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AEC RESEARCH AND DEVELOPMENT REPORT

PROPERTIES OF THULIUM METAL AND OXIDE

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PROPERTIES OF THULIUM METAL AND OXIDE

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ABSTRACT

The properties of thulium metal and thulium oxide were surveyed to establish specifications for reactor target materials for experimental production of ^{170}Tm for isotopic power source applications. Radiation characteristics and chemical, thermophysical, and mechanical properties are presented that are pertinent to the use of the activated targets without reprocessing after irradiation.

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INTRODUCTION

Thulium-170 and thulium-171 are promising heat sources for isotopic power generators. Thulium-170, a β-emitter with a half-life of 128 days, can be made by irradiating natural ^{169}Tm. No separations, fuel compound synthesis, or fuel form fabrication are required after irradiation; the irradiated ^{169}Tm target can be used directly as a heat source fuel. Thulium-171, a β-emitter with a half-life of 1.9 years, can be made by irradiating ^{169}Tm at high flux or by irradiating erbium enriched in ^{170}Er; in the former case the ^{170}Tm may be allowed to decay to increase the 171/170 ratio, and in the latter case chemical separation is required following irradiation.

The Savannah River Laboratory is developing technology for production of ^{170}Tm. In an initial test, thulium oxide was irradiated to 2.48 w/g in the Savannah River high flux demonstration. In subsequent work, both thulium oxide and thulium metal are being irradiated to evaluate cost and production parameters and to provide material for testing and evaluation.

In the course of work to establish specifications suitable for ^{169}Tm target materials for the irradiation studies, a detailed literature survey was made of the properties of thulium oxide and thulium metal applicable to the direct utilization of the irradiated target materials as heat source fuels. The information obtained is presented in the following data compilation.

For the most part, the thermal and mechanical properties reported here are for unirradiated natural thulium. Many of the properties have not been determined, but some could be calculated or estimated reliably from information on related and similar rare earths. The source of each datum is referenced; if calculated or estimated by the authors, the method is indicated. An attempt is made to evaluate the uncertainty in certain critical properties.

SUMMARY

Thulium sesquioxide, with an estimated melting point of 2375°C, is well suited as a target material for production of ^{170}Tm to be used subsequently in high-temperature isotopic heat sources. The oxide is readily available in adequate purity and can be fabricated to high-density solid forms before irradiation. It is highly refractory; inert in oxidizing, vacuum, or reducing environments; and is potentially nonreactive with refractory metal containers up to 2000°C.

Thulium metal, with a melting point of 1545°C, is a convenient target material, but may be limited to lower temperature heat-source applications.

PROPERTIES

			Ref.
Thulium-170	Half-Life:	128 ±1d	2,3
Thulium-171	Half-Life:	700 ±5d	3

A. THULIUM METAL

1. Composition

a. Radionuclide Abundance (calculated for an arbitrarily selected flux of 4×10^{14} n/(cm^2)(sec) at reactor discharge for ^{169}Tm irradiation). 4

Isotope	Specific Activity, w/g →	Concentration, wt % 0.62	2.24	2.94
^{169}Tm		95.3	77.9	60.6
^{170}Tm		4.7	16.3	21.5
^{171}Tm		-	2.9	8.4
^{170}Yb		-	2.9	9.0
^{171}Yb		-	-	0.5

	Ref.
b. Chemical Impurities (commercial samples before irradiation)	4

Elements not listed were <0.001 wt % or were not detected by the spark-source emission spectrograph. Variation in product is within 10X.

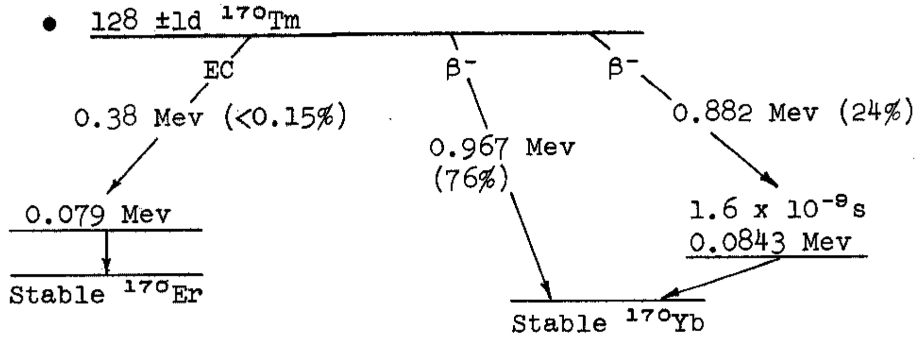
Element	Wt %
Ta	0.107
Yb	.0046
Er	.030
Ho	.0030
Dy	.0010
Tb	.0028
Gd	.063
Nd	.021
La	.0025
Y	.0053
Ni	.0155
Fe	.033
Mn	.0033
Cr	.010
Ti	.0012
Ca	.071
Si	.0055
Al	.0016
Mg	.014
F	0.0034

2. Specific Power

	Ref.
• 2 to 3 w/g of metal at reactor discharge is feasible	1,4
• 900 to 1400 Ci/g of metal at reactor discharge is feasible	1,4
• 445 ±2 Ci/w of ^{170}Tm measured on typical irradiated wafers	4

3. Radiation | Ref.

a. <u>^{170}Tm and ^{171}Tm Decay Processes</u> 5,6 7,8



1) <u>Internal Conversion of ^{170}Tm Gamma</u> 9,10 11

87% of the 0.0843 Mev gamma is internally converted. Conversion coefficients:

α_K = 1.38 ±0.04 (59.7 and 52.0 kev);

1.47 ±0.07; 1.32 ±0.05

α_L = 4.1 ±0.5 (8 kev); 4.11 ±0.21

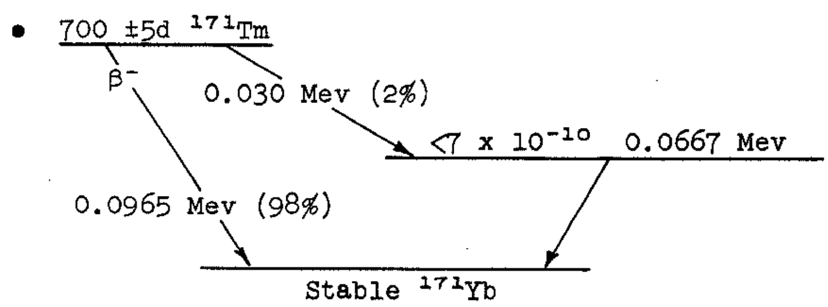
α_M = 1.2 ±0.2

$\alpha_{M + N + \ldots}$ = 1.38 ±0.08

I_K = 4.9 ±0.5%/β-disintegration;

$I_{(L + M)}$ = 16 ±2%/β-disintegration;

I_{γ} = 3.1 ±0.3%/β-disintegration.



Ref. 5,6 7,8

Ref.

2) Internal Conversion of ^{171}Tm Gamma 8

Over 87% of the 0.0667 Mev gamma is internally converted.

$$\alpha_K = 6.5 \pm 0.9$$

b. Radiation by Particle and Type

1) Alpha particles

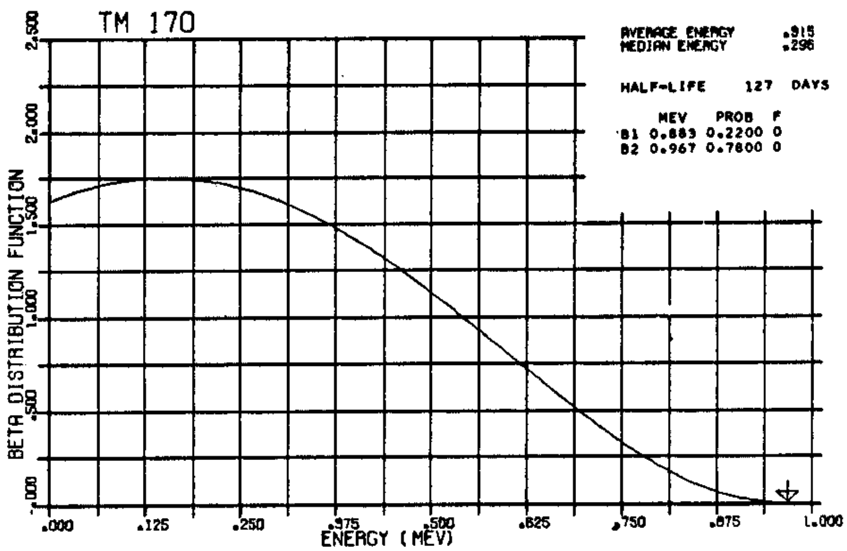
None

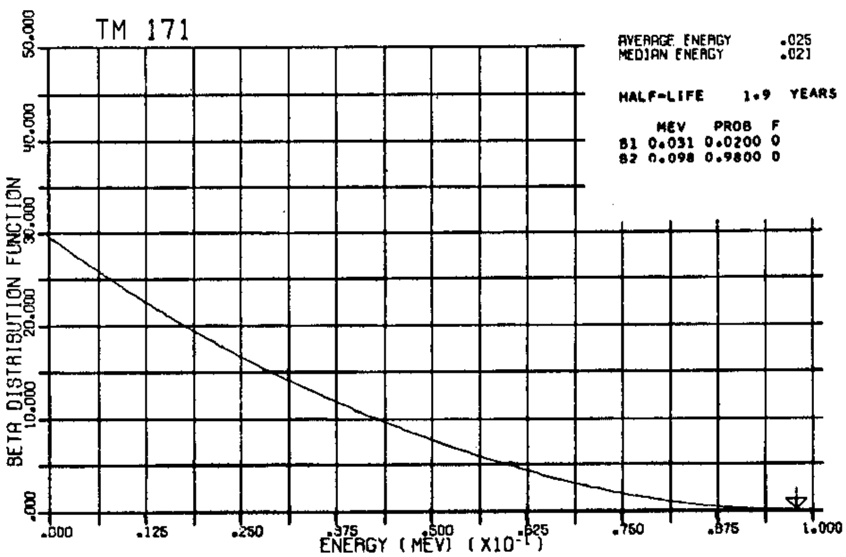
2) Beta particles 2,4,7 12,13

Isotope	Max Energy, Mev	Abundance, %	Average Energy, Mev Calc		Average Energy, Mev Meas	Milliwatts/Curie Calc	Milliwatts/Curie Meas
^{170}Tm	0.969	76	0.33	0.315	0.38	1.91	2.25 ±0.10
	0.885	24	0.30				
^{171}Tm	0.097	98	0.029	0.025		0.17	
	0.03	2	0.01				

Ref. 14

- Calculated Beta Spectrum





Ref.

