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**^{252}Cf SHIELDING WITH
WATER-EXTENDED POLYESTER**

D. H. STODDARD

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Aiken, S. C. 29801**

PREPARED FOR THE U. S. ATOMIC ENERGY COMMISSION UNDER CONTRACT AT(07-2)-1

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ABSTRACT

Dose rates from ^{252}Cf shielded by ten mixtures containing water-extended polyster resins are calculated. The radiation properties of ^{252}Cf used in the calculations are tabulated.

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INTRODUCTION

Water-extended polyester (WEP) was first proposed for ^{252}Cf shielding by Oliver and Moore in 1970.¹ Additives can be used to change the shielding properties of this material.² This report presents ANISN-calculated shielding data on ten different mixtures containing WEP. These shielding curves will allow a shield designer to select the type of WEP mixture that best fits his needs. Table I lists the formulations and densities of the types of WEP for which the shielding calculations were made.

THE RADIATION PROPERTIES OF ^{252}Cf

Californium-252 decays 97% by the emission of alpha particles, and 3% by the spontaneous fission process, which causes the emission of 2.4×10^{12} neutrons/sec from one gram of this material.

Although isotopic compositions will vary somewhat depending on the mode of production and decay time, the penetrating* radiation from californium sources is primarily neutrons and gamma rays from the isotope ^{252}Cf and from its fission products.

The general nuclear properties of the californium isotopes are presented in Table II.³ The penetrating radiations of ^{252}Cf are listed in the following sections.

Gamma Activity of ^{252}Cf

The gamma activity of ^{252}Cf consists of gamma rays from:

- The alpha decay process. Gamma rays reported for the alpha decay of ^{252}Cf are listed in Table III.
- Prompt spontaneous fission. The energy and abundance of prompt gamma rays from spontaneous fission are listed in Table IV.
- Fission products continuously produced by spontaneous fission. The fission products formed from spontaneous fission approach equilibrium within a few hours after separation. Equilibrium fission product gamma activities are listed in Table IV.

*Penetrating radiation is that which will penetrate the horny layer of the skin.

TABLE I
WEP MATERIAL COMPOSITIONS

WEP Mixture	Density (as cast), g/ml	Component	wt %
1	1.07	Water	54.0
		661-P Polyester Resin ^a and Hardener	40.0
		Boric Acid	6.0
2 ²	1.125	661-P Polyester Resin and Hardener	40.2
		Ethylene Glycol	26.0
		Water	24.0
		Boric Acid	8.0
		Sodium Hydroxide	1.8
3	1.45	Water	30.8
		Lead Oxide	30.7
		661-P Polyester Resin and Hardener	30.8
		Boric Acid	7.3
		Oil	0.4
4	1.65	Lead Oxide	40.0
		661-P Polyester Resin and Hardener	26.7
		Ethylene Glycol	13.9
		Water	12.8
		Boric Acid	6.3
		Oil	0.3
5	1.71	Iron	40.0
		661-P Polyester Resin and Hardener	26.7
		Boric Acid	20.8
		Ethylene Glycol	5.8
		Water	5.3
		Oil	1.0
		Sodium Hydroxide	0.4
6	1.926	Iron	50.0
		661-P Polyester Resin and Hardener	25.0
		Boric Acid	18.6
		Water	2.7
		Ethylene Glycol	2.6
		Oil	1.0
		Sodium Hydroxide	0.2
7	2.035	Iron	53.3
		661-P Polyester Resin and Hardener	26.7
		Boric Acid	8.1
		Ethylene Glycol	5.7
		Water	5.5
		Sodium Hydroxide	0.4
		Oil	0.3
8	2.10	Lead Oxide	53.3
		661-P Polyester Resin and Hardener	26.7
		Boric Acid	8.1
		Ethylene Glycol	5.7
		Water	5.5
		Sodium Hydroxide	0.4
		Oil	0.3
9	1.13	661-P Polyester Resin and Hardener	40.0
		Water	40.0
		Lithium Sulfate	20.0
10	1.12	661-P Polyester Resin and Hardener	40.0
		Water	40.0
		Lithium Sulfate	10.0
		Boric Acid	10.0

^a. Ashland Chemical Co.

TABLE II

General Nuclear Properties of Californium Isotopes

Isotope	Spontaneous Fission Half-Life	Neutrons Per Fission of Pure Isotope	Alpha or Beta Decay Half-Life	Specific Activity of Pure Isotope, Ci/g
^{249}Cf	$1.5 \times 10^9 \text{ y}$	3	360 y	4
^{250}Cf	$(1.73 \pm 0.06) \times 10^4 \text{ y}$	3	11 y	1.31×10^2
^{251}Cf	-	-	$\sim 1500 \text{ y}$	0.95
^{252}Cf	$85.5 \pm 0.5 \text{ y}$	3.80 ± 0.035	$2.646 \pm 0.004 \text{ y}$	5.37×10^2
^{253}Cf	-	-	$18 \pm 3 \text{ d}$	2.87×10^4
^{254}Cf	$61.9 \pm 1.1 \text{ d}$	3.9 ± 0.14	-	-

TABLE III

Gamma Rays from ^{252}Cf Alpha Decay

Energy, MeV	Abundance, photons/(sec-g nuclide)
0.043	2.78×10^9
0.100	2.0×10^9

TABLE IV
Gamma Rays from Spontaneous Fission of ^{252}Cf

Energy, MeV	Abundance, photons/(sec-g nuclide)		
	Prompt Gamma	Equilibrium Fission Product Gamma	Total
0 to 0.5	3.3×10^{12}	1.3×10^{12}	4.6×10^{12}
0.5 to 1.0	1.7×10^{12}	4.0×10^{12}	5.7×10^{12}
1.0 to 1.5	7.7×10^{11}	9.1×10^{11}	1.7×10^{12}
1.5 to 2.0	4.2×10^{11}	3.5×10^{11}	7.7×10^{11}
2.0 to 2.5	2.2×10^{11}		2.2×10^{11}
2.5 to 3.0	1.1×10^{11}		1.1×10^{11}
3.0 to 3.5	5.6×10^{10}		5.6×10^{10}
3.5 to 4.0	3.0×10^{10}		3.0×10^{10}
4.0 to 4.5	1.7×10^{10}		1.7×10^{10}
4.5 to 5.0	8.2×10^9		8.2×10^9
5.0 to 5.5	4.9×10^9		4.9×10^9
5.5 to 6.0	1.8×10^9		1.8×10^9
6.0 to 6.5	1.0×10^9		1.0×10^9

Although X-rays are relatively high in abundance, those produced are very low in energy (most are <40 keV) and are not considered in this report.

Beta Activity of ^{252}Cf

No beta radiation has been reported from the decay process. The beta radiation associated with the equilibrium fission products during spontaneous fission is easily absorbed and is not included in this report.

Neutron Activity of ^{252}Cf

The neutron radiation from ^{252}Cf consists principally of neutrons from spontaneous fission. A second minor source of neutrons is the α, n neutrons from the reaction of alpha particles with light elements. Table V lists the neutrons present from spontaneous fission.

