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HANFORD ¹⁴⁶Pm BURNOUT EXPERIMENTS

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ERRATA

Page 14, 4th line from bottom

$$dN_{46}/dt = \sigma N_{47} - \Lambda_{47} N_{46}.$$

should be

$$dN_{46}/dt = \sigma N_{47} - \Lambda_{46} N_{46}.$$

Page 15, top line

$$\text{Since the quantity } (\sigma_{2n} \phi / \Lambda_{46} - \Lambda_{47}) \quad (\lambda_{46} / \lambda_{47})$$

should be

$$\text{Since the quantity } (\sigma_{2n} \phi / [\Lambda_{46} - \Lambda_{47}]) \quad (\lambda_{46} / \lambda_{47})$$

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HANFORD ^{146}Pm BURNOUT EXPERIMENTS

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January 1970

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ABSTRACT

Hanford ^{147}Pm , containing about 0.3 ppm ^{146}Pm , was irradiated in a Hanford K-Reactor in an attempt to burn out the ^{146}Pm present and thereby reduce the gamma dose rate to a level where ^{147}Pm could be used in circulatory assist devices. The ^{146}Pm content was reduced by a factor of two for the longest irradiation, which was less than the expected result. A cross section of 3500 b was found for ^{146}Pm , compared to 8400 b found by Pinajian, which partially explains the low ^{146}Pm burnout. The ^{146}Pm cross section varied slightly with position in the target, suggesting resonance capture of epithermal neutrons. Explanations for the low burnout of ^{146}Pm are examined and possible ways of further reducing the ^{146}Pm content are suggested.

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INTRODUCTION

In the purification of promethium, non-promethium radioactivity can be reduced to an insignificant level by the ion exchange process.⁽¹⁾ Thus, in well-aged promethium (from which ^{148}Pm has decayed), the principal penetrating radiation is due to ^{146}Pm , and this dictates the shielding requirements of a promethium fueled heat source.⁽²⁾ For many applications, the dose rate due to ^{146}Pm is not much more than a nuisance, but for other applications, such as circulatory assist devices, the amount of required shielding may be excessive for allowable dose rates. Promethium generated in power reactors contains substantially larger amounts⁽³⁾ of ^{146}Pm than that produced in AEC production reactors, and will be somewhat more limited in usefulness unless the ^{146}Pm content can be reduced. The ^{146}Pm content cannot be reduced by aging (as can ^{148}Pm) since its half-life⁽⁴⁾ is greater than that of ^{147}Pm .⁽⁵⁾ separation of the promethium isotopes is perhaps technically possible, but not economically attractive.⁽⁶⁾ Therefore, consideration has been given to using the differences in neutron cross sections of ^{146}Pm and ^{147}Pm to achieve a reduction in content of the unwanted isotope. While this possibility has been previously considered for pure thermal flux conditions,⁽⁷⁾ the following sections examine the influence of resonance and fast neutrons, and report the results of experiments which were performed to test promethium burnout.

The three promethium isotopes of practical interest, their half-lives, and neutron cross sections^(8,9,10) are listed in Table 1.

TABLE 1. Neutron Cross Sections of Promethium Isotopes

<u>Isotope</u>	<u>Half-Life</u>	<u>Cross Section, b</u>			
		<u>Thermal</u>	<u>Resonance Integral</u>	<u>Reactor*</u>	<u>Fission Spectrum</u>
^{147}Pm	2.62 years	154	2,300	315	0.0059
^{146}Pm	5.53 years			8,400	
$^{148\text{m}}\text{Pm}$	42 days	22,000	32,000	24,000	