3) Gamma — 7,12

Isotope	Max Energy, Mev	Abundance, %	Milliwatts/Curie, calculated	Photons / watts sec
^{170}Tm	0.0843	3.4	0.02	3.2×10^{12}
^{171}Tm	0.0667	1	0.008	1.86×10^{11}

4) Bremsstrahlung

Bremsstrahlung is the principal contributor to radiation doses. Energy distributions calculated for ^{170}Tm and ^{171}Tm oxide are shown in Section B,3,a. The metal would show a similar distribution.

5) Neutrons

None

4. Interaction with Materials of Containment — 15

No direct measurements are available for interaction with materials of construction for containers. Estimates of the extent of interaction cannot be made easily because of the lack of pertinent phase diagrams and diffusion data.

Material	Interaction
Ta	Probably nonreactive; no intermetallic compounds; occurrence of low-melting eutectics not investigated.
Re,Ru,Os	Reactivity dependent on rate of growth of known intermetallic compounds; occurrence of low-melting eutectics not investigated.
W,Mo,Cr	Occurrence of intermetallic compounds or low-melting eutectics have not been investigated.
"Super-alloys"	Reactivity dependent on rate of growth of known intermetallic compounds, such as TmFe, $TmCo_2$ and $TmNi_2$; occurrence of low-melting eutectics not investigated.

5. Thermophysical Properties

a. Density

9.318 g/cc, X-ray — 16

9.135 g/cc, measured on commercial samples (Section A,1,b.) — 4

Ref.

b. <u>Coefficient of Thermal Expansion</u>

$\bar{\alpha} = 12.4 \times 10^{-6}/^{\circ}C$ (20 to 853°C) 17

The lattice constants increase as follows:

$$a = 3.5372 + 3.19 \times 10^{-5}t - 1.41 \times 10^{-8}t^2 + 1.19 \times 10^{-11}t^3$$

$$c = 5.5619 + 12.51 \times 10^{-5}t - 3.89 \times 10^{-8}t^2 + 3.27 \times 10^{-11}t^3$$

The lattice constants are in Angstroms and the temperature is in °C (20° to 853°C)

c. <u>Specific Heat and Enthalpy</u>

See Table 1. 18

$$H^{o}_{T} - H^{o}_{298.15} = -1712 + 5.408T + 11.52T^2 - 0.65T^3 \quad (298 \text{ to } 1818^{\circ}K)$$ 18

$$= -2418 + 9.890T \quad (1818 \text{ to } 1863^{\circ}K)$$

Debye temperature: 167°K 19

Gruneisen constant: 0.94 15

TABLE 1

Thermodynamic Properties of Thulium 18

°K	$H^o_T - H^o_{298.16}$ (cal/mole)	C_P (cal/mole-deg)	S^o_T (cal/mole-deg)	$(H^o_T - H^o_{298.15})/T$ (cal/mole-deg)	$(G^o_T - H^o_{298.15})/T$ (cal/mole-deg)
Thulium					
298.15	0	6.07	17.371	0	17.37
300	12	6.08	17.41	0.04	17.37
400	631	6.30	19.19	1.58	17.61
500	1,272	6.51	20.62	2.54	18.07
600	1,933	6.72	21.83	3.22	18.60
700	2,616	6.93	22.88	3.74	19.14
800	3,318	7.13	23.82	4.15	19.67
900	4,041	7.33	24.66	4.49	20.17
1000	4,783	7.52	25.44	4.78	20.66
1100	5,544	7.71	26.17	5.04	21.13
1200	6,324	7.89	26.85	5.27	21.58
1300	7,123	8.08	27.49	5.48	22.01
1400	7,939	8.25	28.10	5.67	22.43
1500	8,773	8.43	28.67	5.85	22.82
1600	9,624	8.60	29.22	6.02	23.21
1700	10,492	8.77	29.74	6.17	23.57
1800	11,377	8.93	30.25	6.32	23.93
1818	11,537	8.96	30.34	6.35	23.99
1818	15,562	9.89	32.55	8.56	23.99
1900	16,373	9.89	32.99	8.62	24.37

	Ref.
d. Temperatures of Phase Transformations	
Melting point: 1545 ±5°C	20
Investigations of Tm-Mg alloys indicate a possible transformation from hcp to bcc (a_o = 3.92 Å) near 1300°C.	21
e. Latent Heats of Phase Transformations	
ΔH_{fusion}: 4.025 kcal/mole	18
ΔS_{fusion}: 2.21 eu	18
ΔH_{subl}: 57.44 ±0.20 kcal/mole (809° to 1219°K)	22
$\Delta H_{vap(298)}$: 59.1 kcal/mole	22,23
$\Delta S_{vap(298)}$: 28 ±2 eu	
f. Vapor Pressure	
$\text{Log } P_{mm} = (12552 \pm 45)/T + (9.1761 \pm 0.0457)$ (T in °K)	22
g. Thermal Conductivity	
0.023 ±0.005 (cal-cm)/(sec)(cm²)(°C) at 26 to 30°C, estimated from data on other rare earth metals.	15
h. Thermal Diffusivity	15

0.069 ±0.015 cm²/sec at 26 to 30°C, calculated from the estimate of thermal conductivity.

i. Viscosity

Unknown

j. Surface Tension

Unknown

k. Total Hemispherical Emittance

Unknown

l. Spectral Emissivity

Unknown

	Ref.
m. <u>Crystallography</u>	16
Hexagonal: $a_o = 3.5375 \pm 0.0001$ Å	
$c_o = 5.5546 \pm 0.0004$ Å	
A3 structure, magnesium type	
n. <u>Solubilities</u>	24
Insoluble in water.	
o. <u>Diffusion Rates</u>	
Unknown.	
p. <u>Electrical Characteristics</u>	
Electrical Resistivity: 90×10^{-6} (ohm)(cm) at 25°C	15
Hall Coefficient: $-1.8 \times 10^{-12} \frac{(\text{volt})(\text{cm})}{(\text{amp})(\text{Oer})}$ at 25°C	25
Work function: 3.12 ev	15
Electronegativity: 1.22	15

6. Mechanical Properties

	Ref.
a. <u>Hardness</u>	
48 kg/mm^2 (Vickers DPH)	15
69 Rockwell H	15
53 kg/mm^2 (Brinell, arc melted)	26
b. <u>Strength</u>	
The following values of tensile properties are estimated by comparison with data for other rare earths, as given in Reference 15.	15

- Proportional limit (0.2% offset):

  30,000 to 44,000 psi

	Ref.
• Ultimate tensile strength:	
29,000 to 44,000 psi (in tension)	
~51,800 psi (in compression)	
• Elongation: 1 to 5% in tension	
20% in compression	
• Young's Modulus: 11,500,000 psi	
• Shear Modulus: 4,400,000 psi	
• Poisson's ratio: 0.24	
Compression values are as follows.	
• Ultimate strength: 110,000 psi	26
• Yield strength (0.2%): 20,300 psi	
• Modules of compression: 5,500,000 psi	
c. Compressibility	
$\frac{\Delta V}{V_o} = 2.579 \times 10^{-6}(P-P_o) + 2.160 \times 10^{-11}(P-P_o)^2$	15

$$- 1.527 \times 10^{-18}(P-P_o)^3. \quad (P \text{ is in kg/cm}^2)$$

$P_o = 0$

$P_{max} = 40{,}000 \text{ kg/cm}^2$

Std Deviation $= 2.6 \times 10^{-4}$

Ref.

7. Chemical Properties

a. Heat and Free Energy of Formation, Entropy

Heat of formation: 0

Free energy of formation: 0

Entropy: $S_{298.15}$ = 17.371 cal/mole-deg 18

b. Chemical Reactions and Reaction Rates

These data are for unirradiated thulium and may be different for irradiated material.

Media	Temp, °C	Corrosion Rate, mils per year
Distilled H_2O	30	<1
	boiling	<1
1N H_3PO_4	30	10-100
	boiling	<1
5N H_3PO_4	30	$1\text{-}9 \times 10^3$
1N CrO_3	30	$1\text{-}9 \times 10^3$
	boiling	$1\text{-}9 \times 10^2$
1N $H_2C_2O_4$	30	1-10
	boiling	1-10
1:1 (48%) HF - (65%) HNO_3	25	<1
1N NaOH	30	<1
1N NH_4OH	30	<1
1N $NaNO_2$	30	1-10
	boiling	<1

Oxidation proceeds with a linear rate from 100 to 800°C; moisture accelerates the reaction. Rate constants for oxidation in dry air are: 28

Temp, °C	Const., mg/(cm²)(min)
200	6.4×10^{-5}
300	3.4×10^{-4}
400	5.0×10^{-4}
500	6.8×10^{-4}
700	2.2×10^{-3}

Activation energy is 3.1 kcal from 300 to 700°C.

8. Biological Tolerances

Ref.

