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PROMETHIUM ISOTOPIC POWER DATA SHEETS

J. H. Jarrett and H. H. Van Tuyl

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PROMETHIUM ISOTOPIC POWER
DATA SHEETS

By

J. H. Jarrett and H. H. Van Tuyl
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Chemistry and Metallurgy Division

May 1970

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ABSTRACT

This report summarizes the nuclear, physical, mechanical and chemical properties of promethium metal and promethium oxide. It also covers the projected availability of promethium, summary information on containment materials, and summary information on promethium applications.

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PROMETHIUM ISOTOPIC POWER
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INTRODUCTION

The Division of Isotopes Development of the Atomic Energy Commission has assigned to Pacific Northwest Laboratory (PNL) (Battelle-Northwest) the responsibility of developing technology for the preparation and use of promethium in space, marine, and terrestrial applications.

This report summarizes the nuclear, physical, mechanical, and chemical properties of promethium. Estimates of properties not yet measured have been made based on measured values for neighboring rare earths.

Technology has been developed (and demonstrated on a several hundred gram scale) to permit large scale preparation of highly purified promethium. The half-life and specific heat generation rate for promethium have been measured, and its shielding requirements have been studied. A summary of promethium's major characteristics is shown in Table 1.⁽¹⁾

APPLICATIONS

Applications of promethium and its compounds have previously been demonstrated and reported.⁽²⁾ Both the beta-emission properties and the heat generation properties have been used in various applications. Some of these applications using beta-emission properties include:

- Promethium-activated phosphor light sources^(3,4)
- Beta backscatter thickness gauges^(5,6)
- Beta-excited X-ray applications⁽⁷⁻¹⁰⁾
- Beta voltaic electric power sources⁽¹¹⁻¹³⁾
- Photoelectric battery⁽¹⁴⁾

TABLE 1. Summary of Major Characteristics
of Promethium

Watts/Gram (100% Basis)	0.3330
Half-Life, Years	2.62344
Typical Cation Purity, %	95
Usual Compound Form	Pm ₂ O ₃
Melting Point of Compound, °C	2320
Active Isotope in Compound, wt%	82
Watts/Gram (Compound)	0.2731
Density of Compound, g/cm ³ as produced	6.6
Power Density, W/cm ³ Compound	1.80
Major Radiation Type	β
Estimated Future Price, \$/g (Pure)	75
Present Price, \$/g (Pure)	186
Estimated Future Price, \$/W	220
Curies/Gram (100% Basis)	927.62
Curies/Watt	2786
Total kWh _t Per Initial Gram, 3-Year Mission	6

The following applications using heat generated by promethium in the form of the oxide, Pm₂O₃, have been demonstrated:

- Heaters, (15-17) including a flight qualified unit (18,19)
- Heat pipe operation (20)
- Radioisotet - attitude control microthruster (21,22)
- Thermionic systems (23)
- Dynamic energy conversion systems:
 - Stirling engine operation (20)
 - Vuilleumier refrigerator operation (20)

Numerous potential applications exist for ¹⁴⁷Pm metal. These include thermal applications where a power density higher

than that for the oxide is required, or thermal applications where the heat source material must be machined or cast into a complex shape.⁽²⁴⁾

AVAILABILITY

The availability of promethium has been discussed in general terms in a previous publication.⁽²⁾ Several megacuries of semi-pure, aged promethium are in storage at the Pacific Northwest Laboratory and this material can be fully purified as required. Promethium which has been aged for about one half-life of ^{147}Pm is desirable for many applications. This aging allows the gamma emitting $^{148\text{m}}\text{Pm}$ to decay to negligible levels, greatly reducing the total radiation.⁽²⁾

Promethium is produced as a fission product in the Hanford and Savannah River Production Reactors and in power reactors. The amount available from the Hanford reactors has not been published, but the availability was assumed to be 30 MCi (about 10 kW) per year in the Hanford Isotopes Plant study.⁽²⁵⁾ Savannah River production reactor promethium availability information has not been published. The estimated availability of promethium from power reactors is shown in Figure 1. This is similar to other reported availability estimations.^(26,27)

FIGURE 1. Estimated Growth of Nuclear Power in the United States*
(End of Year Basis)

NUCLEAR PROPERTIESHALF-LIVES

Half lives of principal radioisotopes are given in Table 2.

TABLE 2. Half-Lives of Principal Radioisotopes

Nuclide	Half-Life				Reference
	Days		Years		
¹⁴⁷ Pm	958.19 ± 0.06		2.62344 ± 0.00016		28
¹⁴⁶ Pm	2020	± 18	5.53	± 0.05	29
¹⁴⁸ Pm	5.4	± 0.1	0.0148	± 0.0003	30
^{418m} Pm	43	± 2	0.118	± 0.005	30
¹⁵⁴ Eu	5800	± 1500	16	± 4	31

SPECIFIC POWER

Specific activity and power values for various percentages of promethium are shown in Table 3.