TABLE V

Neutrons from Spontaneous Fission of ^{252}Cf

Energy, MeV	neutrons/ (sec-g of nuclide)
0 to 0.5	2.8×10^{11}
0.5 to 1.0	3.7×10^{11}
1.0 to 2.0	7.6×10^{11}
2.0 to 3.0	4.6×10^{11}
3.0 to 4.0	2.8×10^{11}
4.0 to 5.0	1.6×10^{11}
5.0 to 6.0	5.6×10^{10}
6.0 to 7.0	4.0×10^{10}
7.0 to 8.0	1.3×10^{10}
8.0 to 10.0	9.9×10^9
10.0 to 13.0	2.2×10^9
Total	2.4×10^{12}

Neutrons associated with ^{252}Cf can also be represented by the Watt formula.⁴

$$N(E) = 0.373 \exp(-0.88E) \sinh(2.0E)^{1/2}$$

where $N(E)$ is the fraction of neutrons per unit energy range, and E is the neutron energy in MeV.

ATTENUATION CALCULATIONS

The dose attenuation calculations presented in this section provide a WEP shield reference for ^{252}Cf users. Dose rates are normalized to 1 fission neutron/sec, and multiplied by $4\pi r^2$ to remove the effect of spherical divergence. The dose rate plotted in Figures 4 through 13 is the surface dose rate at the interface between the shield and tissue-like material.

Applications

Dose Rates at the Surface of a Spherical Shield

Total dose rate at the surface of a spherical shield (Figure 1) may be determined by adding the neutron dose rates and the gamma dose rates at the common shield thickness and multiplying by the ^{252}Cf source weight in grams and $(1/4\pi r^2) \times 2.4 \times 10^{12}$ n/(sec-g)

$$D_T \text{ (mrem/hr)} = (D_{n_T} + D_{\gamma_T}) \times \frac{1}{4\pi r^2} \times 2.4 \times 10^{12} \times \text{grams}$$

Dose Rates at the Surface of a Cylindrical Shield

Total dose rate at the surface of a cylindrical shield (Figure 2) will always be less than at the surface of a sphere that can be inscribed within that cylinder. Direct use of the spherical shield dose rate curves presented for cylindrical shields may be expected to yield slightly conservative results (or a higher dose rate).

Dose Rates for a Slab Shield

The dose attenuation curves used for a slab shield (Figure 3) will yield a conservative dose rate ($\sim 2\times$ depending on the shielding material) at short ranges. The data become less conservative and will approach the spherical dose rate as the distance between the shield and receptor increases.

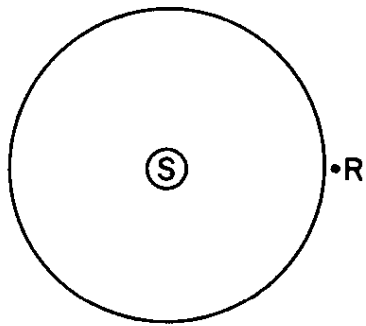


FIGURE 1. Spherical Shield Geometry

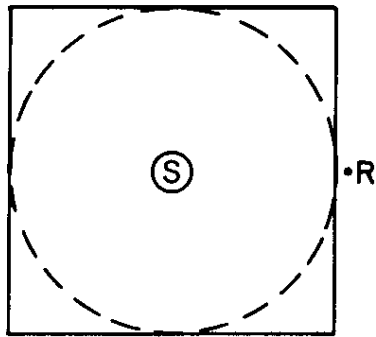


FIGURE 2. Cylindrical Shield Geometry

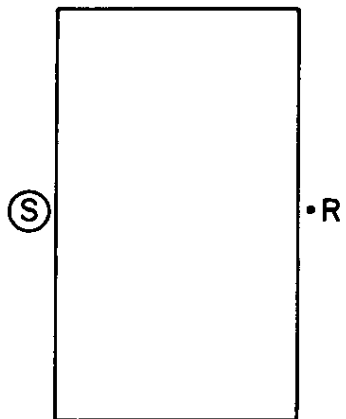


FIGURE 3. Slab Shield Geometry

Calculations

Dose rate attenuation curves $D(r)$ shown in Figures 4 through 13 have been calculated using the ANISN code.⁵

ANISN is an acronym for the discrete ordinates or S_n method of approximating the solution of the energy-dependent linear Boltzmann transport equation with anisotropic scattering. The one-dimensional equation may be applied to slab, spherical, or cylindrical geometry.

In every case, the source was assumed to be in a spherical aluminum matrix, having a volume of 1 cm^3 and a radius of 0.62 cm.

The angular flux was approximated by a sixteenth order quadrature (S_{16}) and the anisotropy of elastically scattered neutrons and of Compton-scattered photons was approximated by a third order Legendre expansion (P_3).

Energy Group Structure

The energy group structure has been derived using the multi-group theory of neutron and gamma-ray transmission. The groups have been divided into 13 neutron energy groups and 9 gamma energy groups. The neutron groups have been coupled with the gamma groups to calculate the inelastic and capture gamma activity that arises from neutron scatter or absorption.

Source

The source used in the calculations includes the gamma rays described in the previous section distributed within their energy intervals and the neutrons as distributed by the Watt formula. Low energy X-rays and beta radiation are not significant for personnel shielding because of their inability to penetrate materials in which sources are usually encapsulated.

Table VI lists the distribution of neutrons and gamma rays as a function of energy for each neutron emitted from ^{252}Cf .

TABLE VI

Twenty-Two Group Source Distribution for ^{252}Cf

<u>Group</u>	<u>Energy Interval, MeV</u>	<u>Type Radiation</u>	<u>Source Distribution</u>
1	10.0 to 14.92	Neutron ↓	0.0036
2	6.70 to 10.00		0.0279
3	5.49 to 6.70		0.0356
4	4.49 to 5.49		0.0547
5	3.68 to 4.49		0.0730
6	3.01 to 3.68		0.0869
7	2.02 to 3.01		0.1894
8	0.91 to 2.02		0.3033
9	0.41 to 0.91		0.1462
10	0.11 to 0.41		0.0676
11	0.015 to 0.11		0.0119
12	0.0000041 to 0.015		0.0
13 ^a	0.0 to 0.0000041	Gamma ↓	0.0
14	6.0 to 10.0		0.0012
15	5.0 to 6.0		0.0056
16	4.0 to 5.0		0.020
17	3.0 to 4.0		0.071
18	2.0 to 3.0		0.094
19	0.9 to 2.0		1.06
20	0.4 to 0.9		2.4
21	0.12 to 0.4		2.0
22	0.01 to 0.12		0.0

^a. Called thermal neutrons.*Cross Sections*

Cross sections for neutron-induced reactions and transitions between groups were taken from a 100-group ENDF/B library. The ANISN code was used to collapse the cross sections from 100 to 13 neutron energy groups. Gamma-ray group-to-group transfer cross-sections were generated by the MUG⁶ code, which includes photo-electric and pair production absorption as well as Compton effects. Nine gamma-ray energy groups were used. Cross sections for neutron-induced secondary and inelastic scattering gamma rays were coupled in the manner of POPOP4⁷ between the neutron and photon downscatter matrices.