* *Westcott Spectral index, $r = 0.07$*

Neodymium-146, -147 and -148 are formed in the fission reaction either directly or as daughters of short-lived parents. Neodymium-147 then decays to form ^{147}Pm . The 147 chain is formed in about 2.1% of the fissions, depending slightly on the fissioning nuclide.⁽¹¹⁾ Most of the 146 and 148 chains result in stable neodymium isotopes. Promethium-146 is formed as a direct fission product in about $10^{-6}\%$ of fissions.⁽¹²⁾ The fission yield of the ^{148}Pm isomers is negligible since much more is formed from ^{147}Pm by neutron capture. Promethium-146 is also formed from ^{147}Pm by (n,2n) reactions. Since this reaction requires high energy neutrons (above 7.684 MeV), the production of ^{146}Pm is dependent on the neutron spectrum in which the promethium is formed. Under favorable conditions, such as relatively short exposures in the well-moderated Hanford reactors, the amount of ^{146}Pm is fission promethium is about 2.5×10^{-7} Ci per curie of ^{147}Pm .

Since the neutron capture cross section of ^{146}Pm is greater than that of ^{147}Pm , irradiation of fission product promethium should reduce the relative ^{146}Pm content. In order to ascertain whether such a process is technically practical, it is necessary to determine (1) the loss of ^{147}Pm during irradiation and (2) the realistic limit of ^{146}Pm depletion.

Promethium-147 loss during irradiation occurs by decay and by capture of both thermal and resonance neutrons. Since the decay process is determined only by the half-life of the isotope, and the capture process by the integrated flux to which the target is exposed, the use of high fluxes and short irradiation times are desirable.

The total loss of ^{147}Pm is dependent on flux characteristics, flux intensity, and the duration of exposure. Fluxes as low as 10^{13} n/cm²-sec result in an excessive loss of ^{147}Pm by radioactive decay during irradiation. Table 2 shows the relative effect of decay versus neutron capture for three neutron fluxes. Irradiation at a flux of 10^{14} n/cm²-sec of purely thermal neutrons will reduce the $^{146}\text{Pm}/^{147}\text{Pm}$ ratio by a factor of 10 for every 35 days of irradiation--if an 8400 b cross section for ^{146}Pm can be assumed. Calculations indicate a 70-day irradiation should reduce the ^{146}Pm content by a factor of 100 with a loss of only 18% of the original promethium (13% by neutron capture and 4.6% by radioactive decay).

TABLE 2. Fraction of Promethium Isotopes Disappearing by Decay in Irradiation in Thermal Neutron Fluxes

Isotope	Loss by Decay as % of Total Loss Rate		
	$\phi = 10^{13}$	$\phi = 10^{14}$	$\phi = 10^{15}$
^{146}Pm	78.0	26.0	3.40
^{147}Pm	5.5	0.6	0.06
^{148}Pm	41.0	6.6	0.70

In the above calculation, it was assumed that no nuclear reactions were regenerating ^{146}Pm . Actually, reactions do exist because ^{146}Pm can be produced by the (n,2n) and (γ,n) reactions of ^{147}Pm with neutrons and gamma rays having energies

EXPERIMENTAL WORK

TARGET ELEMENT DESIGN

Four promethium target elements and one neodymium target element were designed and fabricated. They were used to determine the "burnout" rates (and equilibrium content) of ^{146}Pm in a well-moderated reactor as a function of integrated exposure. The promethium target elements were fabricated semi-remotely from short cooled promethium which had been purified by ion exchange. (During the irradiation, ^{148}Pm and $^{148\text{m}}\text{Pm}$ will quickly assume equilibrium values, so the initial presence of these nuclides will not complicate the interpretation of the results.)

The macroscopic cross section of ^{147}Pm , with its equilibrium daughters at a flux of 10^{14} n/cm²-sec, is about 40 times that of natural uranium. Therefore, the target element must be very small (as compared to a fuel element) to avoid a large depression of the thermal neutron flux. For example, a thickness of 0.07 in. of high density promethium sesquioxide will reduce the thermal flux by a factor of two. The resonance flux depression in the vicinity of the large ^{147}Pm resonance is much more pronounced. Removal of the resonance flux is probably desirable since even with a spectral index of 0.07, much of the ^{148}Pm and $^{148\text{m}}\text{Pm}$ production is due to resonance neutrons. Thermal flux depression would be undesirable if ^{146}Pm burnout were to occur primarily from thermal or near-thermal neutrons.