Maximum Permissible Concentrations of Tm for Continuous Occupational Exposure 29

Isotope	Form	Organ of Reference	Max Permissible Body Burden (Total Body), µCi	Max Permissible Conc. µCi/cm³ For 40-hr week Water	Air	For 168-hr wk Water	Air
^{170}Tm	Soluble	GI[(a)]		10^{-3}	3×10^{-7}	5×10^{-4}	10^{-7}
		Bone	9	0.8	4×10^{-8}	0.3	10^{-8}
		Kidney	30	4	2×10^{-7}	1	6×10^{-8}
		Total body	60	5	2×10^{-7}	2	7×10^{-8}
	Insoluble	Lung		-	3×10^{-8}	-	10^{-8}
		GI[(a)]		10^{-3}	2×10^{-7}	5×10^{-4}	8×10^{-8}
^{171}Tm	Soluble	GI[(a)]		0.01	3×10^{-6}	5×10^{-3}	10^{-6}
		Bone	90	3	10^{-7}	0.9	4×10^{-8}
		Kidney	700	20	8×10^{-7}	6	3×10^{-7}
		Total body	700	20	8×10^{-7}	6	3×10^{-7}
	Insoluble	Lung		-	2×10^{-7}	-	8×10^{-8}
		GI[(a)]		0.01	3×10^{-6}	5×10^{-3}	9×10^{-7}

(a) Gastrointestinal.

Maximum Permissible Inhalation of Tm for Short-Term Exposure 30

(Pertinent to accident situations)

Isotope	Critical Organ	Effective Half-life, days	µCi Inhaled That Will Present a Dose of: 0.3 rem in 1 wk	15.7 rem in 1 yr	150 rem in 70 yr
^{170}Tm + ^{170}Yb	Bone	113	22	54	4.61×10^{2}
^{171}Tm	Bone	410	2.45×10^{2}	3.28×10^{2}	1.44×10^{3}

9. Shielding Data

The contributions, of bremsstrahlung from ^{171}Tm and gamma radiation from ^{171}Yb, to dose rates from a ^{170}Tm heat source are small compared to the bremsstrahlung from ^{170}Tm decay because of the low energy for ^{171}Tm decay and its low concentration in irradiated ^{169}Tm. Further, the gamma radiation from ^{170}Yb is negligible because of its low energy and internal conversion. To a first approximation, shielding requirements for ^{170}Tm fuel are determined only by bremsstrahlung from β-decay of ^{170}Tm.

Calculated gamma plus bremsstrahlung dose rates for unshielded, iron-shielded, lead-shielded, and uranium-shielded sources are shown in Fig. 1-4 for ^{170}Tm sources from 0.1 to 20 kw. Dose rates for unshielded and iron-shielded ^{171}Tm sources are shown in Fig. 5 and 6. 12

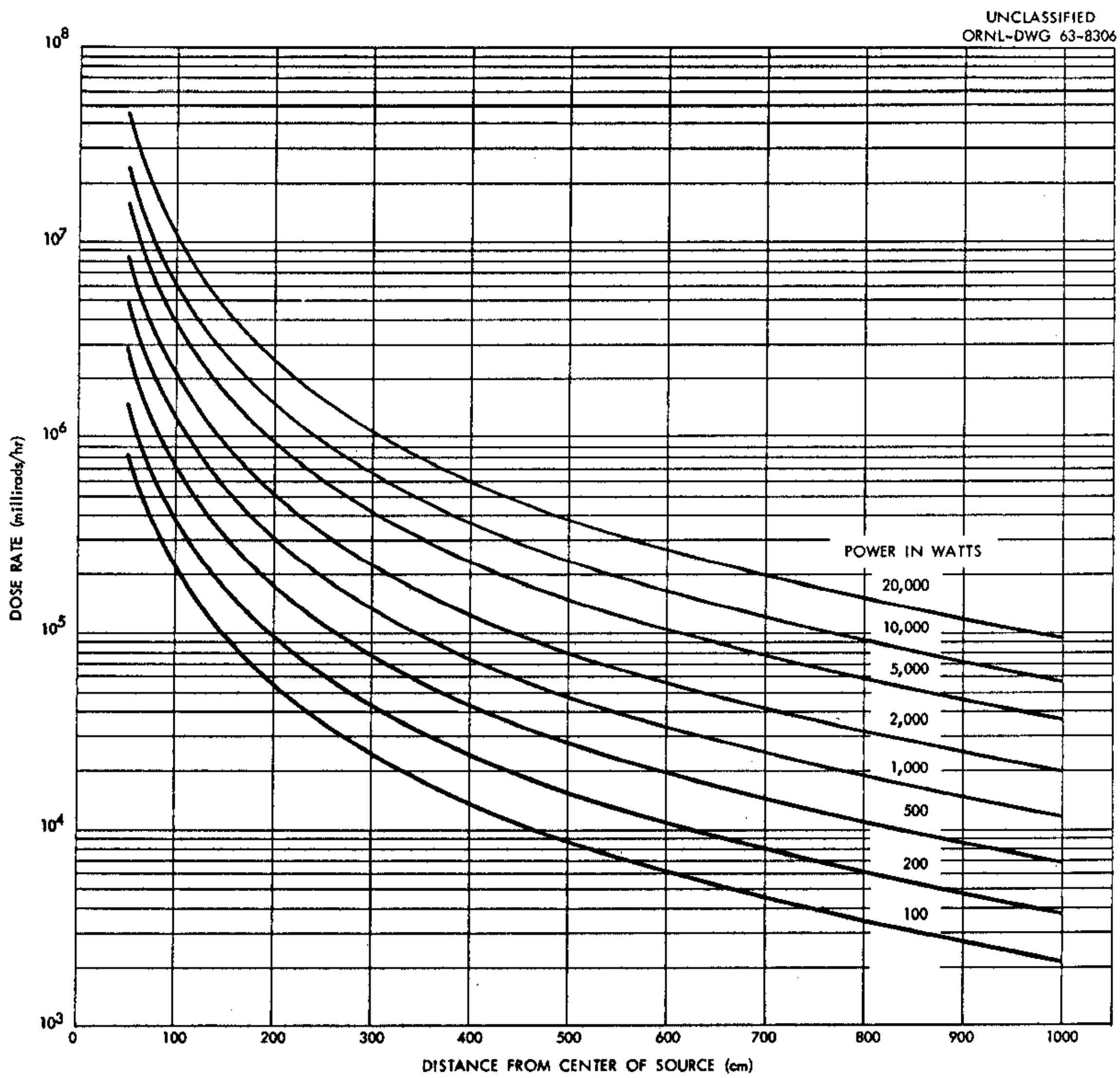


FIG. 1 GAMMA PLUS BREMSSTRAHLUNG DOSE RATES FROM UNSHIELDED ISOTOPIC POWER SOURCES OF THULIUM-170 AS A FUNCTION OF DISTANCE FROM CENTER OF SOURCE [12]

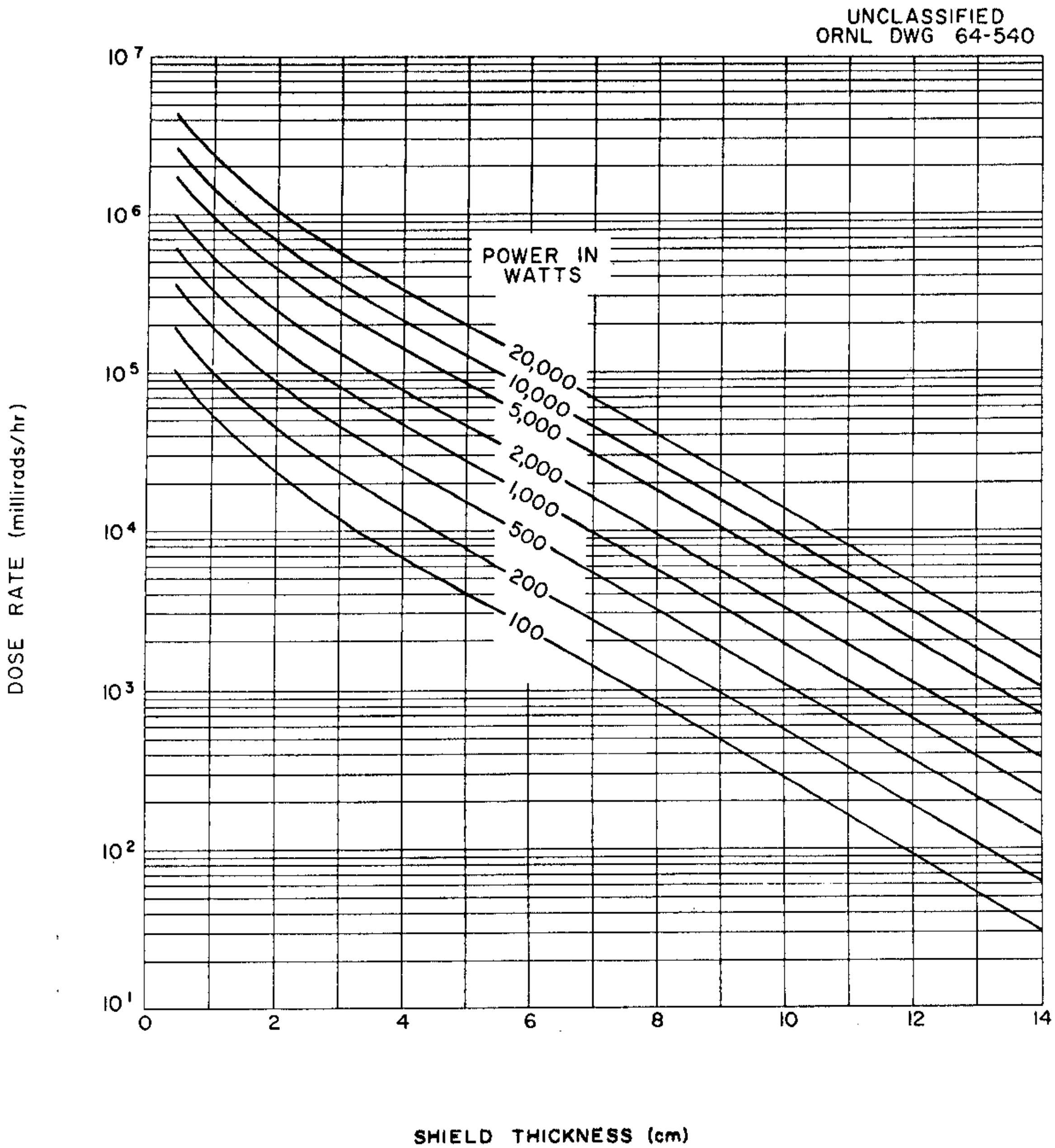


FIG. 2 GAMMA PLUS BREMSSTRAHLUNG DOSE RATES FROM IRON-SHIELDED ISOTOPIC POWER SOURCES OF THULIUM-170. CENTER OF SOURCE TO DOSE POINT SEPARATION DISTANCE = 100 cm [12]

