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RADIATION SHIELDING FOR SMALL POWER SOURCES OF ^{170, 171}Tm

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Health and Safety
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by

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January 1969

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ABSTRACT

Dose equivalent rates measured with a 6.8-watt ^{170}Yb , ^{171}Yb source shielded with tantalum agreed closely with rates calculated from the number and energy distribution of bremsstrahlung photons from the beta decay of ^{170}Yb . This excellent agreement validated the calculational method. Shielding requirements were estimated for a hypothetical 62-watt power source of ^{171}Yb for the projected artificial heart application; the calculations suggest a required purity range of 10^{-4} to 10^{-5} Ci ^{170}Yb Ci ^{171}Yb for ^{171}Yb implanted in the human body.

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INTRODUCTION

Thulium-171, ^{147}Pm , and ^{238}Pu are candidate isotopes for power sources in artificial hearts.^(1,2) Forecasts of the eventual extent of application—potentially over 10^5 users—will require large quantities of the chosen isotope. A prime consideration for implanted isotopic power sources is the dose rate from the radionuclide and any radioactive impurities. Thulium-171 is particularly attractive for use in artificial hearts because its beta particles are low in energy (0.097 Mev) and are accompanied by gamma emission of low energy (0.067 Mev); hence its characteristic radiations are easily shielded.

However, the methods for large-scale production of this isotope also produce a second isotope, ^{170}Tm , an energetic beta emitter (0.970 Mev) that requires considerable shielding because of bremsstrahlung from beta interaction with the source medium. Preliminary calculations of the concentration of ^{170}Tm that could be tolerated in ^{171}Tm for the heart application indicated that this impurity could significantly increase shielding requirements even at a few parts per million (see Appendix). These calculations were based on the complex series of assumptions and approximations that are necessary in estimating the distribution of energies of photons produced in bremsstrahlung interactions. A shielding experiment was designed to test the validity of the resulting dose rate calculations so that a realistic appraisal of the tolerable level of the ^{170}Tm could be determined.

DISCUSSION

Preparation and Dose Rate Measurement of Thulium Source

A ^{170}Tm source consisting of four thulium sesquioxide (Tm_2O_3) wafers in a tantalum container was prepared. Forty-five wafers (~5 g Tm_2O_3 per wafer) had been irradiated in a Savannah River production reactor, to a ^{170}Tm content of 800 Ci/g Tm_2O_3 . The fabrication, pre- and post-irradiation physical properties, and chemical contamination of these targets have been described.⁽³⁾

The ^{170}Tm content of the four selected wafers was measured over a 34-day period using a twin bridge calorimeter. The half-life observed was 128 ± 1 days. A comparison of measurements of the tantalum-contained source with previous measurements of all the Tm_2O_3 wafers made 300 days earlier also indicated, on the average, a half-life of ~130 days. The ^{170}Tm content of the four wafers was 3.05 ± 0.15 kCi ^{170}Tm based on the conversion of watts to curies reported by Cornman⁽⁴⁾ of 445 ± 22 Ci ^{170}Tm /watt. The lack of other interfering radioisotopes was confirmed from the gamma spectrum of the contents of the tantalum cylinder measured at one meter through four inches of lead.

This 6.8-watt source of $^{170}\text{Tm}_2\text{O}_3$ contained 3050 Ci of ^{170}Tm with 1.79 disintegrations of ^{170}Tm per disintegration of ^{171}Tm . The Tm_2O_3 wafers formed a right circular cylinder 1.767 cm in diameter and 1.031 cm high. These were placed in a laminar shield of six telescoping concentric tantalum cylinders having top and bottom the same thickness as the sides. Tantalum was used because it is easier to machine than tungsten.

The exposure rates from this source were measured using small LiF thermoluminescent dosimeters (3.2 mm diameter, 4.0 mm high active area with 0.8 mm polyethylene walls) placed against the side of the cylinders midway between the ends. The center of each dosimeter was about 0.3 cm from the shield surface. The dosimeters were exposed to the source for 5 minutes, and their readings were compared with readings from dosimeters exposed to one roentgen of radium radiation at 42.0 cm to obtain the measured exposure rates.