TABLE 3. Specific Activity and Specific Power Values

Quantity	Value			Reference
	100% Pm	95% Pm	82% Pm	
Watts/gram	0.3330 ± 0.0005	0.3164	0.2731	32
Curies/gram	927.62 ± 0.06	881.2	760.6	28
Watts/kilocurie	0.3590 ± 0.0005	0.3590	0.3590	
Curies/Watt	2786 ± 4	2786	2786	
Grams/kilocurie	1.07803 ± 0.00007	1.1348	1.3147	
Grams/Watt	3.003 ± 0.005	3.161	3.662	

Values for 100% promethium can be closely approached by rapid processing of freshly separated promethium. Values for 95% promethium represent typical as-produced metal delivered about 2 months after separation. Values for 82% promethium represent typical as-produced oxide (containing 14% oxygen and

4% samarium) delivered about 2 months after separation. Other promethium contents can be evaluated from the data in Figure 2 or by direct proportion to the amount present.

FIGURE 2. Effect of ^{147}Pm Content on Specific Power and Activity

COMPOSITION

Promethium isotopes are formed through the reactions shown in Figure 3. Typical contents, expressed as curies of the various nuclides per million curies of ^{147}Pm , are shown in the following table.

FIGURE 3. Methods of Formation of Promethium Isotopes

TABLE 4. Typical Radioisotopic Composition of Promethium

Nuclide	Content, ppm by Activity		Reference
	Nominal	Range	
^{146}Pm	0.25	0.10 - 5	33
$^{148\text{m}}\text{Pm}$	0.025	0 - 0.5	34
^{154}Eu	0.001	0.00001 - 0.01	35

The ^{146}Pm content is about 0.25 ppm from production reactors and 1 to 5 ppm from power reactors. Re-irradiation under selected conditions will reduce this to 0.15 ppm or less, but with a cost increase.⁽³⁶⁾

The $^{148\text{m}}\text{Pm}$ content can be adjusted to any desired value by aging, as illustrated in Figure 4. An arbitrary limit of 10% of the ^{146}Pm activity represents a reasonable compromise between low dose rate and high promethium yields.

Europium 154 is the only significant nonpromethium radioisotope found in as-produced promethium. The europium content can be adjusted to any desired level by chemical processing.

The amount of ^{147}Sm in promethium changes with time since ^{147}Pm decays at the rate of 2.2% per month. The rate of ^{147}Pm decay is shown in Figure 5 and Table 5.

The fraction of ^{147}Pm remaining at times not listed can be derived by multiplying the fractions remaining at appropriate shorter times. The fraction of ^{147}Sm present can be derived by subtracting the fraction of ^{147}Pm from 1. For example, at 123 days the fraction of ^{147}Pm remaining is

$$0.93022 \times 0.98564 \times 0.99783 = 0.91487,$$

and the fraction which has been converted to ^{147}Sm is

$$1.00000 - 0.91487 = 0.08513.$$

RADIATION

Decay schemes for the radioisotopes of interest in promethium sources are shown in Figures 6 through 9. The transitions between various levels are shown by energy and, in parentheses, the number of transitions per 100 disintegrations of the parent (which is the percent abundance).

FIGURE 4. Relative Decay Rates of Promethium Isotopes

FIGURE 5. Rate of Decay of Promethium Isotopes

TABLE 5. Decay of ^{147}Pm to ^{147}Sm

Time, Days	Composition, %		Time, Days	Composition, %	
	^{147}Pm	^{147}Sm		^{147}Pm	^{147}Sm
1	99.928	0.072	150	89.711	10.289
2	99.855	0.145	200	86.530	13.470
3	99.783	0.217	250	83.456	16.544
4	99.711	0.289	300	80.492	19.508
5	99.639	0.361	350	77.632	22.368
6	99.567	0.433	400	74.875	25.125
7	99.495	0.505	450	72.215	27.785
8	99.423	0.577	500	69.649	30.351
9	99.351	0.649	550	67.175	32.825
10	99.279	0.721	600	64.789	35.211
20	98.564	1.436	650	62.487	37.513
30	97.853	2.147	700	60.268	39.732
40	97.148	2.852	750	58.127	41.873
50	96.448	3.552	800	56.062	43.938
60	95.752	4.248	850	54.070	45.930
70	95.062	4.938	900	52.150	47.850
80	94.377	5.623	950	50.297	49.703
90	93.697	6.303	1000	48.510	51.490
100	93.022	6.978	2000	23.532	76.468

Selected data are summarized in Table 6 as percent abundance for important beta particles and gamma or bremsstrahlung photons, and as particles or photons per Watt second from typical promethium (0.25 ppm ^{146}Pm , 0.025 ppm ^{148}Pm , 0.001 ppm ^{154}Eu).