UNCLASSIFIED
ORNL DWG 64-541

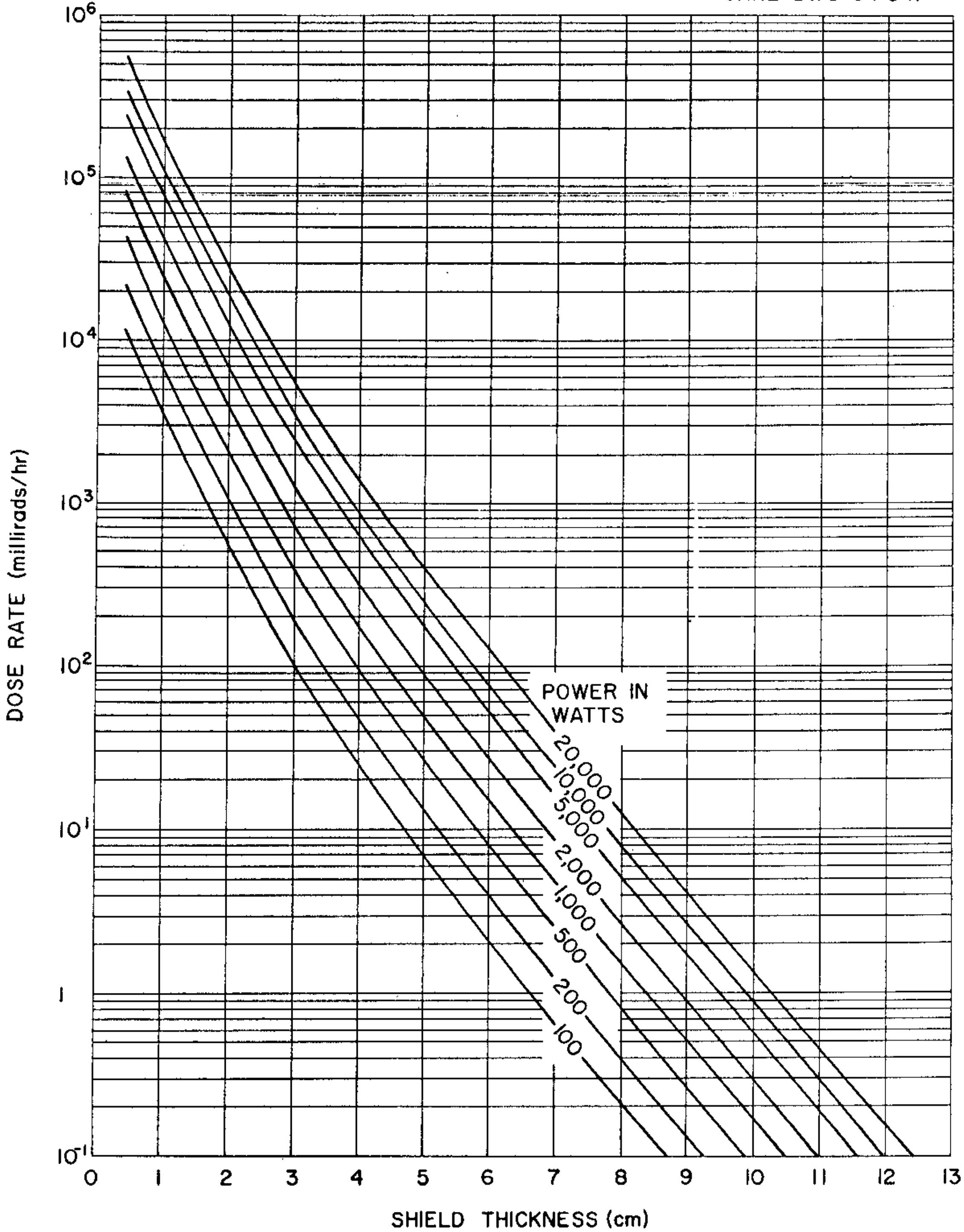


FIG. 3 GAMMA PLUS BREMSSTRAHLUNG DOSE RATES FROM LEAD-SHIELDED ISOTOPIC POWER SOURCES OF THULIUM-170. CENTER OF SOURCE TO DOSE POINT SEPARATION DISTANCE = 100 cm [12]

UNCLASSIFIED
ORNL DWG 64-542

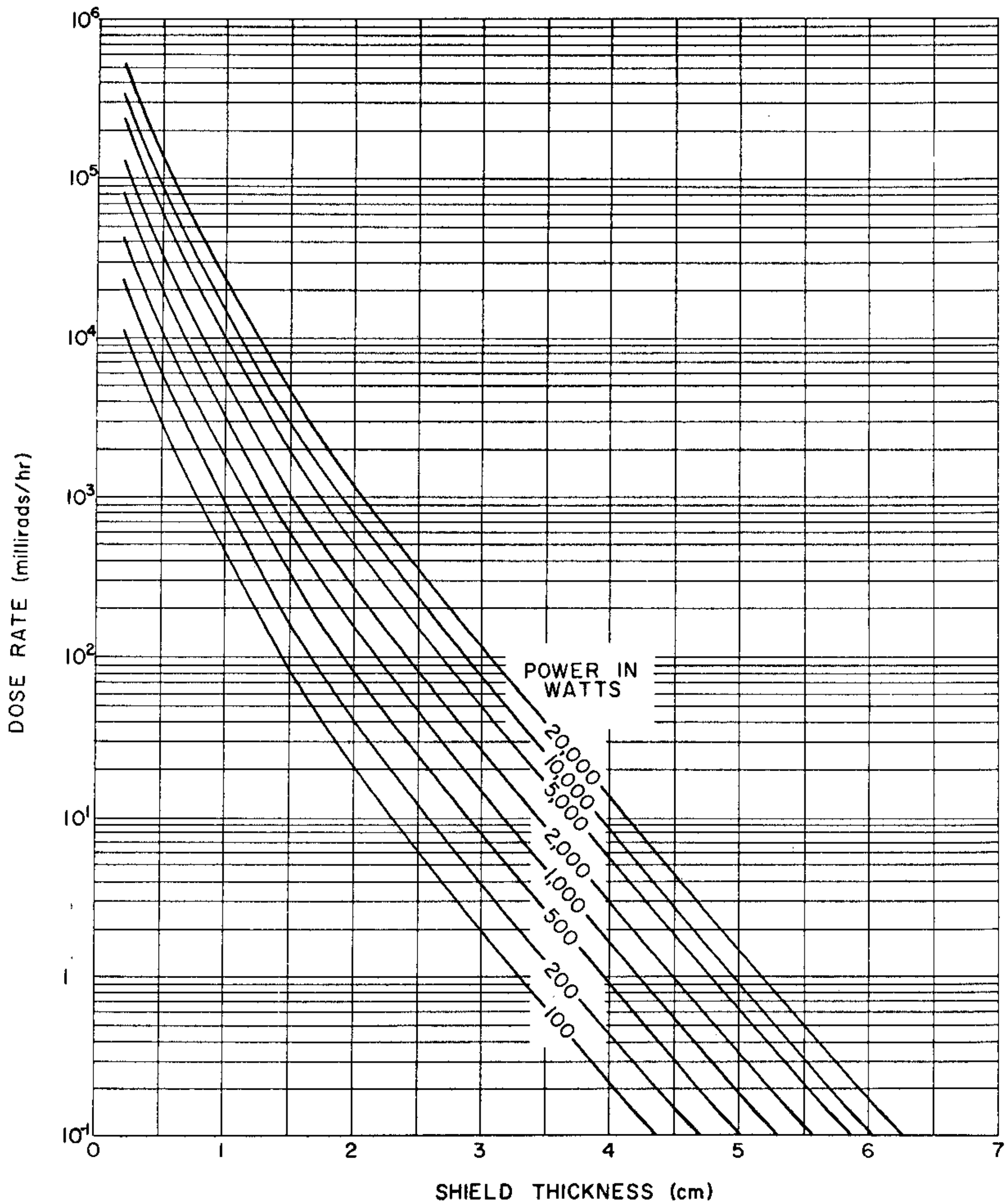


FIG. 4 GAMMA PLUS BREMSSTRAHLUNG DOSE RATES FROM URANIUM-SHIELDED ISOTOPIC POWER SOURCES OF THULIUM-170. CENTER OF SOURCE TO DOSE POINT SEPARATION DISTANCE = 100 cm [12]