Flux-to-Dose Conversion

Dose rates are given in terms of the biological dose rate, $D(r)$, (multiplied by $4\pi r^2$) computed at the interface between the shield and tissue-like material. Thus, the dose rate at that point contains the contributions from tissue backscatter. Conversion factors used to convert to biological dose rate are given in Table VII.

TABLE VII

Dose Rate Conversion Factors

Group ^a	Factor, mrem-hr ⁻¹	cm ²
	particle-sec ⁻¹	
1	0.147	
2	0.147	
3	0.142	
4	0.139	
5	0.134	
6	0.130	
7	0.125	
8	0.130	
9	0.105	
10	0.056	
11	0.020	
12	0.0056	
13	0.00373	
14	0.0081	
15	0.0061	
16	0.0054	
17	0.0045	
18	0.0036	
19	0.0031	
20	0.0014	
21	0.00065	
22	0.00015	

^a. Dose rate conversion factors for
Groups 1-13 were determined from data
in Reference 8; those for Groups 14-22
were determined from data in Reference 9.

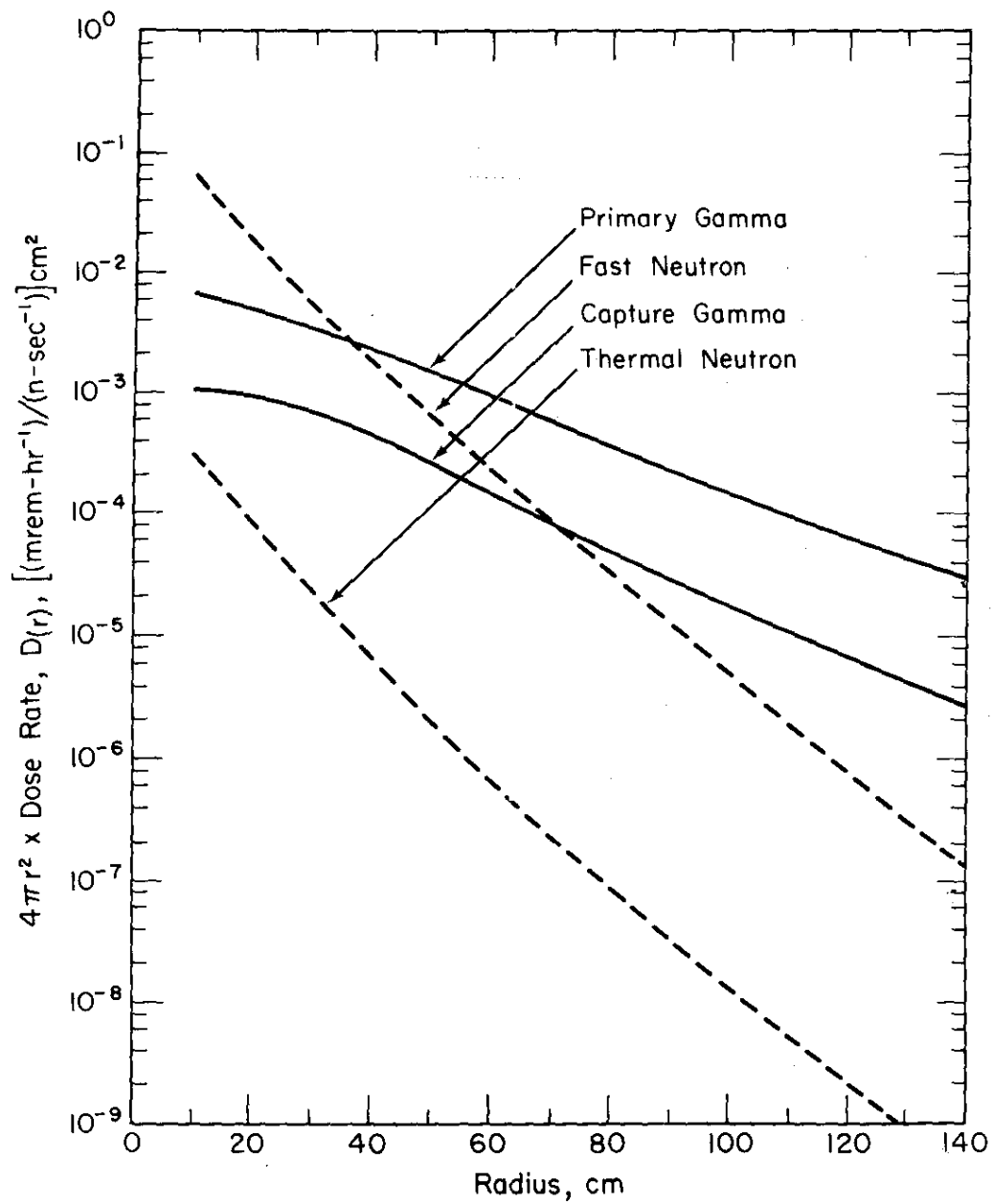


FIGURE 4. Dose Rates for WEP Mixture 1

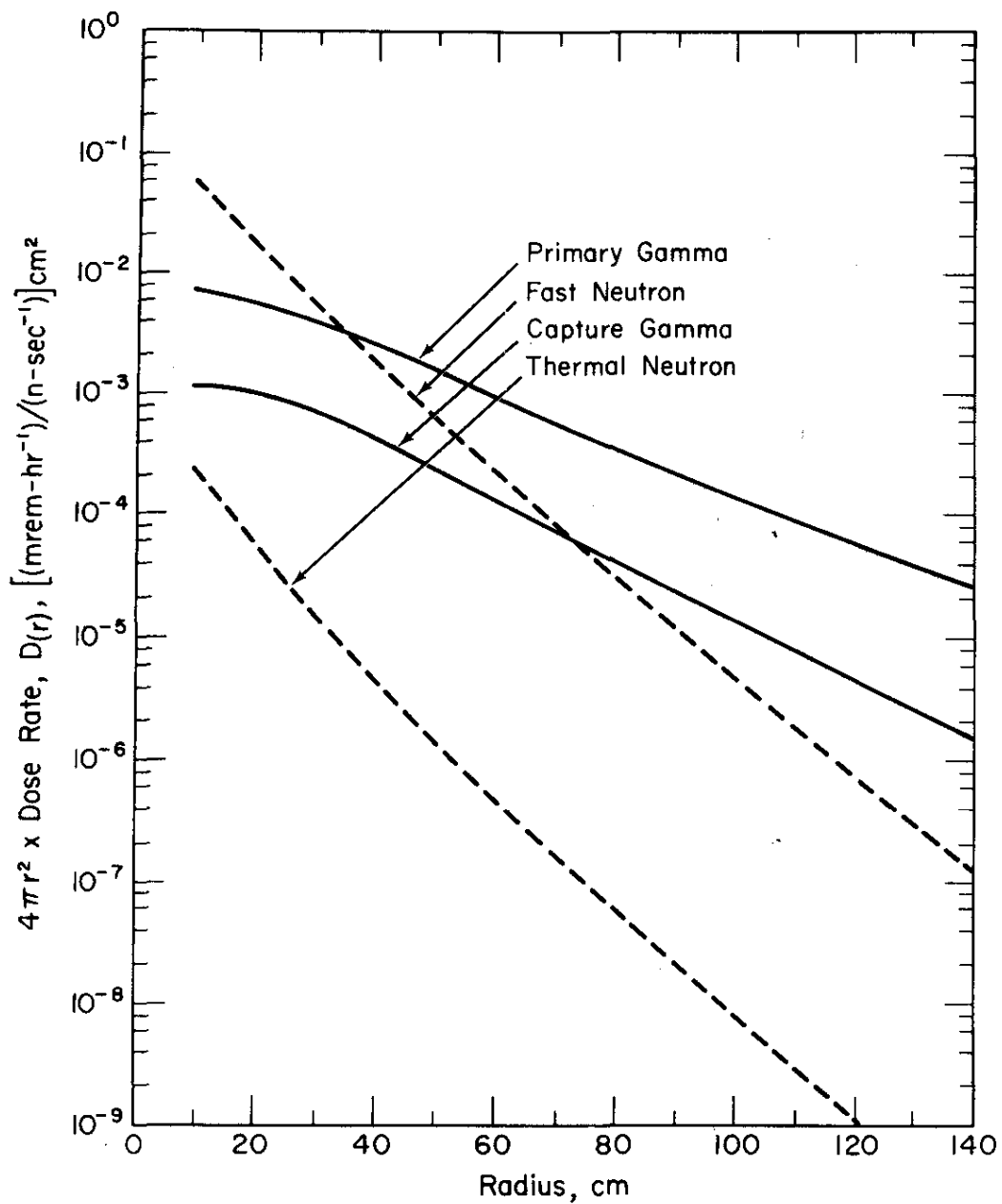


FIGURE 5. Dose Rates for WEP Mixture 2

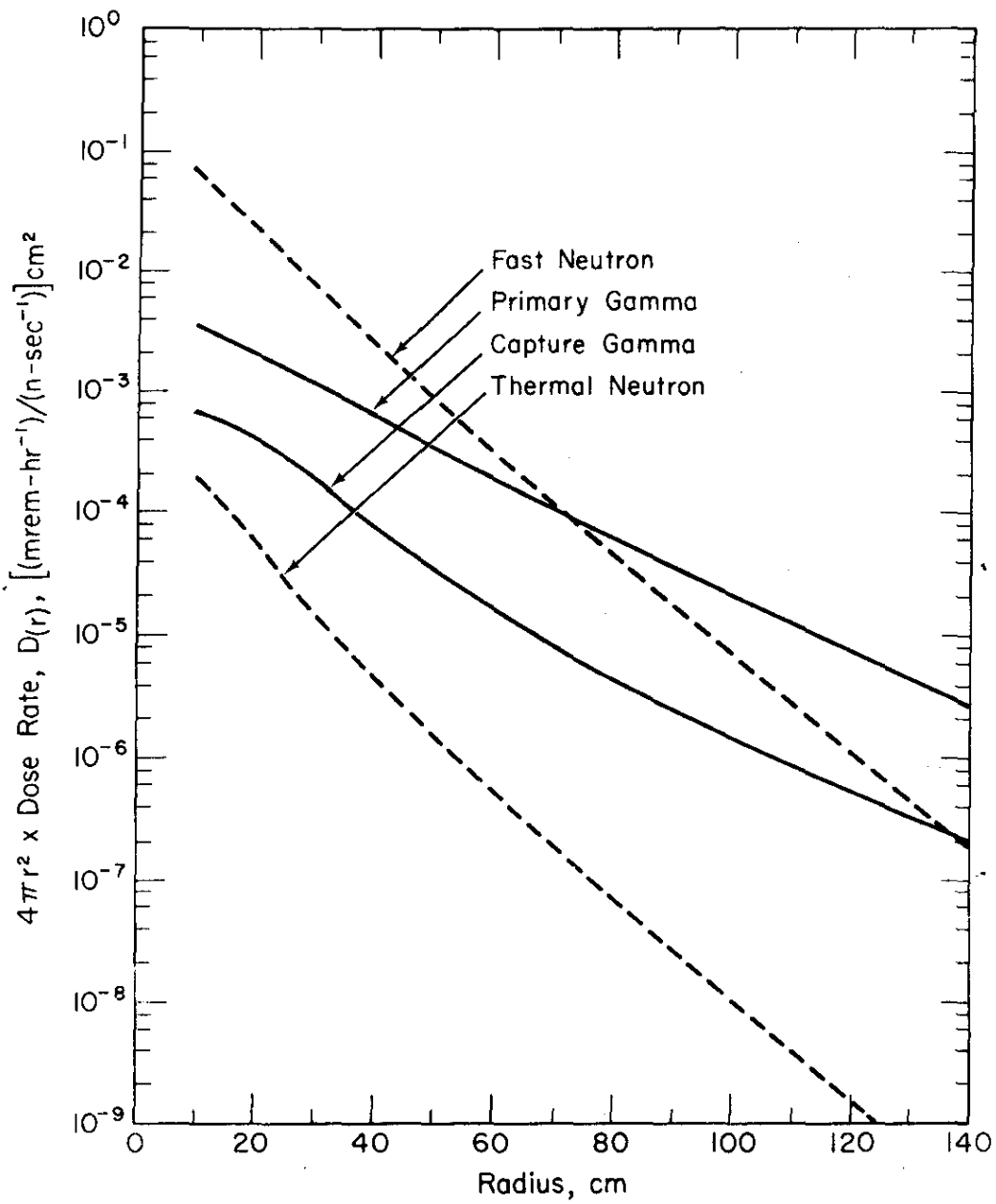