The irradiation capsule (Figure 2) was designed to provide information about the optimum target design. It consists primarily of three individual promethium capsules surrounded by an annular layer of promethium. The inner-most capsule will see only neutrons which have been filtered through promethium oxide, thus drastically depleting the resonance neutrons at promethium resonance energies, depressing the thermal flux somewhat, and increasing the effective thermal

FIGURE 2. Target Element for Promethium Irradiation

neutron temperature. This capsule represents the inner portion of a massive target element, while the annulus represents a thin layer of promethium. Burnout of ^{146}Pm could be greater in the annular portion by about a factor of two, but if the ^{146}Pm cross section is principally due to a resonance which does not coincide with a high ^{147}Pm cross section, the burnout may be similar in both the annular portion and the inner capsules.

Gamma rays in excess of 7.784 MeV, the binding energy of a neutron in ^{147}Pm , will cause a (γ, n) reaction with ^{147}Pm to produce ^{146}Pm . This reaction must be minimized if the ^{146}Pm content is to be reduced as much as possible. The principal sources of high energy photons in a reactor are fission gamma rays and the prompt gamma rays from neutron capture processes. Each nuclide will yield its own characteristic group of capture gamma rays. For many nuclides, the maximum gamma energy is less than 7.684 MeV, while for others, the energy may be over 10 MeV. Capture gamma rays from aluminum were thought to be of sufficiently low energy to be innocuous. Therefore, the massive outer body was made from high purity (1100) aluminum, and the other parts were made from a less pure but more machinable alloy (6061). Flux monitoring wires, containing 1%Co in aluminum were placed in each of two holes in the inner capsule and in two holes in the large block surrounding the annular layer of promethium. The holes were then plugged with 6061 aluminum and welded. No suitable fast flux monitoring materials was found which was free from the decided disadvantage of yielding excessively high energy gamma radiation.

Three of the target elements were loaded with 0.9 g of promethium oxide in the inner capsule, about 3 g in each of the end capsules, and 30 g in the annulus for a total of 37 g/target element. In the fourth target element; the three separate small capsules were replaced by 21 g of oxide in the inner container, surrounded by 29.4 g in the annulus. The fourth capsule was expected to provide a greater supply of high-purity promethium

but slightly less definitive information about neutron cross sections. A single neodymium capsule was fabricated similar to the fourth promethium capsule.

REACTOR IRRADIATION

The samples described above were inserted into an experimental channel in what was considered to be a well-thermalized region of one of the Hanford "K" reactors. This was done in an attempt to minimize $n,2n$ reactions caused by the fast flux contribution. All five targets, four promethium and one neodymium, were put in the reactor at the same time in accord with the schedule of irradiating the first target for about 90 days, the second for 180 days, the third for 270 days, and the neodymium and the last promethium for 360 days. This schedule was approximately maintained, and the targets were discharged and set aside to allow much of the 41.5 day ^{148m}Pm to decay away (since ^{148m}Pm gamma initially masks the ^{146}Pm and ^{147}Pm radiations).

RADIOCHEMICAL ANALYSIS OF TARGETS

The ^{146}Pm and ^{147}Pm contents in the target material were analyzed 1.35 yr after the start of irradiation. Disintegration rates of these radionuclides were determined with a calibrated Ge(Li) detector and multi-channel analyzer. The Ge(Li) detector was calibrated with the 130-keV ^{144}Ce , 430-keV ^{125}Sb , and the 730, 760-keV ^{95}Zr gammas, with absorption of the sample and 10 cm^5 counting vials as part of the calibration. The 453-keV gamma of ^{146}Pm , with a relative abundance of 0.65, and the 122-keV ^{147}Pm , with a gamma-to-beta ratio of $3.5 \pm 0.5 \times 10^{-5}$,⁽¹⁴⁾ were used to determine the $^{146}\text{Pm}/^{147}\text{Pm}$ disintegration rate ratio (3.78×10^{-7}) in the unirradiated sample. Correction back to the start of the irradiation yielded an initial disintegration rate ratio, R , of 3.13×10^{-7} , with the major uncertainty being the error in the gamma-to-beta ratio.