FIGURE 6. Decay Scheme of ^{147}Pm

FIGURE 7. Decay Scheme of ^{146}Pm

FIGURE 8. Decay Schemes of ^{148m}Pm and ^{148}Pm

FIGURE 9. Decay Scheme of ^{154}Eu

TABLE 6. Principal Radiations from Typical Promethium

Type of Radiation	Energy, MeV	Source Nuclide	Abundance, %	Number per Watt-second	Reference
a. Alpha		none			37
b. Beta	0.2246	^{147}Pm	100	1.05×10^{14}	38
	0.103	^{147}Pm	0.008	8.2×10^9	
	0.790	^{146}Pm	32	8.2×10^6	39
	2.467	$^{148\text{m}}\text{Pm}$, ^{148}Pm	5	7.7×10^4	30
	1.855	^{154}Eu	7	7.2×10^3	40
c. Gamma	0.1212	^{147}Pm	0.004	4.1×10^9	38
	1.585	^{146}Pm	0.006	1.5×10^5	39
	1.190	^{146}Pm	0.05	1.3×10^4	
	0.7474	^{146}Pm	34	8.8×10^6	
	0.7562	^{146}Pm	21	5.4×10^6	
	0.4556	^{146}Pm	66	1.7×10^7	
	1.4651	$^{148\text{m}}\text{Pm}$, ^{148}Pm	1.3	5.3×10^4	30
	1.0058	$^{148\text{m}}\text{Pm}$	20	5.2×10^5	
	0.9152	$^{148\text{m}}\text{Pm}$	19	4.9×10^5	
	0.7257	$^{148\text{m}}\text{Pm}$	32	8.2×10^5	
	0.6209	$^{148\text{m}}\text{Pm}$	88	2.3×10^6	
	0.5502	$^{148\text{m}}\text{Pm}$	95	2.4×10^6	
	1.596	^{154}Eu	2	2.1×10^5	40
	1.496	^{154}Eu	0.7	7.2×10^2	
	1.278	^{154}Eu	37	5.8×10^4	
	1.038	^{154}Eu	19	2.0×10^4	
	1.000	^{154}Eu	11	1.4×10^4	
d. Bremsstrahlung	0.21	^{147}Pm	4.2×10^{-7}	4.3×10^5	41
	0.20	^{147}Pm	3.4×10^{-6}	3.5×10^6	
	0.19	^{147}Pm	1.4×10^{-5}	1.4×10^7	
	0.18	^{147}Pm	5.9×10^{-5}	4.0×10^7	
	0.17	^{147}Pm	9.4×10^{-5}	9.7×10^7	
	0.16	^{147}Pm	2.0×10^{-4}	2.1×10^8	
	0.15	^{147}Pm	5.8×10^{-4}	5.9×10^8	
	0.14	^{147}Pm	6.9×10^{-4}	7.1×10^8	
	0.13	^{147}Pm	1.2×10^{-3}	1.2×10^9	
	0.12	^{147}Pm	2.0×10^{-3}	2.1×10^9	
	0.11	^{147}Pm	3.2×10^{-3}	3.3×10^9	
	0.10	^{147}Pm	5.1×10^{-3}	5.3×10^9	
	0.09	^{147}Pm	8.0×10^{-3}	8.2×10^9	
	0.08	^{147}Pm	0.012	1.2×10^{10}	
	0.07	^{147}Pm	0.019	2.0×10^{10}	
	0.06	^{147}Pm	0.031	3.2×10^{10}	
	0.05	^{147}Pm	0.049	5.1×10^{10}	
	0.04	^{147}Pm	0.082	8.5×10^{10}	
	0.03	^{147}Pm	0.13	1.5×10^{11}	
	0.02	^{147}Pm	0.30	3.1×10^{11}	
	0.01	^{147}Pm	0.86	8.9×10^{11}	

NEUTRON CROSS SECTIONSTABLE 7. Neutron Cross Sections of Promethium Isotopes

<u>Isotope</u>	<u>Cross Section, barns</u>				<u>Reference</u>
	<u>Thermal</u>	<u>Resonance Integral</u>	<u>Reactor</u>	<u>Fission Spectrum</u>	
^{147}Pm	154	1,420	315	0.0059	42,43
^{146}Pm			8,400		44
$^{148\text{m}}\text{Pm}$	22,000	32,000	24,000		45

BIOLOGICAL TOLERANCESTABLE 8. Permissible Environmental Concentrations of Promethium

Form	Maximum Permissible Concentration, $\mu\text{Ci/ml}$				Reference
	40-hr Week		168-hr Week		
	Air	Water	Air	Water	
Soluble	6×10^{-8}	6×10^{-3}	2×10^{-9}	2×10^{-4}	46
Insoluble	1×10^{-7}	6×10^{-3}	3×10^{-9}	2×10^{-4}	

TABLE 9. Biological Distribution

<u>Organ</u>	<u>Biological Half-Life, days</u>	<u>Permissible Burden, μCi</u>	<u>Fraction Reaching Organ</u>		<u>Reference</u>
			<u>Ingested</u>	<u>Inhaled</u>	
Total Body	656	300	1×10^{-4}	0.25	47
Bone	1500	60	3.5×10^{-5}	0.09	
Liver	656	300	6×10^{-6}	0.02	
Kidneys	656	200	2×10^{-6}	5×10^{-3}	

SHIELDINGTABLE 10. Comparison of Shielding Effectiveness

Material	Shielding for 100X Attenuation,		Reference
	<u>g/cm²</u>	<u>cm</u>	
Uranium	2.0 to 50	0.11 to 2.6	48
Lead	1.5 to 59	0.13 to 5.2	
Tungsten	1.7 to 66	0.09 to 3.4	
Molybdenum	4.8 to 93	0.47 to 9.1	
Iron	15 to 103	1.9 to 13.1	

(Shielding effectiveness varies with source size and shape and amount of other external shielding.)