FIGURE 6. Dose Rates for WEP Mixture 3

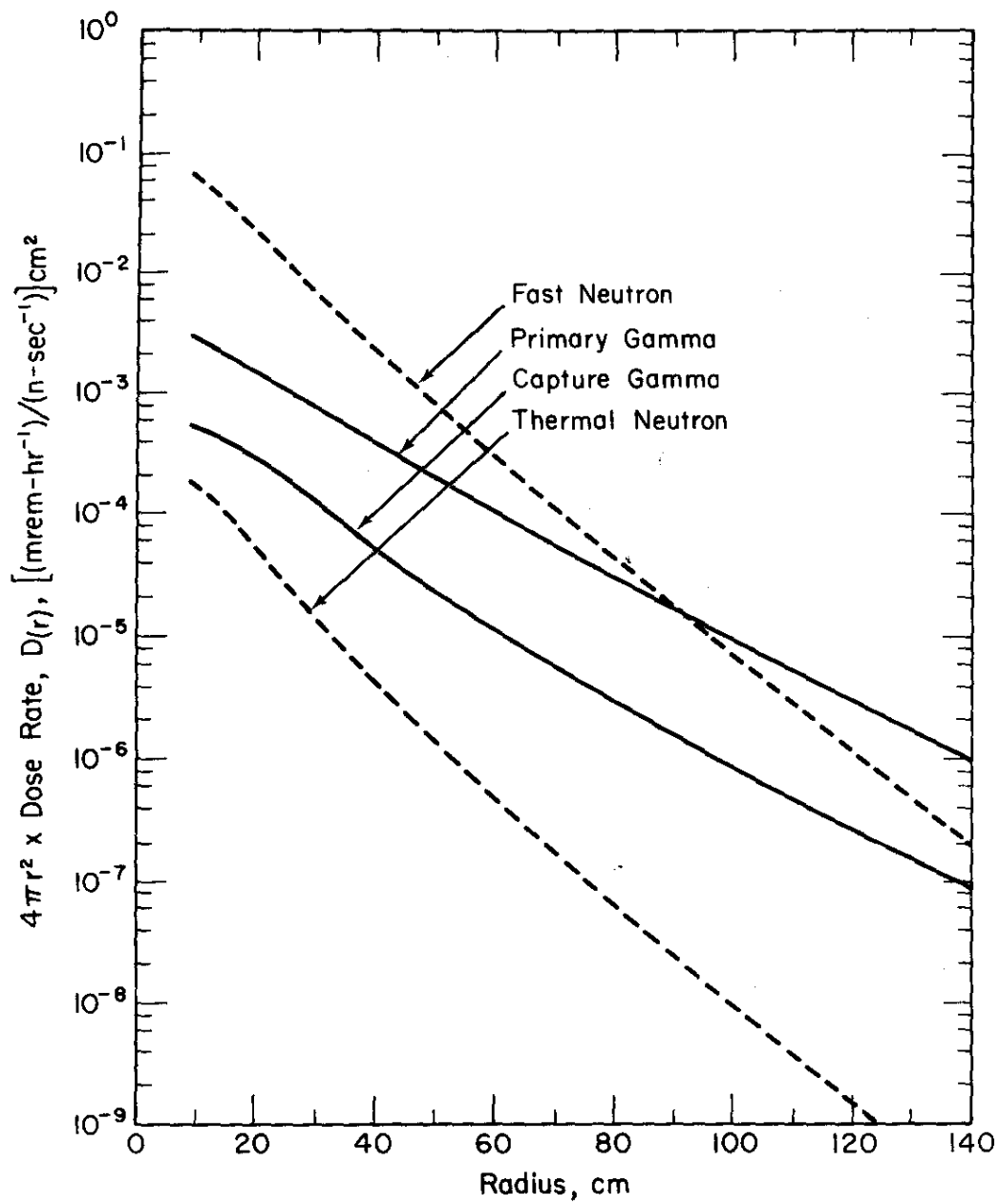


FIGURE 7. Dose Rates for WEP Mixture 4

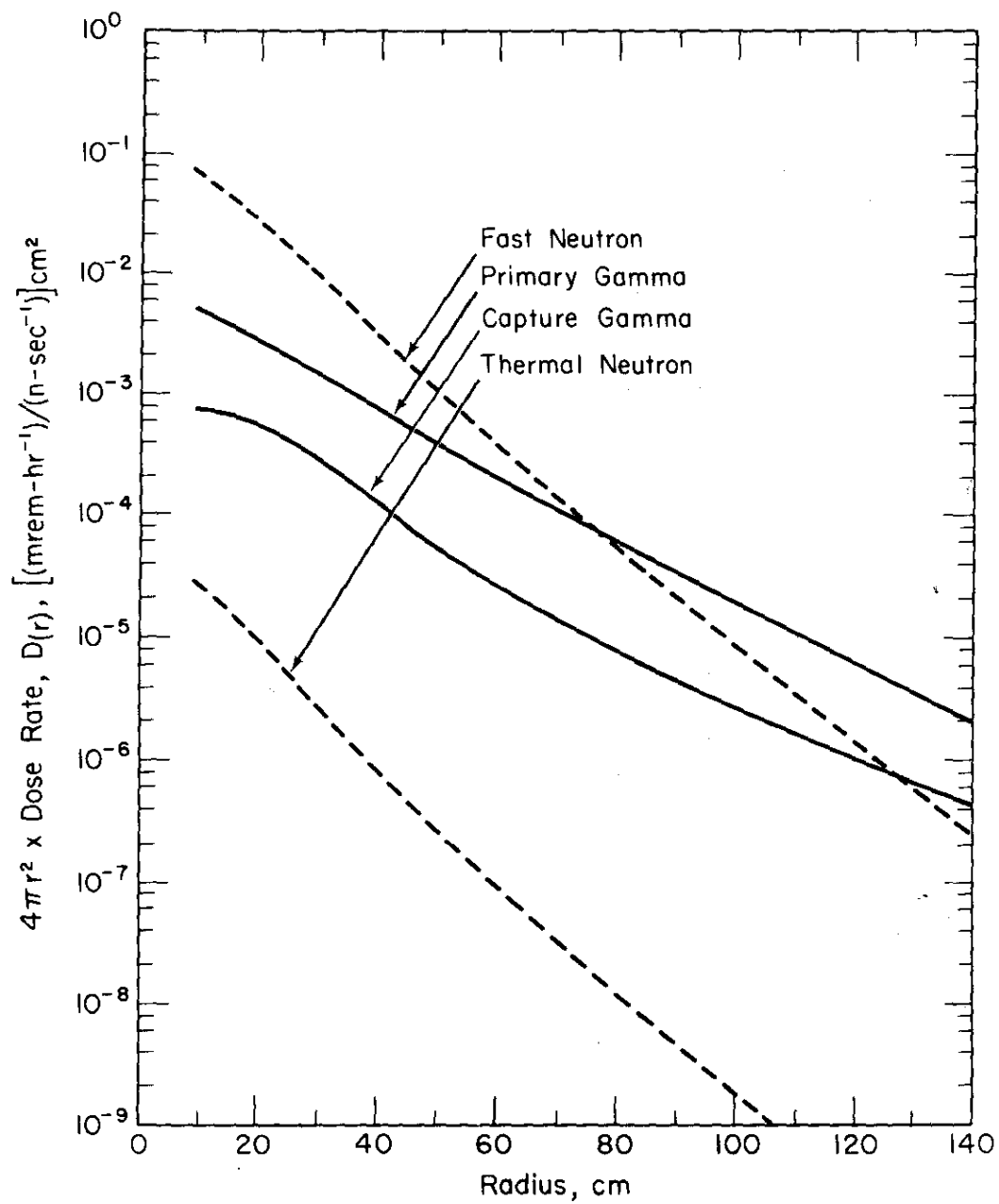


FIGURE 8. Dose Rates for WEP Mixture 5

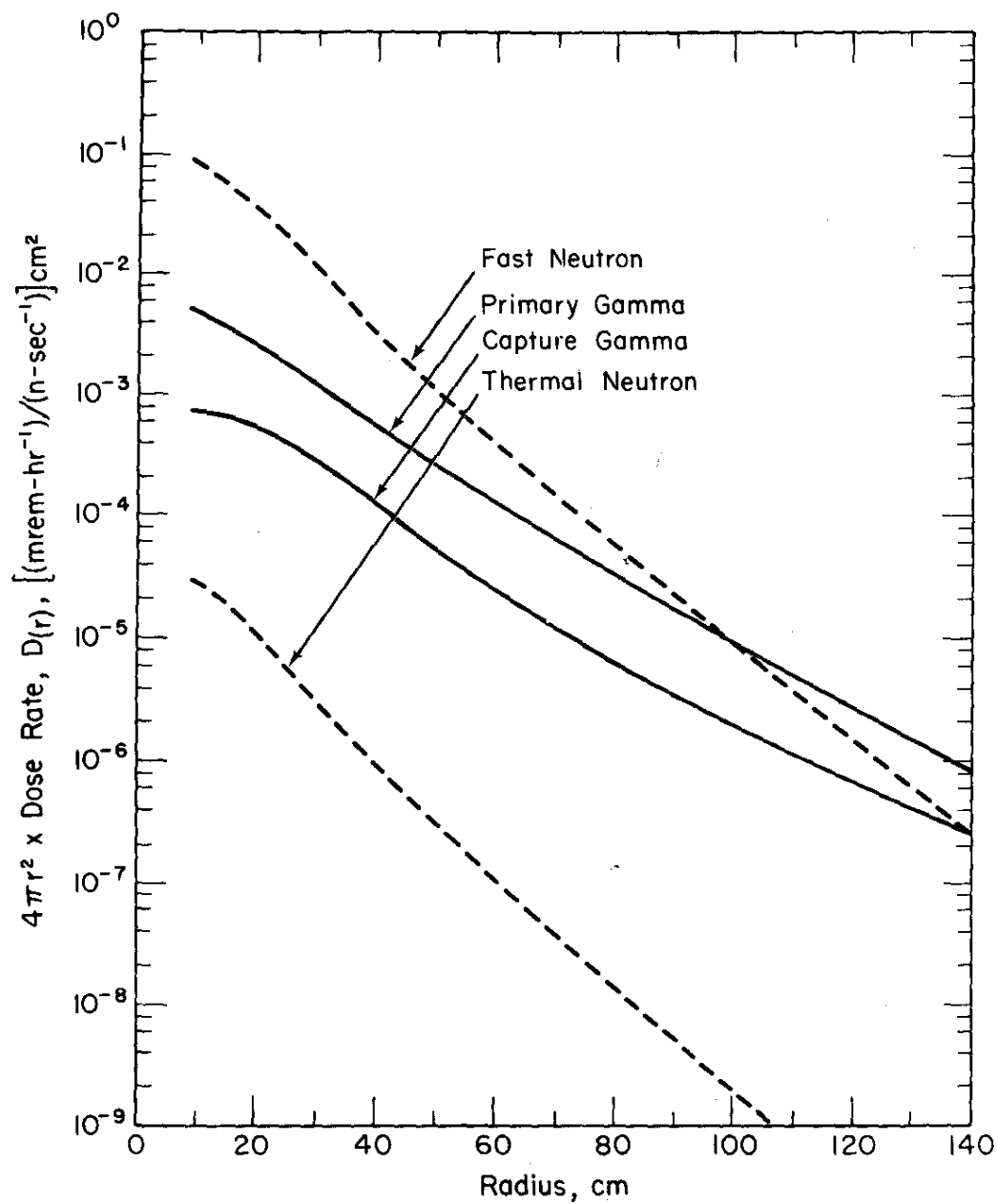


FIGURE 9. Dose Rates for WEP Mixture 6

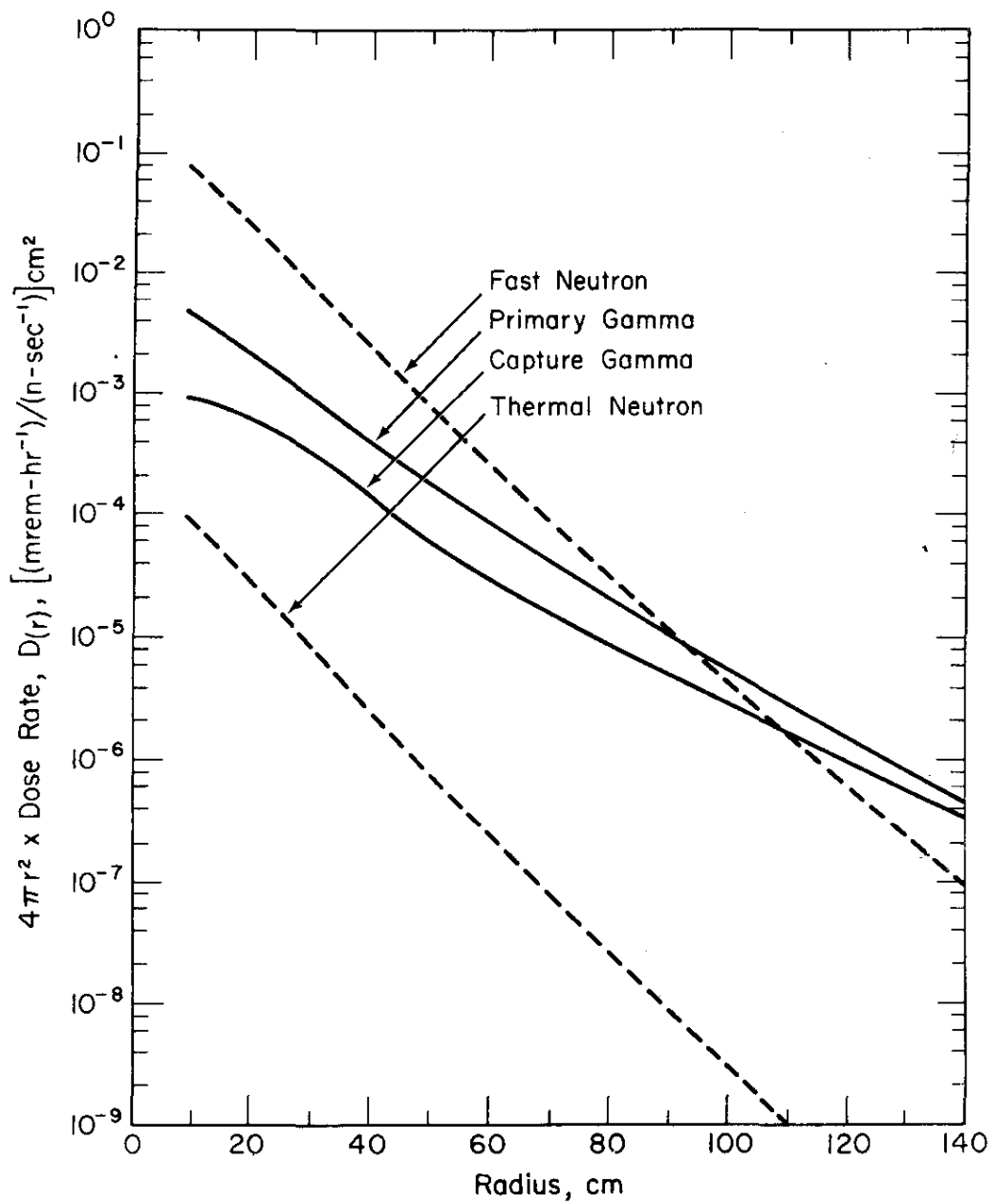


FIGURE 10. Dose Rates for WEP Mixture 7

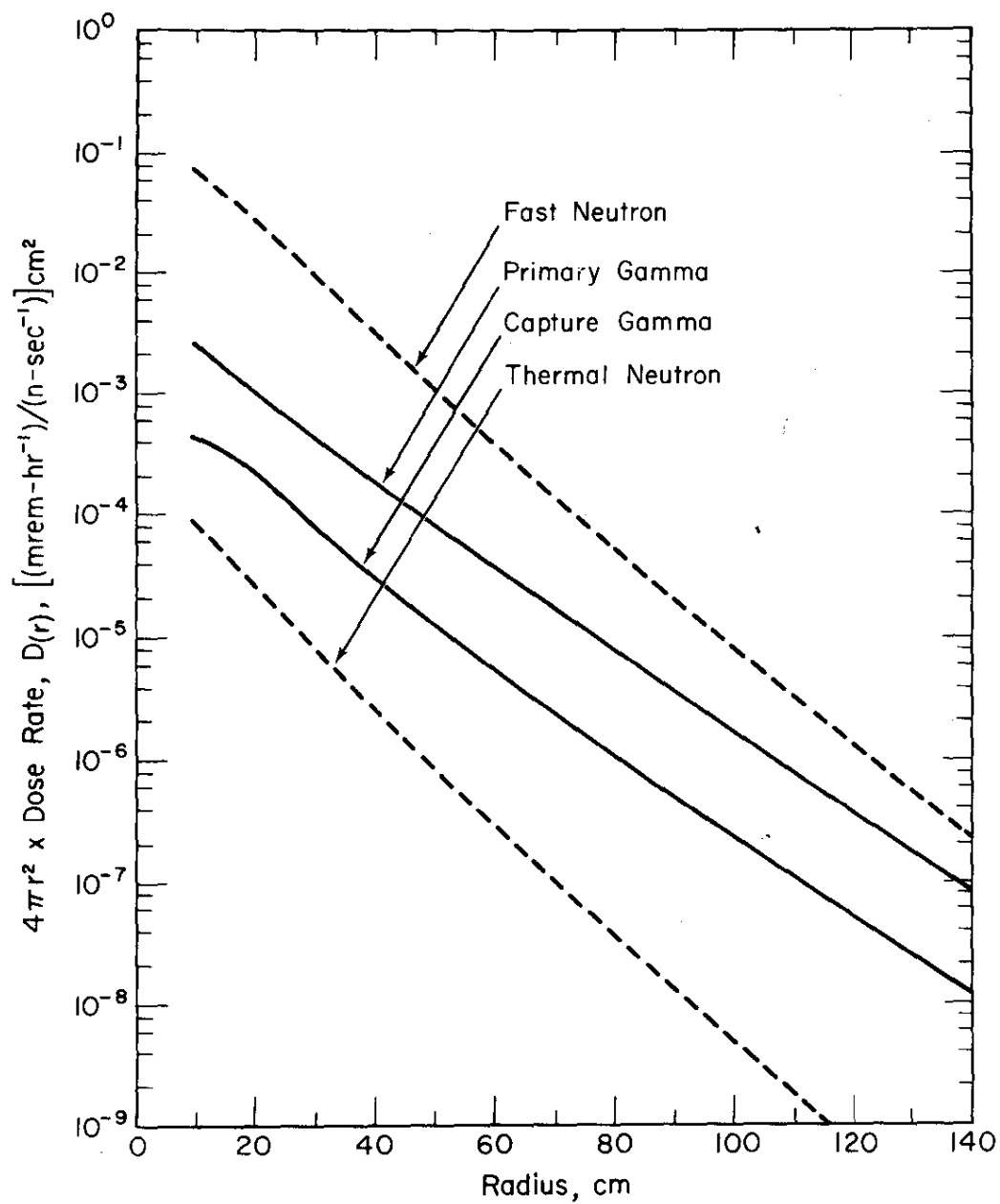


FIGURE 11. Dose Rates for WEP Mixture 8

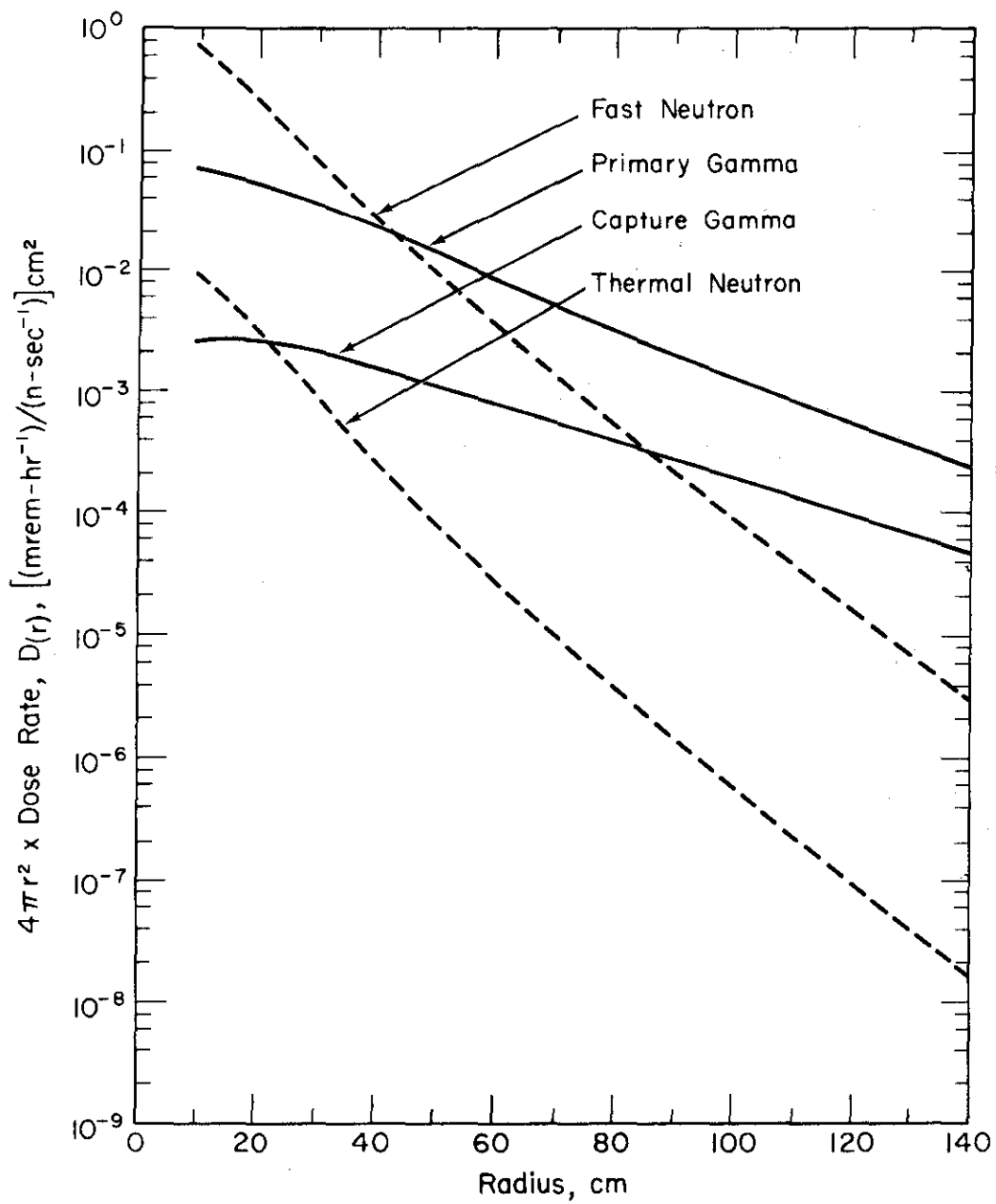


FIGURE 12. Dose Rates for WEP Mixture 9