Each of the targets contained a substantial amount of ^{148m}Pm , even after one year of decay time, and had to be opened

in heavily shielded cells. Small samples of the "inner" and "annular" promethium were taken for analysis. At the same time, the cobalt flux wires were removed from the target elements, and the ^{60}Co contents were determined.

Because the large amount of $^{148\text{m}}\text{Pm}$ gamma activity obscured the small amount of ^{146}Pm present in the samples, the following counting scheme was followed. A weighed portion of Pm_2O_3 was dissolved in nitric acid, diluted to a known volume, and a gamma spectrum was obtained using a $\text{Ge}(\text{Li})$ detector and multi-channel analyzer. Bremsstrahlung from the ^{147}Pm beta radiation was shielded out with a lead absorber. After correction for detector efficiencies and attenuation by the lead absorber, the $^{148\text{m}}\text{Pm}$ decay rate was calculated using the 0.630 MeV gamma counting rate and relative gamma abundance of 0.87.⁽¹⁵⁾ The ^{147}Pm disintegration rate was determined with a proportional counter, previously calibrated against a 4π proportional counter. A $^{148\text{m}}\text{Pm}/^{147}\text{Pm}$ disintegration rate ratio was calculated from these data. The error in this ratio is estimated to be about 10%.

After determination of the $^{148\text{m}}\text{Pm}/^{147}\text{Pm}$ ratio, a sample was prepared for counting in a high-sensitivity anti-coincidence $\text{Ge}(\text{Li})$ gamma ray spectrometer,⁽¹⁶⁾ so that the ^{146}Pm could be detected in the presence of the large $^{148\text{m}}\text{Pm}$ background. This system comprised a 20-cm³ coaxial diode centered inside a 26-in. diameter by 24-in. thick plastic phosphor. The ^{146}Pm , ^{147}Pm and $^{148\text{m}}\text{Pm}$ sample was placed inside the shield, and the coincidence and anti-coincidence spectra were recorded. The anti-coincidence shield reduced the Compton edge, in the vicinity of 700 keV by a factor of 50 to provide a peak-to-Compton edge ratio of about 300 to 1 without significant loss of photopeak efficiency. This counting system enabled the detection of one count of ^{146}Pm in greater than 10^3 of $^{148\text{m}}\text{Pm}$. Counting periods of 24 to 48 hr were required to accumulate about 300 counts of

^{146}Pm , while about 300,000 counts of $^{148\text{m}}\text{Pm}$ were recorded. Figure 3 shows typical results.

In order to minimize differences and uncertainties in Ge(Li) detector efficiencies, the 745-keV gamma of ^{146}Pm and the 727-keV gamma of $^{148\text{m}}\text{Pm}$ were also counted. The 727-keV counts were taken as the sum of those in the coincidence and anti-coincidence modes, and the $^{148\text{m}}\text{Pm}$ disintegration rate was obtained by dividing the observed 727-keV counts by 0.36, the fraction of decays passing through the 1908 keV level of ^{148}Sm . This value was taken from C. M. Lederer, J. M. Hollander, and I. Perlman.⁽¹⁵⁾ Similarly, the ^{146}Pm disintegration rate was obtained by dividing the 745-keV gamma counting rate, observed in the anti-coincidence mode, by 0.32.⁽¹⁵⁾ A $^{146}\text{Pm}/^{148\text{m}}\text{Pm}$ disintegration rate ratio was calculated from the information above.

The $^{148\text{m}}\text{Pm}/^{147}\text{Pm}$ and $^{146}\text{Pm}/^{148\text{m}}\text{Pm}$ ratios were then used to calculate the $^{146}\text{Pm}/^{147}\text{Pm}$ disintegration rate ratio at reactor discharge after suitable decay corrections were made for ^{146}Pm , ^{147}Pm and $^{148\text{m}}\text{Pm}$. Half-lives of 5.53,⁽⁴⁾ and 2.62⁽⁵⁾ yr and 41.5⁽¹⁷⁾ days were used respectively.