Dose Rate Calculations

The absorbed dose rate, including buildup contribution, in millirad/hour from uncollided photons of E initial energy in Mev for a shield t_0 cm thick was calculated using

$$D(t, E) = \Phi(E) E B(E, \mu_o t_o) \frac{\mu_a}{\rho_a} \frac{(1.602 \times 10^{-6})(3.6 \times 10^3)}{(10^{-1})}$$

or

$$D(t, E) = \Phi(E) E B(E, \mu_o t_o) \frac{\mu_a}{\rho_a} 5.767 \times 10^{-2}$$

where

$\Phi(E)$ = the scalar flux in photons $\text{cm}^{-2} \text{sec}^{-1}$.

$B(E, \mu_o t_o)$ = the buildup factor for photon radiation of initial energy E Mev and a shield which has a thickness of $\mu_o t_o$ relaxation lengths.

μ_a = the linear energy-absorption coefficients for muscle in cm^{-1}

μ_o = the linear attenuation coefficient for either tungsten or tantalum in cm^{-1}

ρ_a = the density of muscle in g cm^{-3}

1.602×10^{-6} = the number of ergs per Mev

3.6×10^3 = the number of seconds per hour

10^{-1} = the number of ergs per gram of muscle per millirad.

Because only photon dose is considered, the quality factor is unity, and the absorbed dose rate in mrad/hr and dose equivalent rate in mrem/hr have the same numerical value.

Values for μ_o and μ_a are given in Table I, along with values for the linear attenuation coefficient for Tm_2O_3 , μ_c . Energy-absorption coefficients for muscle were obtained from NBS Handbook 85⁽⁵⁾ by assuming the density of muscle to be 1 g cm^{-3} . The linear attenuation coefficients (μ_o) of tantalum were calculated by converting the attenuation coefficients for tungsten^(6,7) using the following relationships:⁽⁸⁾

$$\tau = \tau_W \left(\frac{Z}{Z_W} \right)^n$$

where τ = the atomic photoelectric cross section of the element considered in barns atom^{-1}

τ_W = the atomic photoelectric cross section of tungsten in barns atom^{-1}

Z = the atomic number of the element considered

Z_W = the atomic number of tungsten

Values of n for various photon energies are given in Figure 1.

$$\sigma = \sigma_W \left(\frac{Z}{Z_W} \right)$$

where σ = the atomic scattering cross sections in barns atom^{-1}

$$\kappa = \kappa_W \left(\frac{Z}{Z_W} \right)^2$$

where κ = the atomic pair production cross sections in barns atom^{-1}

$$\mu = \tau + \sigma + \kappa$$

where μ = the total atomic cross section in barns atom^{-1}

$$\mu_o = \mu \rho \frac{N}{A} 10^{-24}$$

where N = Avogadro's number

A = the atomic weight

$10^{-24} = \text{cm}^2$ per barn

ρ = the density of the attenuating material in g cm^{-3}

The density of tantalum was assumed to be 16.6 g cm^{-3} .

