BNWL-C-45

~~BNWC-45~~

PROMETHIUM ISOTOPIC POWER DATA SHEETS

MARCH 15, 1965

BATTELLE-NORTHWEST

RICHLAND, WASHINGTON

PACIFIC NORTHWEST LABORATORY operated by BATTELLE MEMORIAL INSTITUTE

LEGAL NOTICE

AEC-RLOO RICHLAND, WASH.

BNWC-45

PROMETHIUM ISOTOPIC POWER DATA SHEETS

Compiled by H. H. Van Tuyl
Chemistry Department

March 15, 1965

BATTELLE-NORTHWEST
RICHLAND, WASHINGTON

Work performed under Contract No. AT(45-1)-1830 between the Atomic Energy Commission and Battelle Memorial Institute

Printed by/for the U.S. Atomic Energy Commission

PROMETHIUM ISOTOPIC POWER DATA SHEETS

INTRODUCTION

The Division of Isotopes Development of the Atomic Energy Commission has assigned to Pacific Northwest Laboratory (Battelle-Northwest) the responsibility for developing technology for preparing and using promethium as a radioisotopic fuel in space, marine, and terrestrial power applications. Technology has been developed (and demonstrated on a 20,000 Ci* scale) to permit large scale preparation of highly purified promethium. The half-life and specific heat generation rate were measured. Shielding requirements for promethium have been studied, and the existence of Pm^{146} in production and power fuels was discovered and its consequences investigated. Source preparation and encapsulation is being investigated, along with studies on compatibility with container materials, and reentry considerations. Several megacuries of promethium have been set aside from Purex process solutions to serve, after appropriate aging, as a source of high quality promethium for use in the development program.

The knowledge and technology of promethium as a fuel for radioisotopic power sources is summarized in the following pages. Additional details are available in the references cited. The current data will be reviewed periodically, and revised data sheets will be published as appropriate.

---

*Ci is the recently adopted abbreviation for curie.

NUCLEAR PROPERTIES

Half-Life	Pm^{147}	2.620 ± 0.005 yr[1]
	Pm^{146}	4.5 yr[2]
		1.94 yr[3]
	Pm^{148m}	42 days
	Pm^{148}	5.4 days
Specific Activity	928.4 ± 1.7 Ci/g of Pm^{147}	
Heat Generation	0.3330 ± 0.0005 W/g of Pm^{147}[1]	
	0.3587 ± 0.0010 W/kCi of Pm^{147}	
	2788 ± 8 Ci of Pm^{147}/W	
Radiation Properties	Pm^{147}	0.225 MeV beta (100%)[4]
		0.12 MeV gamma (0.003%)
		0 to 0.225 MeV bremsstrahlung [Spectral distribution is given in Reference (5)]
	Pm^{146}	0.78 MeV beta (35%)[3]
		0.45 MeV gamma (65%)
		0.75 MeV gamma (65%)
	Pm^{148m}	Average values are given in Reference (6). Pm^{148m} is unimportant in well-aged promethium.
Isotopic Composition	Pm^{147}	100%
	Pm^{146}	5×10^{-5}% by activity for Hanford material[7] and 4×10^{-4}% for 13,000 to 32,000 MWd/tonne fuel from Yankee (pressurized water) power reactor. (Unpublished measurement)
	Pm^{148m}	Amount depends on reactor conditions and cooling time. Less than 10% of Pm^{146} activity after cooling about 2.5 yr.
Maximum Permissible Concentration in Air	For continuous exposure, 2×10^{-8} μCi/ml,[9] or tenfold lower by federal regulation.[10] When expressed as watts per unit volume of air, the limit for Pm^{147} is 300 times higher than the limit for Pu^{238}, and ten times higher than the limit for Sr^{90}. On a curie basis the comparison is even more favorable for promethium.	

Shielding

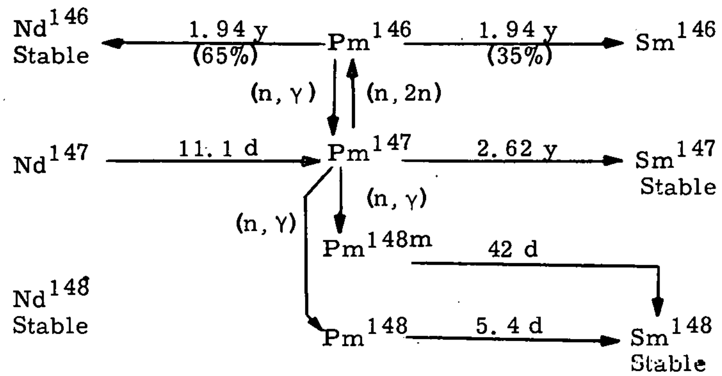
Centimeters of Uranium Required to Reduce Dose Rate at 1 m to Desired Level