A more direct but less precise determination of the $^{146}\text{Pm}/^{147}\text{Pm}$ ratio was done in the following way. After making a 56% correction for absorption of the 122-keV gamma by the 20 mils of cadmium surrounding the sample and accounting for the differences in detector efficiency (a factor of 7.5) between the 122-keV and 745-keV gammas, the ^{147}Pm disintegration ratio was computed from the 122-keV anti-coincidence count by use of the observed gamma-to-beta ratio (3.5×10^{-5}) for ^{147}Pm .⁽¹⁴⁾ This ^{147}Pm disintegration rate was used along with the previously determined ^{146}Pm disintegration rate (also found from the anti-coincidence data on the same tape) to obtain a $^{146}\text{Pm}/^{147}\text{Pm}$ ratio. Due to uncertainties in the

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FIGURE 3. Gamma-Ray Spectrum of ^{148m}Pm

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absorption correction, counter efficiencies, and the gamma-to-beta ratio, these values can only be considered as a check of the results; however, the agreement is surprisingly good.

In one case, the $^{146}\text{Pm}/^{147}\text{Pm}$ ratio was determined using the 453-keV ^{146}Pm gamma counting rate from the anti-coincidence mode. The ^{146}Pm disintegration rate was found by dividing the 453-keV counting rate by 0.40,⁽¹⁸⁾ the fraction of decays which go directly to this level because those gammas in coincidence with the 1190 keV level are cancelled out by the anti-coincidence system. These data were corrected for detector efficiency and used with the 727-keV $^{148\text{m}}\text{Pm}$ disintegration rate to obtain a $^{146}\text{Pm}/^{148\text{m}}\text{Pm}$ ratio. A $^{146}\text{Pm}/^{147}\text{Pm}$ ratio was obtained from the above ratio and the $^{148\text{m}}\text{Pm}/^{147}\text{Pm}$ ratio. The $^{146}\text{Pm}/^{147}\text{Pm}$ ratios, obtained from the counting data along with these calculations, are shown in Table 3.

TABLE 3. $^{146}\text{Pm}/^{147}\text{Pm}$ Disintegration Rate Ratios

Target No.	Net Irradiation Time (days)	Ratio at end of Irradiation $\times 10^7$		
		$\frac{727-745}{(\text{keV})}$	$\frac{727-453}{(\text{keV})}$	$\frac{122-745}{(\text{keV})}$
1-inner	101	2.59	---	3.05
1-annular				
2-inner	181	2.29	---	---
2-annular		2.38		
3-inner	260	2.32	---	2.25
3-annular		1.90		
4-inner	356	1.82	1.53	1.93
4-annular		1.79		
Nd-inner	356	2.14	---	---
Nd-annular		1.80		

Error Analysis

The $^{146}\text{Pm}/^{147}\text{Pm}$ ratios found in Table 3 have a total estimated error of 20%. Statistical counting errors for the

Ge(Li) diodes range from 0.2% for ^{148m}Pm to about 6% for ^{146}Pm , and the ^{147}Pm counting error is estimated to be 3%. Decay corrections, due to differences when samples were counted, are probably less than 5% and are due primarily to an uncertainty in the ^{148m}Pm half-life. Decay corrections of about 1 yr for ^{146}Pm and ^{147}Pm also contribute an estimated error of 1 or 2%. Use of relative gamma abundances introduces the largest error, estimated to be about 20% and comes from an uncertainty of about 7% per transition. Four transitions were used in these calculations.

The method of calculating the $^{146}\text{Pm}/^{147}\text{Pm}$ ratio is reviewed below. The disintegration rate ratio, R, is

$$R = R(146/148m)/R(147/148m) \\ = (C_{146}/C_{148m}) \times (b_2/b_1) / [(C_{147}/C_{148m}) \times (b_3)],$$

where C_{146} , C_{147} and C_{148m} are the observed counting rates, and b_1 , b_2 , and b_3 are the gamma abundances for the 727-keV ^{146}Pm , 745-keV ^{148m}Pm and 630-keV ^{148m}Pm gammas, respectively. If and when "better" gamma abundances become available, the equation above can be used to calculate new $^{146}\text{Pm}/^{147}\text{Pm}$ ratios.