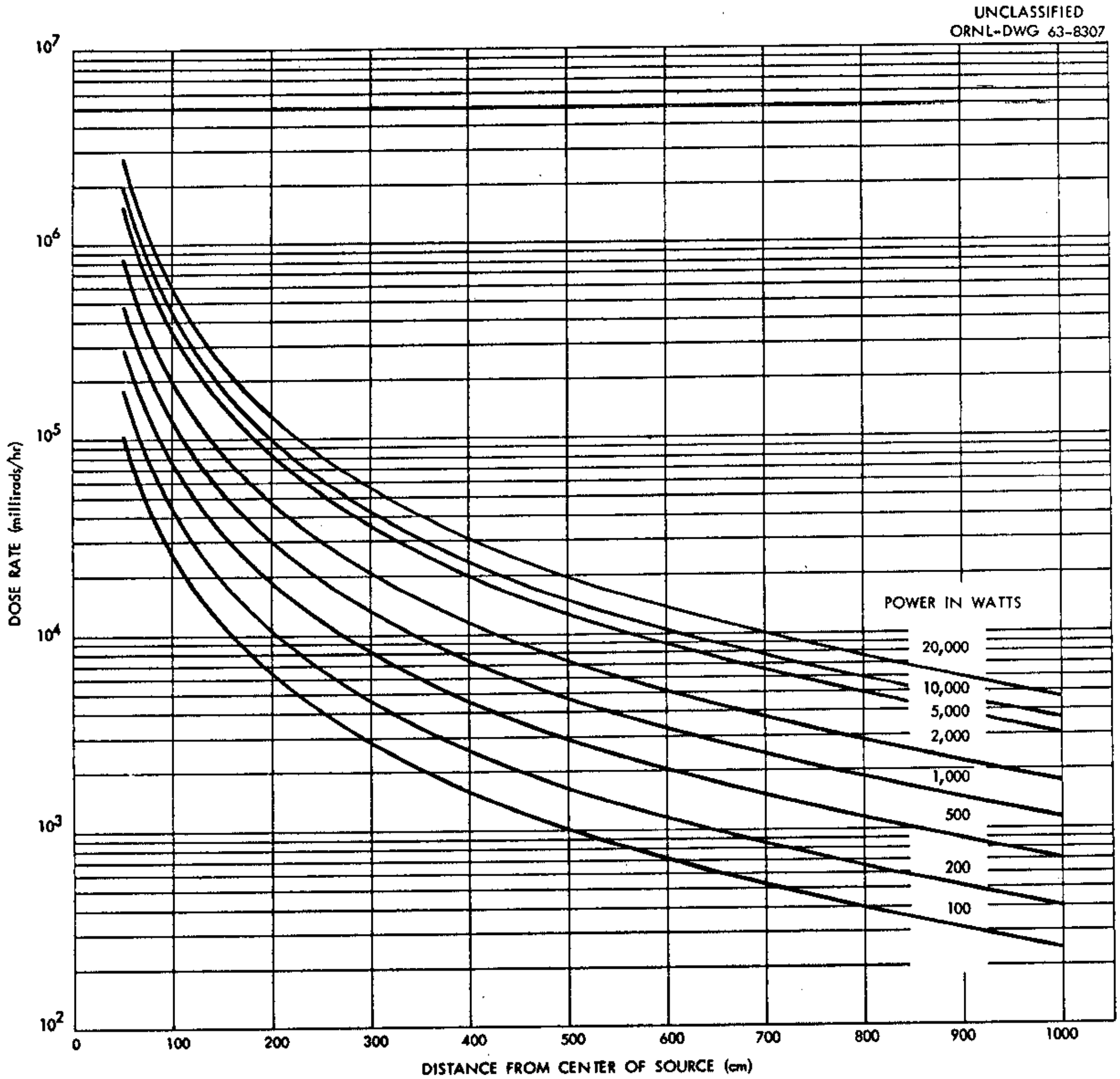


FIG. 5 GAMMA PLUS BREMSSTRAHLUNG DOSE RATES FROM UNSHIELDED ISOTOPIC POWER SOURCES OF THULIUM-171 AS A FUNCTION OF DISTANCE FROM CENTER OF SOURCE [12]

UNCLASSIFIED
ORNL DWG 64-851

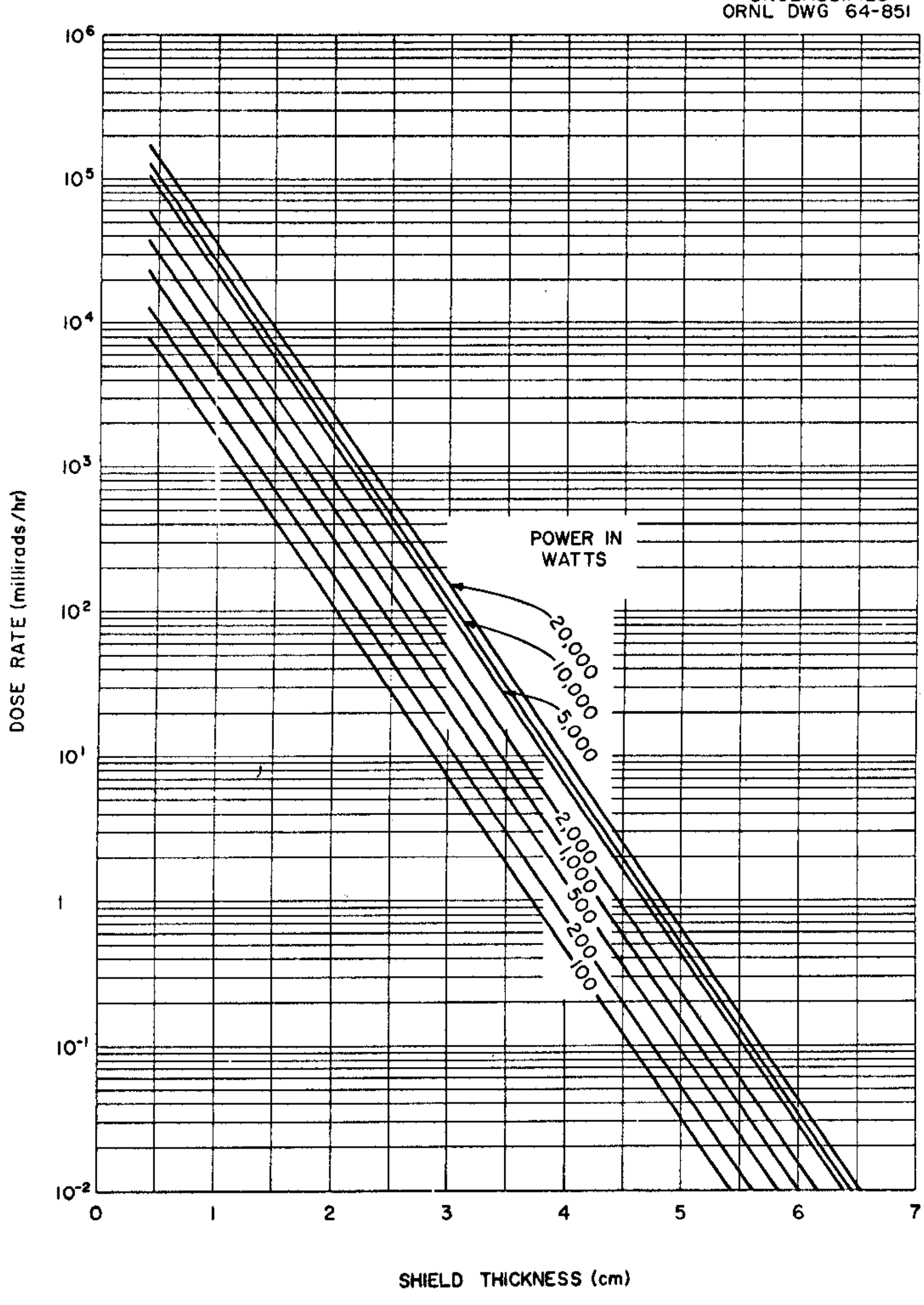


FIG. 6 GAMMA PLUS BREMSSTRAHLUNG DOSE RATES FROM IRON-SHIELDED ISOTOPIC POWER SOURCES OF THULIUM-171 (12)

Center of source to dose point separation distance = 100 cm. Estimate required thickness of lead or uranium shield by using the following relationship: 1 cm of iron = 0.039 cm lead and 0.137 cm of uranium for 0.1-Mev gamma rays

Ref.

B. THULIUM OXIDE - Tm_2O_3

1. Composition

The composition of unirradiated wafers is 99.9% $Tm_{2.00}O_{3.00}$. At temperatures over 1500°C in vacuum, inert gas, or hydrogen, the unirradiated thulium oxide will lose a small amount of oxygen to a composition of $Tm_{2.00}O_{2.90}$. 4

a. Radionuclide Abundance (calculated) for an arbitrarily selected flux of 4×10^{14} n/(cm²)(sec) at reactor discharge for ^{169}Tm irradiation). 4

Isotope	Specific Activity, w/g → Concentration, wt % 0.62	2.24	2.94
^{169}Tm	83.5	68.2	53.1
^{170}Tm	4.0	14.3	18.8
^{171}Tm	-	2.5	7.3
^{170}Yb	-	2.5	7.9
^{171}Yb	-	-	0.4
$^{16,17,18}O$	12.4	12.4	12.4

b. Chemical Impurities (commercial samples before irradiation) 4

Impurity	Range of Conc, wt %
Yb (natural)	0.003 to 0.056
Er	0.001 to 0.052
Si	0.014 to 0.029
Fe	0.003 to 0.019
Ca	0.006 to 0.013
Y	0.005 to 0.009
Al	<0.001 to 0.009
All other impurities	<0.001

Ref.

2. Specific Power

- 2 to 3 w/g of oxide at reactor discharge is feasible 1,4
- 800 to 1200 Ci/g of oxide at reactor discharge is feasible 1,4
- 445 ±2 Ci/w of ^{170}Tm measured on typical irradiated wafers 4

3. Radiation

The gamma and beta radiation is given in Section A,3,b,2).

a. Bremsstrahlung

Calculated bremsstrahlung distributions for the two betas of $^{170}Tm_2O_3$ and for $^{171}Tm_2O_3$ are shown in Tables 2-4. 12

The gamma emission rate from thulium oxide matrices, including bremsstrahlung from the 0.969- and 0.885-Mev ^{170}Tm betas and the 0.1-Mev ^{171}Tm beta, is given in the following table. 12

	Photon Energy, Mev	Photons/w sec
^{170}Tm	0.2 Bremsstrahlung	7.465×10^{11}
	0.475 Bremsstrahlung	5.01×10^{10}
	0.775 Bremsstrahlung	1.158×10^{9}
	0.1 Actual gamma	3.208×10^{12}
^{171}Tm	0.1 Bremsstrahlung	2.325×10^{11}
	0.1 Actual gamma	1.86×10^{12}

Ref.