TABLE I

Linear Attenuation and Energy-Absorption Coefficients

Photon Energy, Mev	μ_o, cm^{-1}		μ_c for $\text{TM}_2\text{O}_3, \text{cm}^{-1}$	μ_a for Muscle, cm^{-1}
	W	Ta		
0.02	1260	1050	500	0.544
0.03	425	353	168	0.154
0.04	193	161	76.8	0.0677
0.05	108	89.3	42.6	0.0409
K-edge	64.8 (0.06 Mev)	53.8 (0.06 Mev)	25.6 (0.0594 Mev)	0.0312 (0.06 Mev)
	44.0 (0.0695 Mev)	38 ^a (0.0674 Mev)	128 ^a (0.0594 Mev)	
	207 (0.0695 Mev)	198 ^a (0.0674 Mev)	127 ^a (0.06 Mev)	
0.08	145	120	57.3	0.0255
0.10	81.3	67.3	32.2	0.0252
0.15	27.8	23.1	11.0	0.0276
0.20	13.7	11.4	5.39	0.0297
0.30	5.65	4.72	2.22	0.0317
0.40	3.36	2.83	1.31	0.0325
0.50	2.41	2.03	0.935	0.0327
0.60	1.95	1.65	0.752	0.0326
0.80	1.47	1.26	0.566	0.0318
1.0	1.24	1.06	0.472	0.0308
1.5	0.950	0.814	0.361	0.0281
2.0	0.843	0.722	0.320	0.0251
3.0	0.782	0.670	0.295	0.0225
4.0	0.776	0.664	0.293	0.0203
5.0	0.789	0.676	0.297	0.0188
6.0	0.807	0.691	0.304	0.0178
8.0	0.845	0.722	0.318	0.0163
10.0	0.897	0.765	0.338	0.0154

^aCoefficients estimated

FIG. 1 VALUES OF n FOR PHOTOELECTRIC CROSS-SECTION EXPRESSION

The linear attenuation coefficient for Tm_2O_3 was obtained by first calculating the mass attenuation coefficient for thulium, $\frac{\mu_o}{\rho}(\text{Tm})$, using the above relationships and substituting this in the

$$\text{equation } \mu_c = \frac{2A(\text{Tm}) \frac{\mu_o}{\rho}(\text{Tm}) + 3A(0) \frac{\mu_o}{\rho}(0)}{2A(\text{Tm}) + 3A(0)} \rho(\text{Tm}_2\text{O}_3)$$

The mass attenuation coefficient for oxygen, $\frac{\mu_o}{\rho}(0)$, was obtained from References 5 and 6. The density of Tm_2O_3 was assumed to be 8.6 g cm^{-3} .

Values for the dose buildup factors, $B(E, \mu_o t_o)$, for 0.5 and 1.0 Mev photons were obtained from Reference 9. These dose buildup factors are given in Figure 2. The factors given in Figure 2 for other energy photons were obtained by linear interpolation.

FIG. 2 DOSE BUILDUP FACTORS FOR TUNGSTEN

Corrections for Source Geometry

The flux, $\Phi(E)$, in photons $\text{cm}^{-2} \text{sec}^{-1}$ was calculated by first assuming the right-circular cylindrical sources of radius R_0 and height H cm to be line sources (line AB in Figure 3) having a uniform source strength of N/H photons $\text{sec}^{-1} \text{cm}^{-1}$. Here N is the total activity in photons sec^{-1} . The line source was then positioned within the cylinder to correctly account for the self-absorption of the cylinder. This was done by calculating t/R_0 , $\mu_0 t_0$, and $\mu_c(t + R_0)$ and using these values to obtain $\mu_c t_c$ from Reference 10. t_c is the self-absorption distance in cm, t_0 is the thickness of the shield in cm, and t is the separation

FIG. 3 SOURCE AND SHIELD
AB is the assumed line source

distance from the edge of the source to the dose point in cm.
From $\mu_c t_c$, the effective self-attenuation distance in cm, t_c ,
was calculated.

The probability that an uncollided photon from an isotopic point source reaching a unit cross sectional area at a distance $t_c + t$ is

$$\frac{e^{-b}}{4\pi (t_c + t)^2}$$

where $\frac{1}{(t_c + t)^2}$ = the solid angle at the source subtended by a unit cross sectional area $t_c + t$ units away.