ANALYSIS OF THE DATA

The number of atoms of ^{146}Pm formed and destroyed during the irradiation is given by the differential equation

$$dN_{46}/dt = \sigma_{2n} \phi N_{47} - \Lambda_{47} N_{46}.$$

Integrating and converting to an activity basis yields

$$R = \frac{A_{46}}{A_{47}} = \frac{\sigma_{2n} \phi}{\Lambda_{46} - \Lambda_{47}} \left(\frac{\lambda_{46}}{\lambda_{47}} \right) [1 - \exp - (\Lambda_{46} - \Lambda_{47}) t] \\ + \frac{A_{46}^0}{A_{47}^0} \exp - (\Lambda_{46} - \Lambda_{47}) t.$$

Since the quantity $(\sigma_{2n} \phi / \Lambda_{46} - \Lambda_{47}) (\lambda_{46} / \lambda_{47})$ is the equilibrium $^{146}\text{Pm}/^{147}\text{Pm}$ ratio, R_{∞} , the equation can be further modified to have the form

$$(R - R_{\infty}) / (R_0 - R_{\infty}) = \exp - (\Lambda_{46} - \Lambda_{47}) t,$$

where R_0 , R , R_{∞} , are the $^{146}\text{Pm}/^{147}\text{Pm}$ ratios at the beginning of the irradiation, at reactor discharge, and at equilibrium, respectively. $\Lambda_{46} = \sigma_{46}\phi + \lambda_{46}$, and $\Lambda_{47} = \sigma_{47}\phi + \lambda_{47}$. These symbols have their usual significance.

Analysis of the cobalt flux wires yielded flux values of 1.44×10^{13} n/cm²-sec and 2.14×10^{13} n/cm²-sec for the inner and annular wires, respectively. Absorption by promethium in the outer annulus causes the difference between the flux at the outer and inner wires. From this flux difference, the macroscopic cross section of the promethium oxide powder was evaluated. Values of the average flux were calculated for both the inner and outer promethium regions and corrected for attenuation within the layer and spatial distribution of the promethium. These average flux values, 1.32×10^{13} and 1.79×10^{13} , were used in analyzing the data from the irradiations.

Examination of the promethium and neodymium irradiations data in Table 3 indicates that the equilibrium concentration of ^{146}Pm in these targets was reached from both directions for the longest irradiations. An average equilibrium ratio (R_{∞}) of 1.80×10^{-7} , the net irradiation times the neutron flux, the initial ratio (R_0) of 3.13×10^{-7} , and the observed ratio (R) were used to calculate the ^{146}Pm cross sections. variances for each cross section were calculated with consideration for counting errors, half-life uncertainties, uncertainty in the initial ratio and uncertainty in irradiation times. The cross section was weighted inversely with its variance to obtain a weighted estimate of the cross section in both the inner and annular promethium. The weighted average cross section was

then used to determine the fraction of saturation at the longest irradiation time, from which a new estimate of the equilibrium ratio was determined. The process was repeated until the change in the equilibrium ratio was less than one percent. The final estimates for the equilibrium ratios were 1.51×10^{-7} and 1.46×10^{-7} for the inner and annular promethium, respectively. Cross sections calculated from these ratios are shown in Table 4, along with the standard deviation for each cross section estimate. The weighted average cross sections are 3670 ± 450 b and 3160 ± 430 b for the inner and annular promethium, respectively, excluding the decay scheme uncertainty.