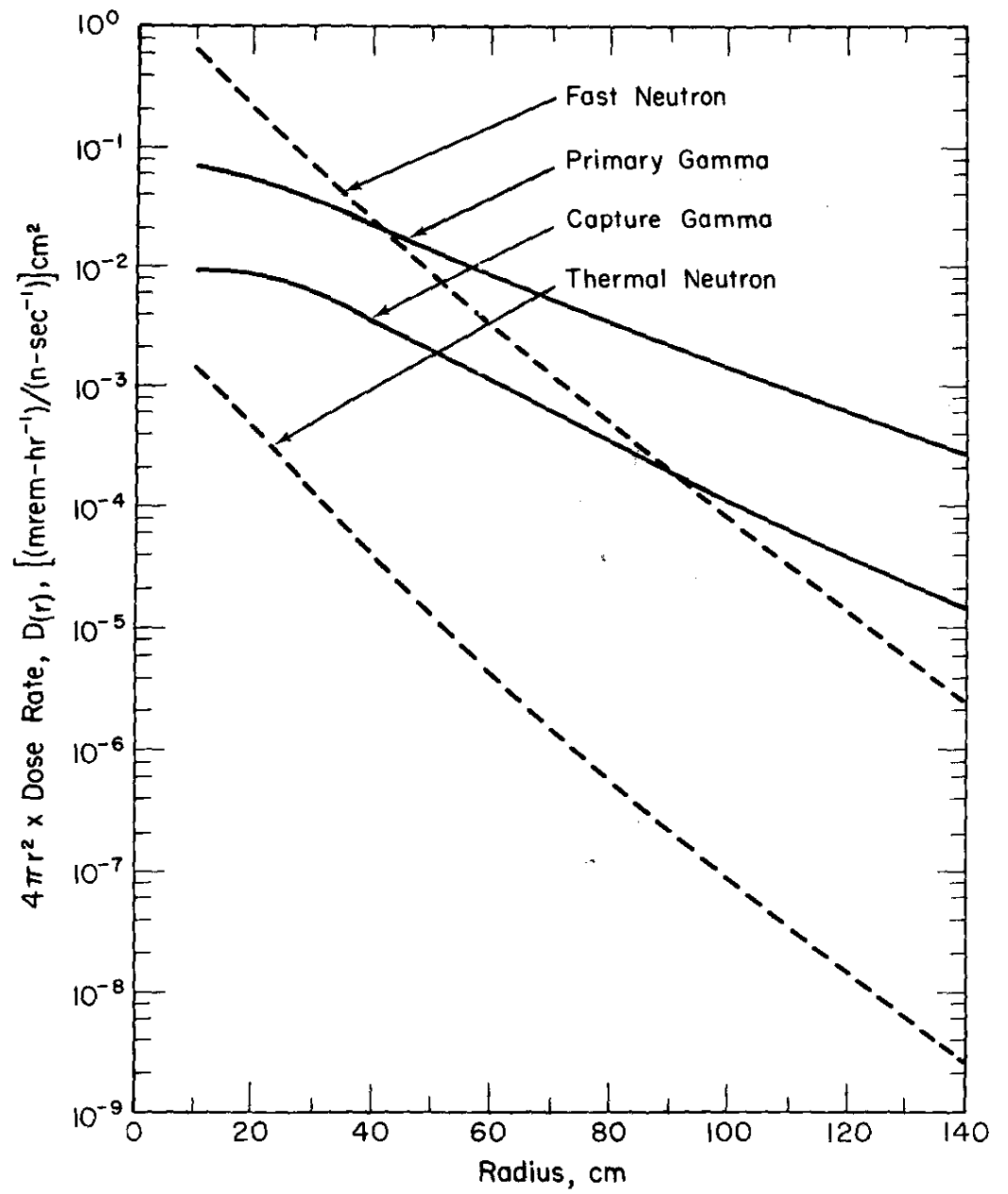


FIGURE 13. Dose Rates for WEP Mixture 10

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REFERENCES

1. G. D. Oliver, Jr. and E. Bailey Moore. "The Neutron Shielding Qualities of Water-Extended-Polyesters." *Health Phys.* 19, 578 (1970).
2. T. S. McMillan. *Thermal Evaluation of Water-Extended-Polyester*. USAEC Report DP-1262, E. I. du Pont de Nemours & Co., Savannah River Laboratory, Aiken, S. C. (1972).