Source Size, W	4×10^{-4}% Pm^{146}			5×10^{-5}% Pm^{146}		
	1 R/hr	0.1 R/hr	0.01 R/hr	1 R/hr	0.1 R/hr	0.01 R/hr
10000	1.0	2.3	3.6	0.05	1.1	2.5
1000	0.2	1.5	2.8	<0.01	0.3	1.6
100	0	0.5	1.8	0	0.01	0.6
10	0	<0.01	0.7	0	0	0.04
1	0	0	<0.01	0	0	<0.01

[Promethium oxide sources. First 0.1 cm of shield is either tantalum or tungsten so that maximum shielding efficiency is obtained. See Reference (6)]

Damage by radiations from promethium is tenfold less to man and 1000 fold less to transistors than the same quantity of fast neutron radiation when compared on an energy absorption basis (i.e., rep or rad).[8]

Methods of Formation



Fission Yields, %

Nd^{147} 2.16 from U^{235}[11]
2.07 from Pu^{239}[12]
2.9 from U^{238}

Pm^{146} 1.2×10^{-6} average from fissions in a thermal reactor, natural uranium fuel.[2]

Pm^{148} and Pm^{148m} have low fission yields since most of the 148 chain stops at stable Nd^{148}.

Neutron Cross Sections

Pm^{147} 235 barns thermal
3220 barns resonance integral[13]
(n, 2n) 5.9 millibarns for fission spectrum neutrons[7]

Pm^{146} 8400 barns for reactor spectrum[14]

Pm^{148m} 29,000 barns for reactor spectrum[13]

PROMETHIUM PRODUCTION TECHNOLOGY

Chemical Purity	99% at time of separation. (Purities as high as 99.99% are obtainable, but are not realistic because Pm^{147} decays at the rate of 2.2% per month to stable Sm^{147}.)
Radiochemical Purity	Less than 1000 disintegrations/min total alpha activity (including Am^{241} and Cm^{242}) per curie of Pm^{147}.(15) 10^{-11} Ci Eu^{154} per curie of Pm^{147} (unpublished measurement). No other activities detectable. Radiation dosages and shielding requirements for promethium of this quality are determined solely by the promethium isotopes themselves.
Availability	Hanford Isotope Production Plant (proposed for completion in 1968) to produce at least 30 MCi of purified promethium per year.(16) Interim facilities will produce small megacurie quantities (FY 1965-66). Interim (before Hanford Isotope Production Plant) capability of about 10 MCi/yr is proposed for Strontium Semiworks installation. Power reactors produce about 3 Ci of promethium (after aging one half-life) per megawatt day; this is equivalent to 15 MCi/yr by 1970.

COMPATIBILITY WITH CONTAINER MATERIALS

(Interpolated from preliminary data with neodymium and samarium. Measurements are continuing.)

	Molten Metal	Solid Metal	Oxide
Time, Days	23	30	42
Temperature, C	1100	600	1100
Tantalum	Excellent	Excellent	----
Molybdenum	----	----	Excellent
Zirconium	----	Excellent	----
Titanium	Fair	Good	----
Platinum	Poor	----	Poor
Hastelloy C	Poor	Fair	Good
Hastelloy N	Poor	----	Good
Hastelloy X	Poor	----	Good
Haynes-25	Poor	----	Good
Stainless Steel 310	Poor	Fair	Fair
Inco-600	Poor	Poor	----
Rene-41	Poor	Poor to Fair	----
Carpenter-20	Poor	----	----
TZM	----	Excellent	----
Naval Brass	----	Fair	----

Excellent:	No attack observed
Good:	Less than 1 mil penetration during test
Fair:	1 to 10 mil penetration during test
Poor:	Over 10 mil penetration during test

PROPERTIES OF CANDIDATE HEAT SOURCE MATERIALS

	Metal	Oxide	References
Density, g/cm^3	7.3	7.3	(17)
Promethium Content, wt%	100	86	
Power Density, W/cm^3	2.43	2.09	
Specific Power, W/g	0.3330[a]	0.286[a]	
Melting Point, C	1080[a]	2270	(17, 18)
Heat of Fusion, kcal/mole	1.8		(17)
Boiling Point, C	3300		(17)
Heat of Vaporization, kcal/mole	60		(17)
Heat of Formation, kcal/mole		-432	
Heat Capacity, cal/mole-C°	(b)	(c)	
Thermal Conductivity, cal/sec-cm-C°	0.031 at 27 C	(d)	(19)
Thermal Expansion Coefficient	6.7×10^{-6} at 400 C	9×10^{-6}	(17, 20, 21)
Vapor Pressure	(e)	(f)	
Surface Tension, dynes/cm	40 to 70 at 2300 C		
Transition Temperature to Body Centered Cubic Form, C	890		(17)
Heat of Transition, kcal/mole	0.73		(17)
Heat of Sublimation, kcal/mole	75 at 960 C		

(a) Measured value, or derived from measured values. Others are estimates based on values for neighboring rare earths.