Effects of uranium shielding on dose rates are shown in Figure 10 as a function of source size. The difference in shielding requirements for ¹⁴⁶Pm and ¹⁴⁷Pm is shown for thin rhenium shields in Figure 11.

PROMETHIUM METALTYPICAL COMPOSITION

<u>Constituent</u>	<u>Concentration, wt%</u>	<u>Reference</u>
¹⁴⁷ Pm and ¹⁴⁷ Sm	>99.78	49
Neodymium	<0.01	
Calcium	<0.02	
Magnesium	<0.04	
Oxygen	<0.10	
Other	<0.05	

The concentration values given are for promethium metal as obtained by the calcium reduction of ¹⁴⁷PmCl₃ in a magnesia container. The ¹⁴⁷PmCl₃ was formed from ¹⁴⁷Pm₂O₃ heart cut of 99.95% purity with respect to all impurities except for its decay product, ¹⁴⁷Sm₂O₃. The as-prepared metal was 97.8 wt% ¹⁴⁷Pm.

FIGURE 10. Dose Rates from Promethium Sources Containing 0.25 ppm ^{146}Pm

FIGURE 11. Expected Dose Rate at 6 in. from a 60 W Promethium Source 1 in. Diameter $\times$ 2.56 in. Long Inside

Higher purity metal may be prepared by this method. ¹⁴⁷Pm metal containing less than 0.02 wt% metallic impurities, besides the ¹⁴⁷Sm decay product, could reasonably be anticipated. (49)

PHYSICAL PROPERTIES

The values enclosed by brackets, [], are approximations derived from measured properties of neighboring rare earths.

	<u>Value</u>	<u>Reference</u>
a. Crystal Structure (Å)		
α-form, double hexagonal (low temperature allotrope)	a = 3.66 ± 0.01 c = 11.80 ± 0.04	50,51
b. Density (g/cm ³), low temperature allotrope, temperature ~35 °C		
calculated from X-ray data	7.13 ± 0.07	50
experimental	7.22 ± 0.02	49,52
c. Phase Transition Temperature (°C)		
[α-β] average of values for neodymium and samarium	[890 ± 8]	53
melting point	1168 ± 6	52,49
boiling point	[2460]	53
d. Heat of Phase Transition (kcal/mole)		
[α-β] average of values for neodymium and samarium	[0.73]	53
heat of fusion	[1.94]	53
	[1.95]	54
heat of sublimation, ΔH ^o ₂₉₈	[64]	53
heat of vaporization	[60]	55
e. Coefficient of Linear Thermal Expansion (cm/cm - °C)	[9.0 × 10 ⁻⁶]	53

	<u>Value</u>	<u>Reference</u>
f. Heat Capacity (cal/mole - °C)	[6.50]	53
g. Vapor Pressure, P(mm Hg), T is in °K		
for neodymium, 1255- 1650 °C	$\log P = 8.102 \pm 0.034$ - $\frac{16320 \pm 59}{T}$	53
for samarium, 612- 949 °C	$\log P = 8.58 \pm 0.20$ - $\frac{10656 \pm 128}{T}$	53
h. Thermal Diffusivity (cm ² /sec)		
Calculated for 27 °C using b, f and i	[0.14 ± 0.02]	--
i. Thermal Conductivity (W/cm - °C)		54
27 °C	0.18 ± 0.02	
727 °C	0.21 ± 0.02	
2000 °C	0.23 ± 0.13	
j. Ionization Potentials (eV)	I = 5.55 II = 10.90	53
k. Grüneisen Constant	[0.81]	53
l. Viscosity	--	
m. Surface Tension (dynes/ cm) at 2,300 °C	[40-70]	56
n. Total Hemispherical Emissivity	--	
o. Spectral Emissivity	--	

MECHANICAL PROPERTIES

a. Hardness, Knoop (KHN) (cast metal)		51
for samarium	45	
for promethium	63	
b. Mechanical Strength	--	
c. Compressibility (cm ² /kg)	[2.78 × 10 ⁻⁶]	53
d. Young's Modulus (kg/cm ²)	[4.3 × 10 ⁵]	53
e. Shear Modulus (kg/cm ²)	[1.7 × 10 ⁵]	53
f. Poisson's Ratio	[0.278]	53

	<u>Value</u>	<u>Reference</u>
--	--------------	------------------

CHEMICAL PROPERTIES

a. Thermodynamic Functions

• S° (cal/mole-deg) 300 °K	[17.1]	57
• $\frac{H^\circ_t - H^\circ_o}{T}$ 300 °K (cal/mole-deg)	[6.07]	57
• $\frac{F^\circ_t - H^\circ_o}{T}$ 300 °K (cal/mole-deg)	[11.0]	57

b. Chemical Reactions

Promethium metal can be expected to react in the same general fashion as other rare earth metals. Typical reactions include the following:

$2 \text{ Pm} + 3/2 \text{ O}_2 \xrightarrow{>25^\circ \text{C}}$	Pm_2O_3	55
$2 \text{ Pm} + 3 \text{ S} \xrightarrow{500^\circ \text{C}}$	Pm_2S_3	58
$\text{Pm} + x/2 \text{ H}_2 \xrightarrow{>25^\circ \text{C}}$	PmH_x	58
$\text{Pm} + 1/2 \text{ N}_2 \xrightarrow{900^\circ \text{C}}$	PmN	58
$\text{Pm} + 3 \text{ H}_2\text{O} \xrightarrow{\hspace{1cm}}$	$\text{Pm}(\text{OH})_3 + 3/2 \text{ H}_2$	59
$\text{Pm} + 3/2 \text{ X}_2 \xrightarrow{200^\circ \text{C}}$	PmX_3	53
X = a halogen		
$\text{Pm} + \text{mineral acid} \xrightarrow{\hspace{1cm}}$	dissolution	53
$\text{or alkaline solution} \xrightarrow{\hspace{1cm}}$	of Pm	

The following promethium compounds have been prepared by various investigators: (2,60,61) $\text{Pm}_2\text{O}_3\text{-A}$, $\text{Pm}_2\text{O}_3\text{-B}$, $\text{Pm}_2\text{O}_3\text{-C}$, PmCl_3 , $\text{PmCl}_3 \cdot x\text{H}_2\text{O}$, PmOCl , PmBr_3 , PmOBr , PmF_3 , PmOF , PmOI , $\text{Pm}(\text{IO}_3)_3$, $\text{Pm}(\text{NO}_3)_3$, $\text{Pm}(\text{OH})_3$, $\text{Pm}_2(\text{C}_2\text{O}_4)_3$,* $\text{Pm}_2(\text{C}_2\text{O}_4)_3 \cdot 3\text{H}_2\text{O}$, PmPO_4 , PmAsO_4 , $\text{Pm}_2(\text{CrO}_4)_3$, PmVO_4 , PmCrO_3 , PmBO_3 , PmScO_3 , PmInO_3 , $\text{Pm}_2(\text{MoO}_4)_3$, $\text{Pm}(\text{WO}_4)_3$, $\text{Pm}(\text{HCOO})_3$, $\text{Pm}(\text{CCl}_3\text{COO})_3$, $\text{Pm}_2(\text{CO}_3)_3 \cdot 2\text{H}_2\text{O}$.

* The decomposition of promethium oxalate to Pm_2O_3 has been investigated using differential thermal analysis and thermogravimetric analysis. (62)

	Value	Reference
c. Solubility in H ₂ O	--	
d. Diffusion Rates, D, (cm ² /sec)		
Through sintered Al ₂ O ₃ ,	D = 1.18 × 10 ⁸	62
T is in °K	× exp(-157,000R/T)	
R = 1.99 cal/g-mole °K		
range, 1350-1500 °C		

COMPATIBILITY WITH MATERIALS OF CONTAINMENT

Compatibility screening tests have been completed using adjacent rare earths as stand-ins for promethium. The tests were conducted at temperatures above and below the rare earth melting point. The results are summarized in Table 11⁽⁶³⁾ and Table 12.^(64,65)

TABLE 11. Metal-Containment Material Compatibility Using Samarium Metal and Neodymium Metal as Stand-ins for ¹⁴⁷Pm Metal

Containment Material	Compatibility ^(a)	
	720 hr at 600 °C ^(b)	575 hr at 1100 °C ^(c)
Tantalum	Excellent	Excellent
Molybdenum	Excellent	Poor
Zirconium	Excellent	Poor
Titanium	Good	Fair
TZM	Excellent	Poor
Platinum	--	Poor
Hastelloy-C	Fair	Poor
Hastelloy-N	--	Poor
Hastelloy-X	--	Poor
Haynes-25	--	Poor
310-Stainless Steel	Fair	Poor
Inco-600	Poor	Poor
Rene-41	Poor	Poor
Carpenter-20	--	Poor
N-Naval Brass	Fair	--

NOTES:

(a) Compatibility

Excellent - no visible attack
 Good - <1 mil penetration
 Fair - 1 to 10 mil penetration
 Poor - >10 mil penetration

(b) 600 °C - below the melting point of the rare earth.

(c) 1100 °C - above the melting point of the rare earth:

Rare Earth	Melting Point, °C
Neodymium	1018 ± 5
Samarium	1073 ± 1

**TABLE 12. Metal-Containment Material Compatibility Using
Samarium Metal as a Stand-in for ^{147}Pm Metal**

Containment Material	Compatibility ^(a)							
	1000 hr 400 °C	4400 hr 400 °C	8800 hr 400 °C	1000 hr 700 °C	4400 hr 700 °C	8800 hr 700 °C	1000 hr 1000 °C	4400 hr 1000 °C
Tantalum	<1	<1	<1	<1	<1	<1	<1	<1
Ta-10W	<1	<1	<1	<1	<1	<1	<1	<1 ^(c)
TZM	<1	<1	<1	<1	<1	10	<1	<1 ^{(b),(c)}
Titanium	<1	<1	<1	<1	<1	<1	<1	<1 ^(c)
Zirconium	<1	<1	<1	<1	<1	<1	<1	-
Inconel	<1	<1	80	125	125	125	75	-
Haynes-25	<1	<1	40	60	50	50 ^(c)	75	-
Hastelloy-X	<1	<1	75	65	63	65 ^(c)	100	-
304L SS	<1	<1	20	40	53	50	125	-
316L SS	<1	<1	30	60	61	55	125	-