3. D. H. Stoddard. *Radiation Properties of Californium-252*. USAEC Report DP-986, E. I. du Pont de Nemours and Co., Savannah River Laboratory, Aiken, S. C. (1965).
4. E. K. Hyde. *The Nuclear Properties of the Heavy Elements*. Vol. III, p. 240, Prentice Hall, Englewood Cliffs, N. J. (1964).
5. W. W. Engle, Jr. *ANISN Users Manual: A One Dimensional Discrete Ordinates Transport Code with Anisotropic Scattering*. USAEC Report K-1693, Oak Ridge Gaseous Diffusion Plant, Oak Ridge, Tenn. (1967).
6. J. R. Knight and F. R. Mynatt. *MUG, A Program for Generating Multigroup Photon Cross Sections*. USAEC Report CTC-17, Oak Ridge National Laboratory, Oak Ridge, Tenn. (1970).
7. W. E. Ford and D. H. Wallace. *POPOP4: A Code for Converting Gamma-Ray Spectra to Gamma-Ray Production Cross-Sections*. USAEC Report CTC-12, Oak Ridge National Laboratory, Oak Ridge, Tenn. (1969).
8. *The National Bureau of Standards Handbook 63*. U. S. Government Printing Office, Washington, D. C. (1957).
9. H. C. Claiborne and D. K. Trubey. "Gamma-Ray Dose Rates in a Slab Phantom." *Trans. Amer. Nucl. Soc.* 12, 383 (1969).