NOTE: All following temperatures in K.

(b) $(4.0 + 0.00925T - 2.9 \times 10^{-9}T^3)$ for promethium interpolated from Reference (22) data.

(c) $(28.99 + 0.00576T - 415,900/T^2)$ for neodymium, Reference (23).

(d) $[0.0045 + 1.414 \times 10^{-8} (1020 - T)^2]$ for samarium, Reference (24).

(e) $(\log_{10}P = 6.50 - 13,000/T)$ where P is vapor pressure in mm of mercury. Derived from estimated boiling point and heat of vaporization.

(f) $(\log_{10}P = 15.62 - 39,800/T)$ for lanthanum, Reference (25).

MECHANICAL PROPERTIES OF PROMETHIUM

[Interpolated from values for neodymium and samarium, Reference (26)]

Tensile Strength, psi	Cast	Cold Reduced
27 C	22,000	30,000
200 C	21,000	22,000
430 C	9,000	13,000
Yield Strength, psi		
27 C	19,000	----
200 C	18,000	18,000
430 C	8,000	12,000
Elongation, %		
27 C	7	2
200 C	10	12
430 C	9	14
Vickers Hardness, 27 C	74	76
Poissons Ratio	0.306	0.352

REFERENCES

1. E. J. Wheelwright, D. M. Fleming, and F. P. Roberts, to be published in April or May issue of J. Phys. Chem. 1965.
2. I. M. H. Pagden, R. Jakeways, and F. C. Flack. Nucl. Phys., vol. 48, pp. 555-60. 1963.
3. E. G. Funk, Jr., J. W. Mihelich, and D. F. Schwerdtfeger. Phys. Rev., vol.120, p. 1781. 1960.
4. D. Stromminger, J. M. Hollander, and G. T. Seaborg. Rev. Mod. Phys., vol. 30, pp. 585-904. 1958.
5. H. H. Van Tuyl. HW-83784. 1964.
6. H. H. Van Tuyl, F. P. Roberts, and E. J. Wheelwright. HW-77375. 1963.
7. F. P. Roberts, E. J. Wheelwright, and H. H. Van Tuyl. J. Inorg. Nucl. Chem., vol. 25, p. 1298. 1963.
8. E. D. Arnold. ORNL-3576. 1964.
9. U.S. National Bureau of Standards Handbook 69. 1959.
10. Title 10, Code of Federal Regulations, Part 20, Appendix B. 1963.
11. H. Farrar and R. H. Tomlinson. Nucl. Phys. 34, pp. 367-381. 1962.
12. Seymour Katcoff. Nucleonics, vol. 18, no. 11, pp. 201-208. 1960.
13. R. P. Schuman and J. R. Berreth. Nucl. Sci. Eng., vol. 12, pp. 519-22. 1962.
14. Oak Ridge National Laboratory. ORNL-3762. 1964.
15. E. J. Wheelwright and F. P. Roberts. HW-78651 REV. 1963.
16. J. R. LaRiviere, et al. HW-77770 Pt. 1. 1963.
17. F. H. Spedding and A. H. Daane. The Rare Earths, Wiley. 1961.
18. F. Weigal. Angew. Chem., vol. 75, p. 451. 1963.
10. S. Legvold and F. H. Spedding. AEC Report SC-508. 1954.
20. P. L. Witlong, L. O. Dominique, L. R. Furlong, and J. A. Finlayson. BM-RI-6180.
21. G. L. Ploetz, C. W. Krystyniak, and H. E. Dumes. KAPL-M-GLP-1. 1957.
22. F. H. Spedding, J. J. McKeown, and A. H. Daane. J. Phys. Chem., vol. 64, p. 289. 1960.
23. J. O. Blomeke and W. T. Ziegler. J. Am. Chem. Soc., vol. 73, p. 5099. 1951.
24. W. E. Kingery and F. H. Norton. NYO-6450, AD-73-173. 1955.
25. S. J. Yosim and T. A. Milne. NAA-SR-1797. 1957.
26. A. C. Davis (Editor). Design Engineering, vol. 60, p. 163. October 1964.