NOTES:

- (a) *Compatibility - maximum penetration of the interaction zone into the 125 mil containment material wall, mil.*
- (b) *3400 hr test.*
- (c) *There was evidence that samarium metal may have been released from the capsule--probably in the weld area. The samarium release appeared to be due to lack of weld integrity since the interaction between the capsule wall material and samarium metal was, in most cases, minimal.*

PROMETHIUM SESQUIOXIDE, Pm_2O_3 TYPICAL COMPOSITION

<u>Constituent</u>	<u>Concentration, wt%</u>	<u>Reference</u>
$^{147}\text{Pm}_2\text{O}_3$ and $^{147}\text{Sm}_2\text{O}_3$	>98.00	66
Nd_2O_3	<1.00	
Sodium	<0.01	
Other	<0.99	

The concentration values given are for a typical 50,000 Ci batch purification run made at the Pacific Northwest Laboratory. Batches of up to 200,000 Ci may be processed in the same equipment with no anticipated change in purity. At the time of shipment from PNL, the typical batch was 97.0% $^{147}\text{Pm}_2\text{O}_3$, and 2.0% $^{147}\text{Sm}_2\text{O}_3$ decay product.

The Nd_2O_3 impurity is present because neodymium is next to promethium in the ion-exchange elution sequence.^(35,67) Lower neodymium contents may be obtained by adjusting the relative amounts of high purity (over 99.9%) promethium fraction and promethium-neodymium mid-fraction recovered in the purification run.

PHYSICAL PROPERTIES

The values enclosed in brackets, [], are approximations derived from measured properties of the neighboring rare earth sesquioxides.

	<u>Value</u>	<u>Reference</u>
a. Crystal Structure ($\text{\AA}$)		68
C-form bcc	$a = 10.99$	
B-form monocl	$a = 14.22$ $c = 8.91$	
	$b = 3.65$ $\beta = 100.1^\circ$	
A-form hcp	$a = 3.81$ $c = 6.03$	
H-form	--	
X-form	--	

	<u>Value</u>	<u>Reference</u>
b. Density (g/cm^3)		69
calculated from	C-form 6.85	
X-ray data	B-form 7.48	
	A-form 7.49	
	H-form --	
	X-form --	
c. Phase Transition Temperature ($^{\circ}\text{C}$)		
C-B	Figure 12	68
B-A	} Table 13 and Figure 12	68
A-H		
H-X		
Melting Point		
Boiling Point	[>3000 $^{\circ}\text{C}$]	57

TABLE 13. Temperatures ($^{\circ}\text{C}$) for B $\rightleftharpoons$ A, A $\rightleftharpoons$ H, H $\rightleftharpoons$ X, and X $\rightleftharpoons$ Liquid on Compositions Containing 98, 53, 26, 9, and 0 wt% $^{147}\text{Pm}_2\text{O}_3$.

<u>Composition</u>	<u>B $\rightleftharpoons$ A</u>	<u>A $\rightleftharpoons$ H</u>	<u>H $\rightleftharpoons$ X</u>	<u>X $\rightleftharpoons$ Liquid</u>
($\text{Pm}_{0.98}\text{Sm}_{0.02}$) $_2\text{O}_3$	1740	2135 ^(a)	2225	2320
($\text{Pm}_{0.53}\text{Sm}_{0.47}$) $_2\text{O}_3$	1740	2115	2250 ^(b)	2360
($\text{Pm}_{0.26}\text{Sm}_{0.74}$) $_2\text{O}_3$	1780 ^(c)	2065 ^(d)	2245	2330
($\text{Pm}_{0.09}\text{Sm}_{0.91}$) $_2\text{O}_3$	1880	2112	2252	2325
Sm_2O_3	1930	2160	—	2365

NOTES:

- (a) Originally peak at 1930 which changed to 2135 $^{\circ}\text{C}$ after a few heating - cooling cycles.
- (b) Originally peak at 2035 which disappeared after a few heating - cooling cycles.
- (c) Originally peak at 1825 which changed to 1780 $^{\circ}\text{C}$ after a few heating - cooling cycles.
- (d) Originally peak at 2140 which changed to 2065 $^{\circ}\text{C}$ after a few heating - cooling cycles.

FIGURE 12. The Pm_2O_3 - Sm_2O_3 Phase Diagram. (The two phase regions are extremely narrow and are represented by a single line)