TABLE 2

12

Production of Bremsstrahlung Photons from Thulium-170 Beta 1 in Thulium Oxide Matrix

Maximum beta-particle energy, Mev 0.968
Average beta-particle energy, Mev 0.326

Bremsstrahlung Energy Group (Mev)	Number of Photons per Beta Particle Within ΔE Energy Group
0.050 ±0.025	5.736×10^{-2}
0.100 ±0.025	2.085×10^{-2}
0.150 ±0.025	1.026×10^{-2}
0.200 ±0.025	5.686×10^{-3}
0.250 ±0.025	3.340×10^{-3}
0.300 ±0.025	2.020×10^{-3}
0.350 ±0.025	1.236×10^{-3}
0.400 ±0.025	7.551×10^{-4}
0.450 ±0.025	4.562×10^{-4}
0.500 ±0.025	2.696×10^{-4}
0.550 ±0.025	1.542×10^{-4}
0.600 ±0.025	8.410×10^{-5}
0.650 ±0.025	4.296×10^{-5}
0.700 ±0.025	2.000×10^{-5}
0.750 ±0.025	8.138×10^{-6}
0.800 ±0.025	2.680×10^{-6}
0.850 ±0.025	6.111×10^{-7}
0.900 ±0.025	6.278×10^{-8}
0.950 ±0.025	2.196×10^{-10}
Total bremsstrahlung energy, Mev/beta particle	1.033×10^{-2}

Ref.

TABLE 3

Calculated Production of Bremsstrahlung Photons from Thulium-170 Beta 2 in Thulium Oxide Matrix 12

Maximum beta-particle energy, Mev 0.884
Average beta-particle energy, Mev 0.293

Bremsstrahlung Energy Group (Mev)	Number of Photons per Beta Particle Within ΔE Energy Group
0.050 ±0.025	4.999×10^{-2}
0.100 ±0.025	1.765×10^{-2}
0.150 ±0.025	8.429×10^{-3}
0.200 ±0.025	4.522×10^{-3}
0.250 ±0.025	2.562×10^{-3}
0.300 ±0.025	1.487×10^{-3}
0.350 ±0.025	8.674×10^{-4}
0.400 ±0.025	5.011×10^{-4}
0.450 ±0.025	2.828×10^{-4}
0.500 ±0.025	1.538×10^{-4}
0.550 ±0.025	7.907×10^{-5}
0.600 ±0.025	3.753×10^{-5}
0.650 ±0.025	1.584×10^{-5}
0.700 ±0.025	5.592×10^{-6}
0.750 ±0.025	1.459×10^{-6}
0.800 ±0.025	2.091×10^{-7}
0.850 ±0.025	4.896×10^{-9}
Total bremsstrahlung energy, Mev/beta particle	8.310×10^{-3}

Ref.

TABLE 4

Calculated Production of Bremsstrahlung Photons from Thulium-171 Beta in Thulium Oxide Matrix 12

Maximum beta-particle energy, Mev 0.097
Average beta-particle energy, Mev 0.029

Bremsstrahlung Energy Group (Mev)	Number of Photons per Beta Particle Within ΔE Energy Group
0.010 ±0.005	1.632×10^{-3}
0.020 ±0.005	4.124×10^{-4}
0.030 ±0.005	1.367×10^{-4}
0.040 ±0.005	4.785×10^{-5}
0.050 ±0.005	1.599×10^{-5}
0.060 ±0.005	4.632×10^{-6}
0.070 ±0.005	9.973×10^{-7}
0.080 ±0.005	1.068×10^{-7}
0.090 ±0.005	0.000
Total bremsstrahlung energy, Mev/beta particle	3.126×10^{-5}

Ref.

4. Interaction with Materials of Containment 4

Preliminary evaluations of rare earth oxide interactions with tungsten, tantalum, molybdenum, and rhenium, and their alloys indicate that Tm_2O_3 probably will not react with these metals up to 2000°C in sealed systems.

5. Thermophysical Properties

a. Density

8.884 g/cc at 25°C, X-ray 31

b. Coefficient of Thermal Expansion

The following values were derived by Stecura and Campbell from the lattice parameter changes for Tm_2O_3 from 0°C, as determined by high-temperature X-ray diffraction. The (10.6.2) and (12.4.2) hkl reflections were used to calculate the expansion. The maximum deviation between values calculated from each line was ±2% above 500°C. 32

Temperature, °C	Linear Expansion, %	Linear Expansion Coefficient (multiply by 10^{-6})
0	0.00	-
28	0.019	6.8
134	0.090	6.6
264	0.189	7.1
364	0.272	7.4
477	0.360	7.5
575	0.444	7.7
695	0.546	7.8
814	0.652	8.0
911	0.743	8.1
1031	0.849	8.2
1132	0.943	8.3
1196	1.005	8.4
1271	1.084	8.5
1295	1.104	8.5

Ref.

The following values were derived by W. C. Mosley at the Savannah River Laboratory from lattice expansion of Tm_2O_3 from 28°C using high temperature X-ray diffraction. Over thirty diffraction peaks were used to calculate the lattice constant at each temperature. 4

Temperature, °C	Linear Expansion, %
28	0.00
535	0.51
1135	0.92
1380	1.17
1745	1.48

A value of 6.3×10^{-6}/°C has been reported for the linear expansion coefficient of a 97% dense ceramic, but the temperature range was not given. 33

c. Specific Heat and Enthalpy

- Heat capacity:

$$C_p = 31.00 + 0.78 \times 10^{-3}T - 3.42 \times 10^{5}T^{-2}$$ cal/(mole)(deg) (298 to 1680°K) 34

$$C_p = 32.00$$ cal/mole (1680 to 2000°K) 34

$$C_p = 30.58 + 2.32 \times 10^{-3}T - 4.07 \times 10^{5}T^{-2}$$ cal/(mole)(deg) (298.1 to 1606.5°K) 35

- Heat content:

$$H_T - H_{298.15} = 31.00T + 0.39 \times 10^{-3}T^2 + 3.42 \times 10^{5}T^{-1} - 10{,}424$$ cal/mole (±0.1%; 298 to 1680°K) 34

$$H_T - H_{298.15} = 32.00T - 10{,}490$$ cal/mole (±0.1%; 1680 to 2000°K) 34

$$H_T - H_{298} = 30.58T + 1.16 \times 10^{-3}T^2 + 4.07 \times 10^{5}T - 10{,}584$$ cal/mole (298.1 to 1606.5°K) 35

Ref.

d. Temperatures of Phase Transformations

Cubic to hexagonal: 2280°C 36

Melting point: 2375 ±25°C, estimated by analogy with other rare earth sesquioxides. 37

Boiling point: 4150 ±200°C 37

e. Latent Heats of Phase Transformations

Unknown.

f. Vapor Pressure

Approximately 10^{-6} atm at 2300°K. 38

The vapor pressure has been estimated within a range of three decades to be 37

$$\text{Log } P_T(\text{atm}) = 7.36 - 31730T^{-1} (T \text{ in } ^\circ K)$$

This relationship is for the predominant vaporization process, formation of the gaseous elements. Calculated pressures at three temperatures are listed in the following table.

Temperature, °K	Pressure, atm
2000	3.2×10^{-9}
2500	4.4×10^{-6}
3000	6.0×10^{-4}

g. Thermal Conductivity

Eastly and Matson report a value for thermal conductivity of 0.125 (cal-cm)/(sec)(cm^2)(°C) at 500°C. However, this value appears too high by a factor of 10 when compared to most other oxides. A value of 1.3 to 4.0 (Btu)(ft)/(ft^2)(hr)(°F) [0.006 to 0.019 (cal)(cm)/(cm^2)(sec)(°C) at 1000°C] was estimated based on values for yttria and other oxides as shown in Fig. 7. 33,39

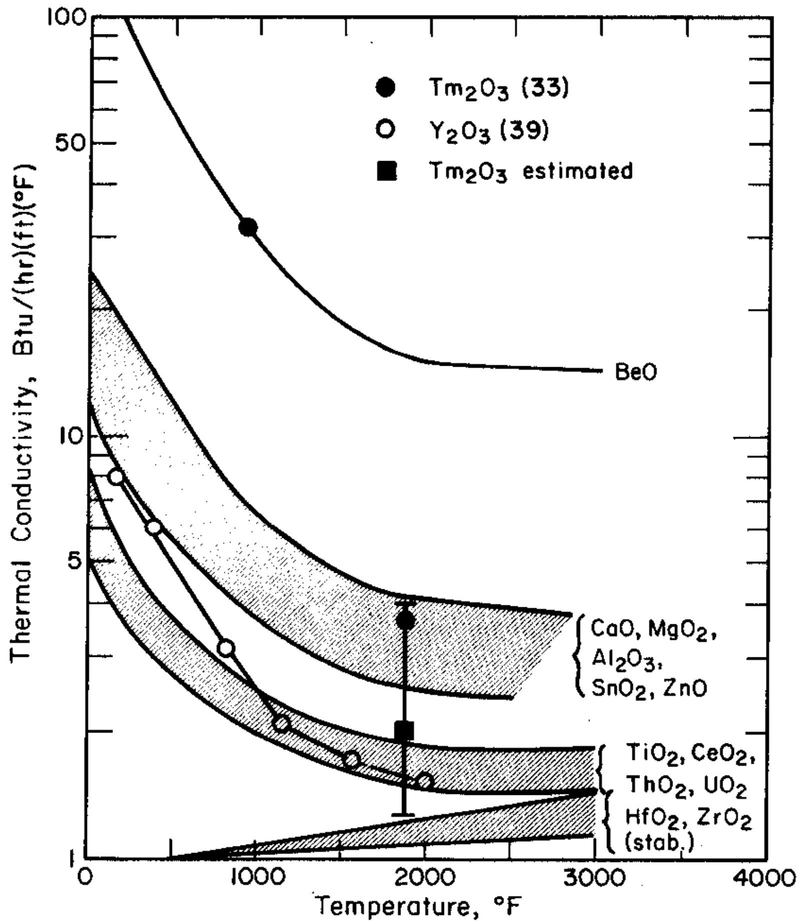


FIG. 7 THERMAL CONDUCTIVITY OF Tm_2O_3 AND OTHER OXIDES
(Figure and data for other oxides from Reference 39)

h. Thermal Diffusivity | Ref.