4π = the total steradians in a sphere

$$b = \mu_c t_c + \mu_o t_o$$

Because the source is a line rather than a point, the contribution from a line segment dl is

$$d\Phi = \frac{N dl e^{-b \sec \theta}}{H 4\pi (t_c + t)^2 \sec^2 \theta}$$

The relation of line segment dl to distance from line $t_c + t$ is

Since

$$dl = (t_c + t) \sec^2 \theta d\theta$$

$$d\Phi = \frac{N e^{-b \sec \theta}}{4\pi (t_c + t) H}$$

and

$$\Phi(E) = \frac{N}{2\pi (t_c + t) H} \int_0^\theta e^{-b \sec \theta} d\theta$$

this equation can be written simply

$$\Phi(E) = \frac{N F(\theta, b)}{2\pi (t_c + t)H}$$

where $F(\theta, b) =$ Sievert's integral

Values for $F(\theta, b)$ were obtained from figures on pages 385-390 in the Reactor Shielding Design Manual.⁽¹⁰⁾

Other Assumptions

The nuclear properties used in these calculations were obtained from the sixth edition of the Table of Isotopes⁽¹¹⁾ and are given in the decay schemes in Figure 4. The number of photons per disintegration from thulium-171 and thulium-170 are given in Table II. The number of bremsstrahlung photons per 50-kev energy interval per disintegration of thulium-170 were taken from calculations by Arnold.⁽¹²⁾ All photons in each energy interval were assumed to have an energy equal to the midpoint of the interval.

FIG. 4 DECAY SCHEMES FOR ^{171}Tm AND ^{170}Tm

TABLE II

Photons per Disintegration of ^{171}Tm or ^{170}Tm

<u>Isotope</u>	<u>Radiation</u>	<u>Energy, kev</u>	<u>Photons per Disintegration</u>
^{171}Tm	gamma	67	2.532×10^{-3}
	X-rays	52	1.747×10^{-2}
	bremsstrahlung	50	2.206×10^{-3}
^{170}Tm	gamma	84	3.459×10^{-2}
	X-rays	52	5.050×10^{-2}
	X-rays	9	1.349×10^{-1}
	bremsstrahlung	50	5.574×10^{-2}
	"	100	2.014×10^{-2}
	"	150	9.857×10^{-3}
	"	200	5.430×10^{-3}
	"	250	3.169×10^{-3}
	"	300	1.903×10^{-3}
	"	350	1.155×10^{-3}
	"	400	6.992×10^{-4}
	"	450	4.180×10^{-4}
	"	500	2.441×10^{-4}
	"	550	1.377×10^{-4}
	"	600	7.386×10^{-5}
	"	650	3.700×10^{-5}
	"	700	1.683×10^{-5}
	"	750	6.669×10^{-6}
	"	800	2.136×10^{-6}
	"	850	4.778×10^{-7}
	"	900	4.897×10^{-8}
	"	950	1.713×10^{-10}

Comparison of Measured and Calculated Dose Rates

The comparison of the calculated and measured dose equivalent rates is given in Table III and plotted in Figure 5. The good agreement validates the calculational method and the associated assumptions. The small differences are not significant in terms of shield weight.

TABLE III
Calculated and Measured Dose Rates

Shield Thickness, cm	Measured Exposure Rate, R/hr	Dose Equivalent Rates in Muscle, ^a rem/hr	Calculated Dose Equivalent Rates, rem/hr	Calculated-to-Measured Ratio
0.361	2,070	1,987	2,350	1.18
0.677	630	605	752	1.24
0.970	232	223	283	1.27
1.549	46.9	45.0	54.1	1.20
2.138	11.4	10.9	12.5	1.15
2.717	3.11	2.99	3.28	1.10

^a Exposure rate measured in air x 0.96⁽⁵⁾

FIG. 5 DOSE EQUIVALENT RATES

APPENDIX

Applications

These calculations were used to determine shielding requirements for hypothetical ^{171}Tm sources containing various quantities of ^{170}Tm impurity.

A simple geometry for the source was assumed for the artificial heart application: a right circular cylinder with height equal to diameter, clad by shielding material. The dose point was assumed to be on the outer surface of the shield midway between the two ends of the cylinder.