	Value	Reference
d. Heat of Transition (kcal/mole)		
C-B	--	
B-A	--	
A-H	--	
H-X	--	
Fusion	--	
Sublimation	266	
assuming $^{147}\text{Pm}_2\text{O}_3(\text{s}) \longrightarrow 2 \text{ } ^{147}\text{PmO}(\text{g}) + \text{O}(\text{g})$		68
e. Coefficient of Linear Thermal Expansion (cm/cm-°C)	Figure 13	70
($^{147}\text{Pm}_{0.88} \text{ } ^{147}\text{Sm}_{0.12}$) 2O_3	9.4×10^{-6}	70
($^{147}\text{Pm}_{0.68} \text{ } ^{147}\text{Sm}_{0.32}$) 2O_3	9.8×10^{-6}	70
($^{147}\text{Pm}_{0.00} \text{ } ^{147}\text{Sm}_{1.00}$) 2O_3	10.6×10^{-6}	71
f. Heat Capacity (cal/mole-°C)	Figure 14	68
	Figure 15	68
g. Vapor Pressure (Atm)	--	
h. Rate of Effusion (g/cm ² -sec)	Figure 16	68
i. Thermal Conductivity, k, (Watts/cm-°C)	Figure 17	72
Values of A and B for $k = 1/(AT+B)$ and 100 °C < T °C < 1000 °C:		72
<u>Sample Composition</u>	<u>A</u>	<u>B</u>
($^{147}\text{Pm}_{0.98} \text{ } ^{147}\text{Sm}_{0.02}$) 2O_3	0.0261	38.6
($^{147}\text{Pm}_{0.88} \text{ } ^{147}\text{Sm}_{0.12}$) 2O_3	0.0259	42.8
($^{147}\text{Pm}_{0.84} \text{ } ^{147}\text{Sm}_{0.16}$) 2O_3	0.0231	46.8
($^{147}\text{Pm}_{0.80} \text{ } ^{147}\text{Sm}_{0.20}$) 2O_3	0.0280	45.2

FIGURE 13. Thermal Expansion of $(^{147}\text{Pm}, ^{147}\text{Sm})_2\text{O}_3$

FIGURE 14. The Heat Capacity of Cubic Pm₂O₃ Estimated from the Data Shown for Hexagonal Nd₂O₃ and Cubic Sm₂O₃. The data for cubic Gd₂O₃ illustrate the heat capacity of a material with essentially no electronic contributions.

FIGURE 15. The Heat Capacity of Monoclinic Pm_2O_3 Estimated from the Data Shown for Hexagonal Nd_2O_3 and Monoclinic Sm_2O_3

FIGURE 16. Rate of Effusion of (¹⁴⁷Pm_{0.96} ¹⁴⁷Sm_{0.04})₂O₃ from a Rhenium Knudsen Cell

FIGURE 17. Thermal Conductivity of (^{147}Pm , ^{147}Sm) 2O_3

	Value	Reference
j. Thermal Diffusivity (cm ² /sec)	Figure 18	73
k. Viscosity	--	
l. Surface Tension (dynes/cm) for Sm ₂ O ₃	962 ± 53	68
m. Total Hemispherical Emissivity	--	
n. Spectral Emissivity	--	
o. Electrical Resistivity (ohm-cm)	Figure 19	72,74

MECHANICAL PROPERTIES

a. Hardness	--
b. Crush Strength	--

CHEMICAL PROPERTIES

a. Thermodynamic Functions

• Enthalpy of formation, ΔH_f° , (kcal/mole) at 300 °K		
Nd ₂ O ₃ - Hexagonal	-432.36 ± 0.21	75
Sm ₂ O ₃ - Monoclinic	-433.88 ± 0.48	75
• Gibbs energy of forma- tion, ΔG_f° , (kcal/ mole) at 300 °K		
Nd ₂ O ₃ - Hexagonal	-411.03 ± 0.25	75
Sm ₂ O ₃ - Monoclinic	-412.68 ± 0.49	75
• Entropy of formation, ΔS_f° , (cal/mole-deg) at 300 °K		
Nd ₂ O ₃ - Hexagonal	-71.10 ± 0.46	75
Sm ₂ O ₃ - Hexagonal	-70.66 ± 0.35	75
• Entropy, S°_{298} , (cal/mole-deg) for ¹⁴⁷ Pm ₂ O ₃	[37.2]	76

FIGURE 18. Thermal Diffusivity of $^{147}\text{Pm}_2\text{O}_3$ - $^{147}\text{Sm}_2\text{O}_3$ Solid Solutions

FIGURE 19. Resistivity of $^{147}\text{Pm}_2\text{O}_3$

	Value	Reference
b. Chemical Reactions		
Promethium oxide can be expected to react in the same general fashion as the oxides of samarium and neodymium. Typical reactions include:		
$\text{Pm}_2\text{O}_3 + 6 \text{HCl (aq)} \longrightarrow$	$2 \text{PmCl}_3 \cdot x \text{H}_2\text{O}$	
$\text{Pm}_2\text{O}_3 + 6 \text{HNO}_3 \longrightarrow$	$2 \text{Pm}(\text{NO}_3) + 3 \text{H}_2\text{O}$	
$\text{Pm}_2\text{O}_3 + 6 \text{HF} \xrightarrow{700^\circ\text{C}}$	$2 \text{PmF}_3 + 3 \text{H}_2\text{O}$	
$\text{Pm}_2\text{O}_3 + 7 \text{C} \xrightarrow{1000^\circ\text{C}}$	$2 \text{PmC}_2 + 3 \text{CO}$	
c. Solubility in Distilled Water (g/liter)		
at 25 °C, pH = 8	0.003	73
d. Dissolution Rates (µg/cm ² -hr)		
• 80% theoretical density Pm_2O_3 pellets sintered at 1400 °C		
seawater	0.04	2,73
distilled water	3.80	2,73
• Pm_2O_3 powder calcined at 850 °C		
distilled water	>8.0	2,73
e. Diffusion Rates	--	

COMPATIBILITY WITH MATERIALS OF CONTAINMENT

Compatibility test results are summarized in Tables 14 through 17.

