>	<u>Intensity, Photons/Alpha Decay</u>
<i>L X-rays</i>	$0.597 \pm 0.006^{(1)}$
	$0.543 \pm 0.013^{(1)}$
29.6	0.13
45.5 } 46.7 } 48.5 }	1×10^{-3}
55.2 } 57.1 }	6×10^{-4}
62.9 } 64.8 } 68.8 }	2×10^{-3}
71.0 } 73.1 } 75.7 }	5×10^{-3}
80.6 } 86.6 }	0.13
94.5	4×10^{-2}
143.6	4×10^{-3}

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