Three mean dose equivalent rates used in the evaluation of potential artificial heart power sources⁽¹⁾ were considered. They were:

- (1) 1 mrem/hr (8.766 rem/yr) for individuals 30 years old or less
- (2) 5 mrem/hr from 31 to 50
- (3) 15 mrem/hr for 51 and over.

A minimum power output of 4 watts was selected. Physical exertion or emotional disturbance can raise the output as high as 7.0 watts. However, excluding violent physical activity from the regimen of artificial heart patients, 1.5 to 4.0 watts should be adequate.⁽²⁾

If a 13% overall system efficiency is assumed, a minimum thermal output of 30 watts would be required. Studies by Thermo Electron Engineering Corporation have shown that in this case, a Rankine cycle, reciprocating engine with water as the working fluid is the most efficient means of converting thermal energy to work.⁽¹⁾ Experience operating these engines indicates an overall system efficiency of at least 12% and possibly 15% is feasible.

To obtain a minimum of 30 watts, the initial thulium-171 power input should be 62 watts assuming a two-year refueling cycle and a half-life of 1.92 years. The power density of $^{171}\text{Tm}_2\text{O}_3$ is 1.54 watts/cm³; ⁽¹²⁾ thus a 62-watt source will occupy 40 cm³, and will have a total activity of 3.48×10^5 Ci.

The mean dose equivalent rate, $\bar{D}(t,E)$, received over the two-year refueling cycle was considered rather than the initial dose equivalent rate for a 62-watt source. This was obtained from the relationship

$$\bar{D}(t,E) = \frac{D(t,E)}{T} \int_0^T e^{-\lambda T} dT$$

or

$$\bar{D}(t,E) = \frac{D(t,E)}{\lambda T} (1 - e^{-\lambda T})$$

where λ = the radioactive decay constant in years⁻¹

T = the refueling cycle or 2 years.

The mean dose equivalent rates were calculated for each photon energy level and tungsten shield thickness. The mean dose rate equivalent at each shield thickness was then obtained by summing the mean dose rate equivalent for all photon energy levels. These are given in Figure 6. Shield thicknesses, t , needed to

FIG. 6 MEAN DOSE EQUIVALENT RATE OF A ^{171}Tm SOURCE WITH VARIOUS ^{170}Tm CONTENTS

obtain mean dose equivalent rates of 1, 5, and 15 mrem/hr were taken from Figure 6. The weight of tungsten in pounds, W, represented by these thicknesses was obtained from the relation

$$W = 2\pi [(R_0 + t)^3 - R_0^3] 0.04255$$

where $0.04255 = \text{the density of tungsten in pounds cm}^{-3}$.

Calculations were made of the weight of tungsten shielding needed for a source containing pure thulium-171 and sources containing one disintegration of thulium-170 per 100, 1,000, and 10,000 disintegrations of thulium-171. When these $^{170}\text{Tm}/^{171}\text{Tm}$ ratios appeared to give unacceptable shield weights, ratios which result in mean absorbed dose rates of 1, 5, and 15 mrem/hr using a 10-pound tungsten shield were also calculated. Results of these calculations are given in Table IV.

TABLE IV

Shield Thicknesses and Weights

$\frac{\text{Ci } ^{170}\text{Tm}}{10^6 \text{ Ci } ^{171}\text{Tm}}$	Mean Dose Rates, ^a mrem/hr	Shield Thickness, cm	Shield Weight, lb
0	15	0.228	0.70
0	5	0.256	0.80
0	1	0.295	0.94
73	15	1.68	10.0
24	5	1.68	10.0
5	1	1.68	10.0
100	15	1.83	11.6
100	5	2.34	17.9
100	1	3.09	30.5
1,000	15	2.90	26.9
1,000	5	3.42	37.3
1,000	1	4.17	56.5
10,000	15	3.98	51.2
10,000	5	4.52	67.3
10,000	1	5.28	95.0

^aMean dose equivalent rate over a 2-year refueling cycle

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