TABLE 4. Cross Sections as Calculated from Final Estimates of Equilibrium Ratios

Target No.	Irradi- ation Time (days)	$\frac{R - R_{\infty}}{R_0 - R_{\infty}}$	$(\Lambda_{46} - \Lambda_{47})$ $\times 10^8 - \text{sec}^{-1}$	σ_6 (b)
1-Inner	101	0.667	4.65	4110 ± 1480
1-Annular		0.674	4.52	3030 ± 1260
2-Inner	181	0.482	4.67	4130 ± 760
2-Annular		0.548	3.85	2660 ± 660
3-Inner	260	0.500	3.08	2930 ± 670
3-Annular		0.258	6.03	3870 ± 930
4-Inner	356	0.191	5.38	4670 ± 1270
4-Annular		0.192	5.37	3500 ± 910

An alternate approach was also used in analyzing the data. The initial ratio used above was determined by a different technique, so that some decay scheme uncertainties enter into the analysis of variance. This source of uncertainty can be eliminated by using the values at the end of the first irradiation as the initial time in analyzing subsequent irradiation times. The same process of calculating a cross section and using it to obtain a revised equilibrium ratio was used in this analysis of the data. Final estimates were 1.55×10^{-7} and

1.57×10^{-7} for the equilibrium ratios, and 3350 ± 1900 b and 4030 ± 1600 b, respectively, for the annular and inner promethium.

The best estimate of cross sections for ^{146}Pm are 3220 ± 420 b for the annular promethium and 3650 ± 440 b for the inner promethium, based on both methods of analyzing the data.

CONCLUSIONS

The difference between our cross section and that found by Pinajian⁽¹⁰⁾ may be accounted for by differences in neutron flux distributions in the samples and the reactors used. This difference indicates that much of the ^{146}Pm burnout is due to resonance capture of epithermal neutrons. This relationship is suggested by the differences in the cross section for the inner and annular promethium. The thermal flux is lower in the inner promethium region, while the epithermal flux is unchanged except at those energies at which ^{147}Pm absorbs neutrons. The reaction rate of ^{146}Pm (product of flux and cross section) is more nearly constant than would be expected from the thermal flux differences. This shows that the flux at energies responsible for ^{146}Pm reactions is depleted less than the thermal flux. By analogy to $^{148\text{m}}\text{Pm}$, which is also an odd-odd nuclide, the cross section is expected to be largely due to resonance absorption at energies slightly above thermal. Such a cross section structure is entirely compatible with the observed data. If the cross section were largely thermal, the cross section would be about the same in the inner and annular regions, but the reaction rate in the inner region would be lowered in proportion to the thermal flux depression.

The higher equilibrium ratio obtained by irradiating neodymium may also be explained on the basis of cross section structure. If one or more of the neodymium isotopes have

large cross sections which coincide with the ^{146}Pm resonance energy, the flux at this energy will be depleted, thus decreasing the reaction rate with ^{146}Pm and increasing the equilibrium amount. This would result in a slight increase in the annular material, and a larger increase in the inner material where all of the neutrons are filtered through an appreciable amount of neodymium. Because neodymium has a much lower cross section than ^{147}Pm , the thermal flux in the annular and inner neodymium samples is not greatly different.

Another explanation of the difference in the neodymium samples could be production of gamma rays of sufficiently high energy to cause γ, n reactions. Energetic gamma rays would also increase the equilibrium amount of ^{146}Pm by increasing the rate of formation, while leaving the rate of decay unchanged. One neodymium isotope, ^{143}Nd , releases 7.82 MeV by neutron capture. However, the maximum gamma ray energy reported in its decay is 7.11 MeV. A small amount of higher energy gammas is possible, and could account for the observed high ^{146}Pm content.

The reduction in ^{146}Pm content is much less than anticipated. Part of the ^{146}Pm content could be explained on the basis of high energy gamma rays from the massive amount of aluminum surrounding the sample. When the experiment was designed, the best estimate of the binding energy of a neutron in ^{147}Pm was 7.83 MeV, but later data show a value of 7.684 MeV to be more reasonable. A major branch of capture gammas from aluminum is at 7.72 MeV, which could cause γ, n reactions according to the revised threshold value.

Another partial explanation of the high equilibrium ratio is that the cross section for ^{146}Pm is lower than was assumed in calculating the effect of irradiation. A decrease in cross section by a factor of two would cause a factor of two increase in equilibrium ratio. Two additional explanations can be given.