0.009 to 0.027 cm^2/sec at 1000°C calculated from thermal conductivity estimate of Section B.6.g.

i. Viscosity

Unknown.

j. Surface Tension

Unknown.

k. Total Hemispherical Emittance

Unknown

l. Spectral Emissivity

Unknown.

m. Crystallography

- Cubic: Thallium oxide type structure (rare earth oxide type C), space group Ia3 (No. 206), 16 molecules per unit cell. 31

  a = 10.488 Å at 25°C for 99.9% pure, stoichiometric, Tm_2O_3 annealed at 1100°C for 16 hours.

- Hexagonal (2280°C to melting): 36

  c = 6.04 Å at 2280°C

  Assuming c/a = 1.60 as for Y_2O_3 at 2380°C and Er_2O_3 at 2300°C, one would have for Tm_2O_3 a = 3.78 Å at 2280°C

- Monoclinic: a = 13.18 Å, b = 3.447 Å, 40

  c = 8.505 Å, β = 100.2°.

  Stable above 1100°C at 20 kb pressure and above 400°C at 40 kb pressure.

n. Solubility

Soluble in mineral acids 24

Low solubility in water

Ref.

e. Diffusion Rates

Unknown.

6. Mechanical Properties

a. Hardness

Unknown.

b. Crush Strength

o Modulus of rupture: 20,000 psi on 97% dense ceramic 33

7. Chemical Properties

a. Heat and Free Energy of Formation, Entropy

$\Delta H^o_{f_{298}} = -451.4 + 1.4$ kcal/mole 41

$S^o_{298} = 36.3 \pm 0.3$ eu (estimated from data on rare earth oxides) 42

b. Heat and Free Energy of Vaporization to Gaseous Atoms, Entropy 37

$\Delta H^o_{v_{298}} = 748.3$ kcal/mole

$\Delta S^o_{v_{298}} = 169 \pm 5$ eu

c. Chemical Reactions and Reaction Rates 4

No measurable reaction with oxygen, nitrogen, water, steam, or hydrogen.

8. Biological Tolerances

Radiobiological tolerances are given in Section A,8.

9. Shielding Data

Dose rates for unshielded, iron-shielded, lead-shielded, and uranium-shielded sources are specified in Section A,9. Computer calculations of the fraction of bremsstrahlung absorbed internally in pure $^{170}Tm_2O_3$ and pure $^{171}Tm_2O_3$ are shown in Fig. 8 and 9. 13

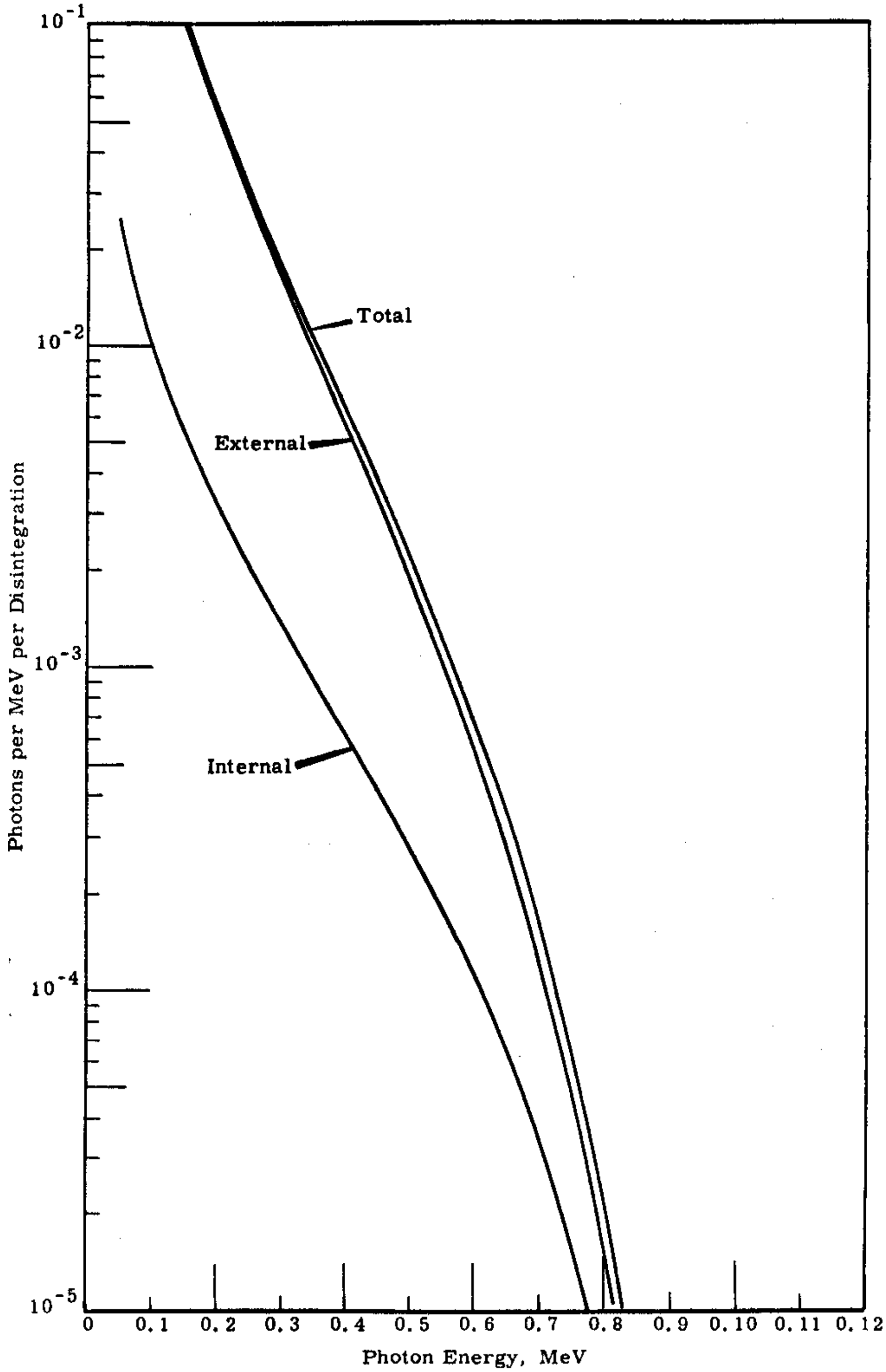


FIG. 8 BREMSSTRAHLUNG FROM ^{170}Tm BETAS STOPPED IN THULIUM OXIDE [13]

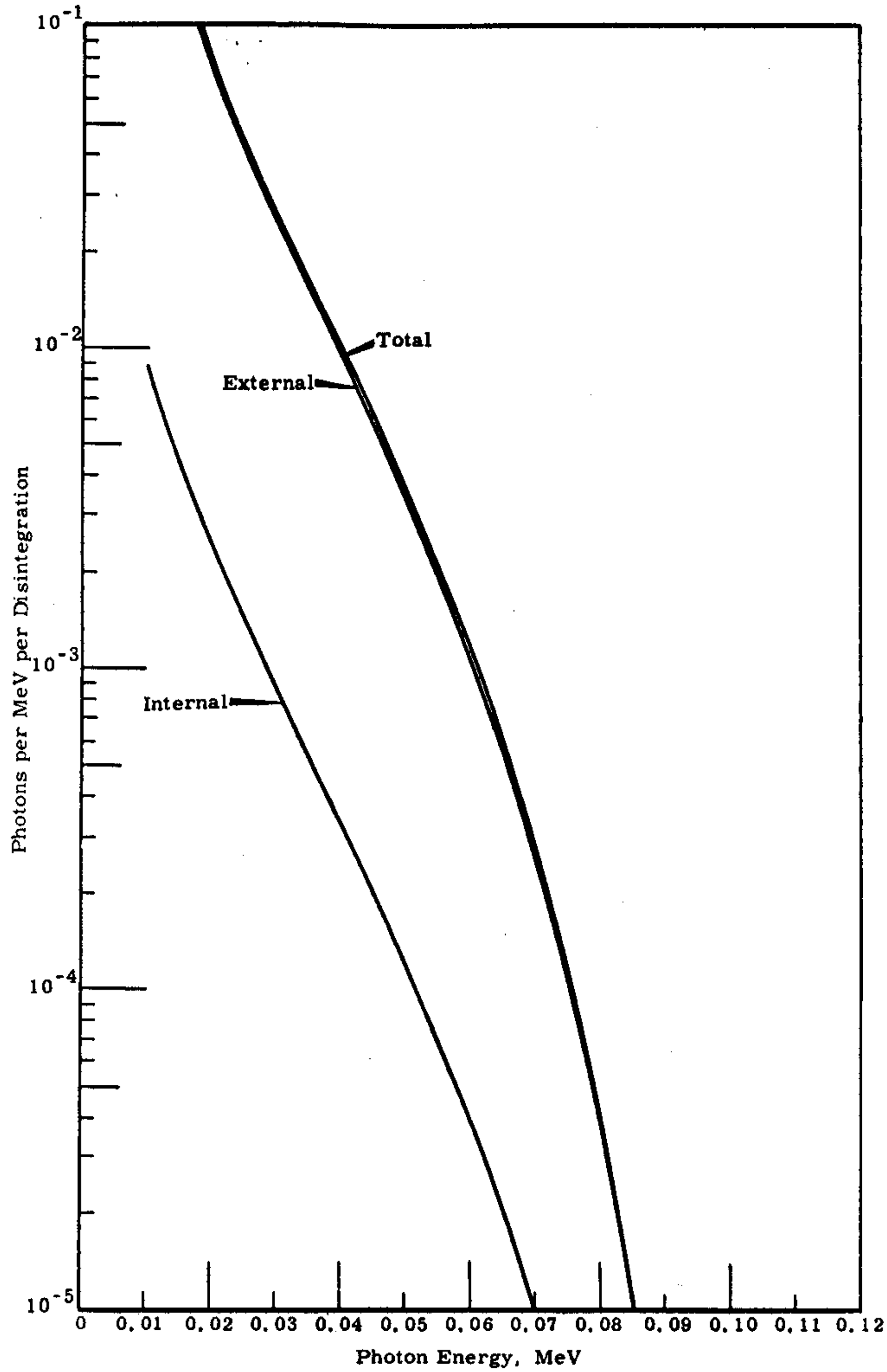


FIG. 9 BREMSSTRAHLUNG FROM ^{171}Tm BETAS STOPPED IN THULIUM OXIDE [13]