TABLE 14. (52, 77)

Oxide-Containment Material Compatibility
at 1100 °C Using Sm_2O_3 as a Stand-in
for Pm_2O_3

Containment Material	Fabrication Method	Compatibility ^(a)			
		1500 hr	4300 hr	8800 hr	17,600 hr
304 L	SP ^(b)	-	<1	<1	<1
304 L	PI ^(c)	1	2	1	1
Haynes-25	SP	-	<1	1	<1
Haynes-25	PI	<1	1	2	<1
Hastelloy-X	SP	-	<1	1	<1
Hastelloy-X	PI	1	1	2	1
Tantalum	SP	-	1	2	1
Tantalum	PI	2	2	2	2
Rhenium	SP	-	0	0	0
Rhenium	PI	0	0	0	0
TZM	SP	-	<1	<1	<1
TZM	PI	<1	<1	<1	<1
W-25Re	SP	-	<1	<1	<1
W-25Re	PI	<1	<1	<1	<1
Ta-10W	SP	-	<1	1	1
Ta-10W	PI	-	<1	1	1
Tungsten	SP				<1(d)

NOTES:

- (a) Compatibility - maximum penetration of the interaction zone into the containment material, mil.
- (b) SP - oxide step-pressed into capsule.
- (c) PI - oxide step-pressed into capsule and capsule pneumatically impacted.
- (d) Result of 1200 hr test.

Greater interaction was observed in some tests when test couples were subjected to the thermocycling. In Sm_2O_3 - containment material compatibility tests, where the temperature was cycled between 700 °C and 1100 °C every 24 hours, interaction zones in some tests penetrated deeper into the containment

material than the penetration depths shown in Table 14 for equivalent material-test time combinations.⁽⁵²⁾

TABLE 15. ^(52,77) Oxide-Containment Material Compatibility
Using Sm_2O_3 as a Stand-in for Pm_2O_3

<u>Containment Material</u>	<u>1000 hr 1550 °C</u>	<u>4300 hr 1550 °C</u>	<u>1000 hr 2000 °C</u>	<u>3000 hr 2000 °C</u>
Tantalum	2	2	<1	1
Ta-10W	1	1	<1	2
TZM	1	<1	-	-
Tungsten	-	0	<1	1
Rhenium	0	0	0	<1
W-26Re	0	1	<1	<1

NOTES:

- (a) *Compatibility - maximum penetration of the interaction zone into the containment material, mil. The oxide was step-pressed into the capsule and pneumatically impacted.*

TABLE 16. (78) Oxide-Containment Material Compatibility Using Sm_2O_3 or Nd_2O_3 as a Stand-in for Pm_2O_3

Containment Material	Compatibility - Average Penetration into Containment Material, mil									
	1600 °C			2000 °C			2500 °C			
	Nd_2O_3	Sm_2O_3	Sm_2O_3	Sm_2O_3	Sm_2O_3	Sm_2O_3	Sm_2O_3	Nd_2O_3	Sm_2O_3	Nd_2O_3
	2000 hr	2800 hr	5000 hr	1300 hr	4400 hr	5100 hr	72 hr	72 hr	132 hr	132 hr
Mo-50Re	<0.2		<<0.1							
Molybdenum	0.2		1							
W-30Mo-30Re	0.5		<0.1							
Tantalum		0.6		4						
Ta-10W		0.4		<4						
Tungsten						4	25		12	3-4
W-25Re						<0.1		5	10	
Rhenium					<0.1		2	~10		

TABLE 17.⁽⁷⁷⁾ Pm_2O_3 -Containment Material Compatibility

Containment Material	Fabrication Method	Compatibility ^(a)		Fabrication Method	13200 hr 1100 °C
		8800 hr 1100 °C			
Tantalum	PS ^(b)	0		PS	0
Tantalum	PI ^(c)	<0.5		--	—
Ta-10W	PS	0		PS	0
Ta-10W	PI	0		--	—
Tungsten	PS	0 ^(d)		PS	0 ^(d)
Rhenium	PS	0		PS	0
W-26Re	PS	0		PS	0
TZM	PS	0		--	—
Haynes-25	PS	0		--	—
Haynes-25	PI	<0.5		--	—
Hastelloy-X	PS	0		--	—
Hastelloy-X	PI	<0.5		--	—
304L SS	PS	<0.5		--	—
304L SS	PI	<0.5		--	—

NOTES:

- (a) Compatibility - maximum penetration of the interaction zone into the containment material, mil.
- (b) PS - $^{147}Pm_2O_3$ was cold-pressed into pellets which were sintered for 16 hr at 1250 °C, and encapsulated in contact with the containment material.
- (c) PI - $^{147}Pm_2O_3$ was cold-pressed into pellets which were sintered for up to 32 hr at up to 1250 °C. The pellets were then encapsulated in contact with the containment material and subsequently pneumatically impacted.⁽⁷⁶⁾
- (d) Some intergranular cracking was observed.

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