The first is that the choice of target element position was not a good one and that the "fast" flux was high enough to make the contribution of the $^{147}\text{Pm}(n,2n)^{146}\text{Pm}$ reaction just slightly less than the $^{146}\text{Pm}(n,\gamma)^{147}\text{Pm}$ reaction. The second is that the cross section for the $^{147}\text{Pm}(n,2n)^{146}\text{Pm}$ reaction is larger than anticipated. Our data do not make it possible to distinguish among the explanations given above.

The flux of 7.72-MeV gamma rays from neutron capture in aluminum was calculated for the promethium layers. Input parameters were 0.23 b for neutron capture cross section of aluminum, 7.72 MeV gamma in 35% of the captures, and 3in. O.D. and 0.5 in. I.D. for the aluminum cyclindrical annulus. The flux of 7.72-MeV gammas in the promethium was calculated to be 0.024 times the thermal neutron flux. The photoneutron cross section for ^{147}Pm at 7.72-MeV cannot be estimated with good accuracy, but data for other rare earths indicate that a cross section of 20 mb is not unreasonable. As measured just outside the annular promethium, the flux in the aluminum is at least 2.14×10^{13} n/cm²-sec and probably somewhat higher in the outer portions of the aluminum container. These values indicate that the contribution from aluminum gammas is equivalent to a fast flux of about 0.1 times the average thermal flux in the annular promethium. From Figure 1, the observed ratio is 0.3, based on a ^{146}Pm capture cross section of 8400 b.(10) Correcting for the observed ^{146}Pm cross section yields a value of 0.11, which is in good agreement with the expected contributions from fast neutrons and aluminum gammas.

Burnout of ^{146}Pm to a value of about 0.16 ppm (activity basis) has been demonstrated to be possible in the Hanford reactors. By such an irradiation, power-reactor promethium could be converted to material of even better quality than as-produced production-reactor promethium. The possibility of further improvement has not been demonstrated, but appears

likely since γ, n reactions probably contributed to production of ^{146}Pm , and this source of ^{146}Pm can be reduced.

Further reduction of the ^{146}Pm content in ^{147}Pm is expected by the use of graphite cladding to reduce the γ, n reaction on ^{147}Pm . This might make it possible to achieve an equilibrium ^{146}Pm value of about 0.08 ppm, instead of the 0.15 ppm demonstrated with aluminum cladding, and thus reduce the gamma dose rate from promethium sources by an additional factor of two. Power reactor promethium can be improved by a factor of about 30 by reirradiation in aluminum, while use of graphite cladding is expected to yield a factor of 50 reduction in ^{146}Pm content. Additional irradiations would be required to evaluate the equilibrium ^{146}Pm content more accurately.

Even with the 4- to 5-ppm ^{146}Pm expected from power reactors, promethium is competitive with ^{238}Pu for many applications. Promethium from production reactors contains substantially less ^{146}Pm , about 0.25 ppm, and is competitive with ^{238}Pu for even more applications. Either type of promethium can be improved to less than 0.16 ppm by reirradiation, but the ultimate purity obtainable is somewhat uncertain. Since the reirradiated promethium will be highly competitive with ^{238}Pu , even at 0.16 ppm, an exact definition of the ultimate improvement achievable might not be necessary.

Circulatory assist devices are expected to use sources of about 30 watts with light shielding to minimize weight. Promethium is one candidate for this application, even though it is more attractive in larger sources and where somewhat thicker shielding can be tolerated. The expected dose rate from ^{238}Pu in circulatory assist devices is equivalent to about 0.03-ppm ^{146}Pm in ^{147}Pm . A reduction to 0.16 ppm by reirradiation would make the dose rate from promethium only five times that from ^{238}Pu , while further reduction in the ^{146}Pm would bring the dose rates into even closer agreement. Promethium

is thus expected to yield only a slightly higher does rate than ^{238}Pu , even in this application. The desirability of additional research on irradiation of ^{147}Pm depends upon the need for more accurate data as discussed above.

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