REFERENCES

1. Thulium metal and oxide were produced at these activity levels in experimental irradiations in "The Savannah River High Flux Demonstration." See USAEC Report DP-999, E. I. du Pont de Nemours and Co., Savannah River Laboratory, Aiken, S. C. (1965).

2. W. J. Kerrigan. "Calorimetrically Measured Half-Life of Thulium-170." J. Inorg. Nucl. Chem., in press (1967).

3. K. F. Flynn, L. E. Glendenin, and E. P. Steinberg. "Half-Life Determinations by Direct Decay." Nucl. Sci. and Eng. 22, 416-19 (1965).

4. Previously unpublished data, Savannah River Laboratory.

5. D. Strominger, J. M. Hollander, and G. T. Seaborg. "Table of Isotopes." Rev. Mod. Phys. 30, 585-904 (1958).

6. A. M. Hellwege and K. H. Hellwege. "Energy Levels of Nuclei: A = 5 to A = 257." Landolt-Bornstein, I, Springer-Verlag, Berlin (1961).

7. Nuclear Data Sheets, 1959-1965. pp 1867, 1868, 1887. Academic Press Inc., New York (1966).

8. R. S. Dingus, W. L. Talbert, Jr., and E. N. Hatch. Measurements of Some Internal Conversion Coefficients Using Scintillation Counter, Coincidence Techniques. USAEC Report IS-1032, Iowa State University, Ames Laboratory, Ames, Iowa (1965).

9. R. L. Graham, J. L. Wolfson, and R. E. Bell. "The Disintegration of Tm^{170}." Canadian J. Phys. 30, 459-75 (1952).

10. E. N. Hatch, G. W. Eakins, G. C. Nelson, and R. E. McAdams. "Method for Measuring Internal Conversion Coefficients Using a Bent Crystal Gamma-Ray Monochromator and a Magnetic Electron Spectrometer." Internal Conversion Processes. pp 183-192. J. H. Hamilton, Academic Press, New York, New York, 669 pages (1966).

11. J. F. W. Jansen and A. H. Wapstra. "Determination of the Conversion Coefficients in ^{170}Yb with Scintillation Methods." "Internal Conversion Processes. pp 237-247. Academic Press, New York, New York, 669 pages (1966).

12. E. D. Arnold. Handbook of Shielding Requirements and Radiation Characteristics of Isotopic Power Sources for Terrestrial, Marine, and Space Applications. USAEC Report ORNL-3576, Oak Ridge National Laboratory, Tenn. (1964).

13. H. H. Van Tuyl. Bremrad-A Computer Code for External and Internal Bremsstrahlung Calculations. USAEC Report HW-83784. General Electric Co., Hanford Atomic Products Operation, Richland, Wash. (1964).

14. O. H. Hogan, P. E. Zigman, and J. L. Mackin. Beta Spectra. II. Spectra of Individual Negatron Emitters. USAEC Report USNRDL-TR-802, U. S. Naval Radiological Defense Laboratory, San Francisco, Calif. (1964).

15. K. A. Gschneidner. Rare Earth Alloys. D. Van Nostrand Co., Inc., Princeton, N. J. (1961).

16. F. H. Spedding, A. H. Daane, and K. W. Herrmann. "The Crystal Structures and Lattice Parameters of High-Purity Scandium, Yttrium and the Rare Earth Metals." Acta Cryst. 9, 559-63 (1956).

17. F. H. Spedding, J. J. Hanak, and A. H. Daane. Cited by Ref. 15.

18. D. H. Dennison, K. A. Gschneidner, Jr., and A. H. Daane. "High-Temperature Heat Contents and Related Thermodynamic Functions of Eight Rare-Earth Metals: Sc, Gd, Tb, Dy, Ho, Er, Tm, and Lu." J. Chem. Phys. 44, 4273-82 (1966).

19. Ames Laboratory Staff. Semi-Annual Summary Research Report in Chemistry for January - June 1958. USAEC Report ISC-1049, Ames Laboratory, Iowa State College, Ames, Iowa (1958).

20. A. H. Daane. "Physical Properties of the Rare Earth Metals." Paper presented at Rare Earth Symposium, ASM Annual Meeting, Chicago, Ill. Cited by Ref. 15. (November 1959).

21. A. E. Miller and A. H. Daane. "The High-Temperature Allotropy of Some Heavy Rare-Earth Metals." Trans AIME 230, 568-72 (1964).

22. F. H. Spedding, R. J. Barton, and A. H. Daane. "The Vapor Pressure of Thulium Metal." J. Am. Chem. Soc. 79, 5160-63 (1957).

23. C. E. Habermann and A. H. Daane. "Vapor Pressures of the Rare-Earth Metals." J. Chem. Phys. 41, 2818-27 (1964).

24. Handbook of Chemistry and Physics. 47th Ed. Chemical Rubber Co. (1966-67).

25. G. S. Anderson, S. Legvold, and F. H. Spedding. "Hall Effect in Lu, Yb, Tm, and Sm." Phys. Rev. 111, 1257-8 (1958).

26. B. Love, comp. "Properties of Rare Earth Metals" in Metals Handbook. Vol. I. 8th ed., pp 1230-1, American Society for Metals. Cleveland, Ohio (1961).

27. L. Lee and N. D. Greene. "Corrosion Characteristics of the Rare Earth and Yttrium Metals." Corrosion 20, 145t-149t (1964).

28. N. D. Greene and F. G. Hodge. "Oxidation Characteristics of the Lanthanide Metals." Corrosion 22, 206-13 (1966).

29. K. Z. Morgan, W. S. Snyder, and J. A. Auxier (Editors). "Report of ICRP Committee II on Permissible Dose for Internal Irradiation (1959), with Bibliography for Biological, Mathematical, and Physical Data. Health Physics 3, 380 pages (June 1960).

30. Calculations performed by T. W. Fowler at the Savannah River Laboratory that extend work of Morgan, et al. (K. Z. Morgan, W. S. Snyder, and M. R. Ford. "Maximum Permissible Concentration of Radioisotopes in Air and Water for Short Period Exposure." Proc. U. N. Intern. Conf. Peaceful Uses Atomic Energy, 2nd Geneva, 13, 139-158 (1955).)

31. H. E. Swanson, M. I. Cook, T. Issacs, and E. H. Evans. Standard X-Ray Diffraction Powder Patterns Data for 43 Substances. U. S. National Bureau of Standards Circular 539, Vol. 9. U. S. Department of Commerce. Washington, D. C. (1960).

32. S. Stecura and W. J. Campbell. Thermal Expansion and Phase Inversion of Rare-Earth Oxides. USAEC Report BM-RI-5847, Bureau of Mines, College Park Metallurgy Research Center, Md. (1960).

33. M. R. Eastly and M. W. Matson. "Engineering Properties of Tm_2O_3." Abstracted in Amer. Ceram. Soc. Bull. 44, (4), 389 (1965).

34. L. B. Pankratz and E. G. King. High-Temperature Heat Contents and Entropies of the Sesquioxides of Erbium, Holmium, Thulium, and Ytterbium. USAEC Report BM-RI-6175, Bureau of Mines, Berkeley Thermodynamics Lab., Berkeley, Calif. (1962).

35. D. Sh. Tsagareishvili and G. G. Gvelesiani. "Enthalpies and Heat Capacities of Oxides of Rare-Earth Metals." Russian J. Inorganic Chem. 10, 171-2 (1965).

36. M. Foëx and J. P. Traverse. "High-Temperature Study of the Allotropic Transformations of the Sesquioxides of Yttrium, Erbium, and Thulium." Compt. Rend. 261, 2490-3 (1965).

37. P. K. Smith. High Temperature Stability of Thulium Oxide. USAEC Report, DP-1116, E. I. du Pont de Nemours and Co., Savannah River Laboratory, Aiken, S. C. (1967).

38. M. B. Panish. "Vaporization of the Rare Earth Oxides." J. Chem. Phys. 34, 2197-8 (1961).

39. J. R. Hague, et al., eds. Refractory Ceramics for Aerospace. The American Ceramic Society. Columbus, Ohio (1964).

40. H. R. Hoekstra. "Phase Relationships in the Rare Earth Sesquioxides at High Pressure." Inorg. Chem. 5, 754-7 (1966).

41. E. J. Huber, Jr., E. L. Head, and C. E. Holley, Jr. "The Heat of Combustion of Thulium." J. Phys. Chem. 64, 379-80 (1960).

42. E. F. Westrum, Jr. and Fredrik Gronvold. "Thermochemical Data of Actinide Element Chalcogenides." Thermodynamics of Nuclear Materials. pp 3-39. International Atomic Energy Agency, Vienna (1962).