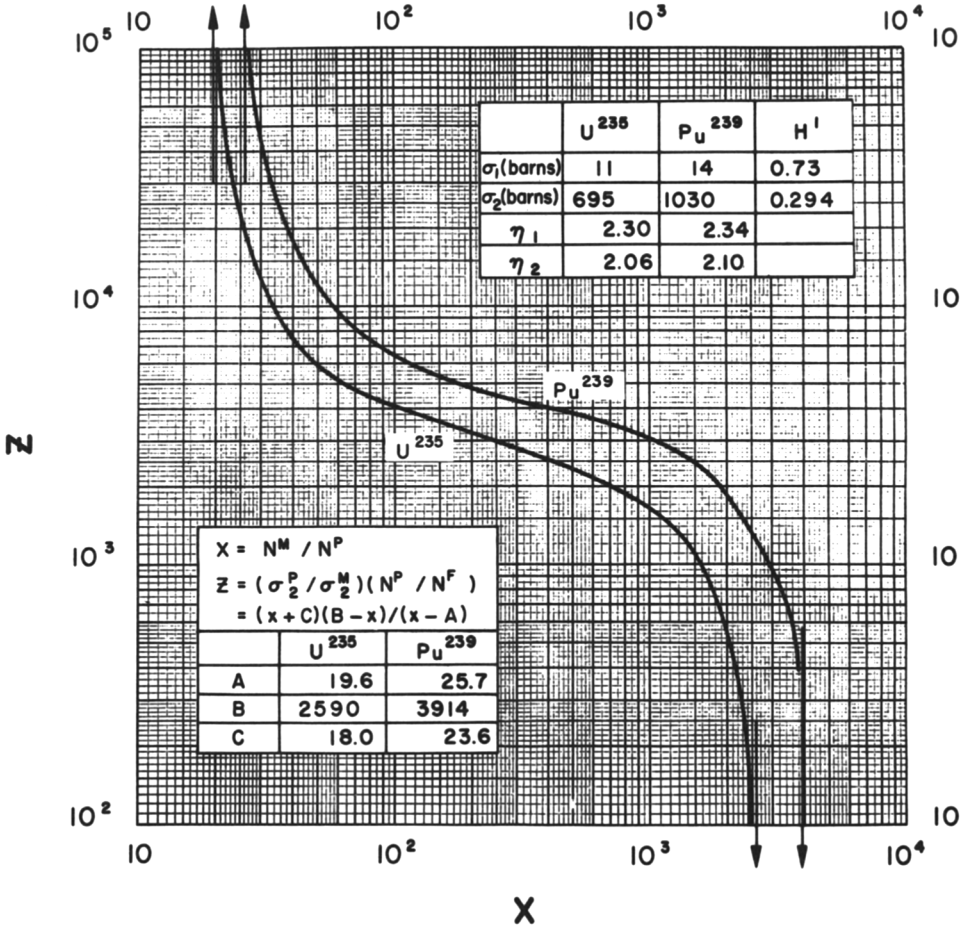

	U²³⁵	Pu²³⁹	H¹
σ_1(barns)	11	14	0.73
σ_2(barns)	695	1030	0.294
η_1	2.30	2.34	
η_2	2.06	2.10	

	U²³⁵	Pu²³⁹
A	19.6	25.7
B	2590	3914
C	18.0	23.6

Fig. 1.

hence

$$\frac{N^P}{N^F} = \frac{4.80}{2.57} = 1.87.$$

Similar curves can be constructed for other fissionable materials.

The parameters used in obtaining the curves of Fig. 1 are given in Table I.

The thermal cross sections are taken from ANL 5800 Table 2.1. The fast cross sections were obtained from the Reactor Physics Branch of Phillips. These are considered reasonable but conservative values for the intended purpose.

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Thermal Power of Promethium-147*

INTRODUCTION

More extensive use of large radioactive sources to produce heat (in thermoelectric generators for satellites and automatic weather stations, for example) point up the need for accurate knowledge of the rate of heat production of such materials.

Promethium-147, which is readily available from uranium fission products and has a convenient half-life of 2.67 years[1], is one of the most attractive isotopes for use in radioactive heat

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*Research sponsored by the USAEC under contract with the Union Carbide Corporation.

[1] F. P. ROBERTS, E. J. WHEELWRIGHT and W. Y. MATSUMOTO, "The Half-Life of Promethium-147," USAEC Rpt. HW-77296, General Electric, Hanford Atomic Products Operation (April 1963).

sources. Furthermore, since Pm^{147} produces very little penetrating radiation, it requires comparatively little shielding.

The average beta energy (from which the thermal power can be directly calculated) was previously determined from the beta spectrum to be 0.00641 MeV and by calculations from Fermi's theory by Hovi[2] to be 0.0630 MeV. Later the value 0.0704 ± 0.0040 MeV was determined by Hovi and Niemela[3], who used a calorimetric method. This latter value compares with 0.070 ± 0.001 MeV obtained in the present study by calorimetry and 4π-beta proportional counting.

EXPERIMENTAL PROCEDURES

Materials and Analyses

The Pm^{147} used in this study was analyzed by gamma spectrometry and found to contain 3×10^{-7} C of Pm^{146} and 1.0×10^{-7} C of Pm^{148} for each curie of Pm^{147}. The decay schemes of Funk *et al.*[4], for Pm^{146} and of Eldridge and Lyon[5] for Pm^{148}, were used in this analysis. The ≈ 0.120-MeV gamma radiation from Pm^{147} was measured and found to be $< 0.06\%$ of the total Pm^{147} disintegrations. Mass spectrographic analyses showed that Sm^{151} was present in quantities of < 0.02 at.%. The energy from these radioactivities, although absorbed in the source and calorimeter, made a negligible contribution to the total energy calorimetrically measured; no other radioactive impurities were found. All radioactivity values were corrected to May 21, 1963, the date of the calorimetric measurements.

Sample Preparation and Counting

A 3.934-g sample of $Pm^{147}{}_2O_3$ was pressed into two 1.0-cm pellets and doubly encapsulated in 20-mil stainless-steel cans. The encapsulated source was 1/2 in. diam by 1 in. high.

After calorimetry the pellets were dissolved in HCl and the solution diluted to one liter in a volumetric flask. Duplicate aliquots were taken at several different times over a four-month period. The number of curies of Pm^{147} in each of the aliquots was determined by 4π-beta proportional counting. A value for the total number of curies was calculated from those obtained on each of the aliquots.

Calorimetric Determination

The calorimeter used in this work was of a type that has been used extensively by the Isotopes Development Center of ORNL[6,7]. The design of this all-copper instrument is shown in Fig. 1. The upper portion consists of a metal cup mounted on a metal shaft. At the base of the shaft is a heat sink cooled by a stream of water at constant temperature. The heat produced by radioactive decay of the source is conducted down the shaft to the heat sink. At steady state the temperature difference between the top and base of the shaft is essentially proportional to the rate at which heat is produced

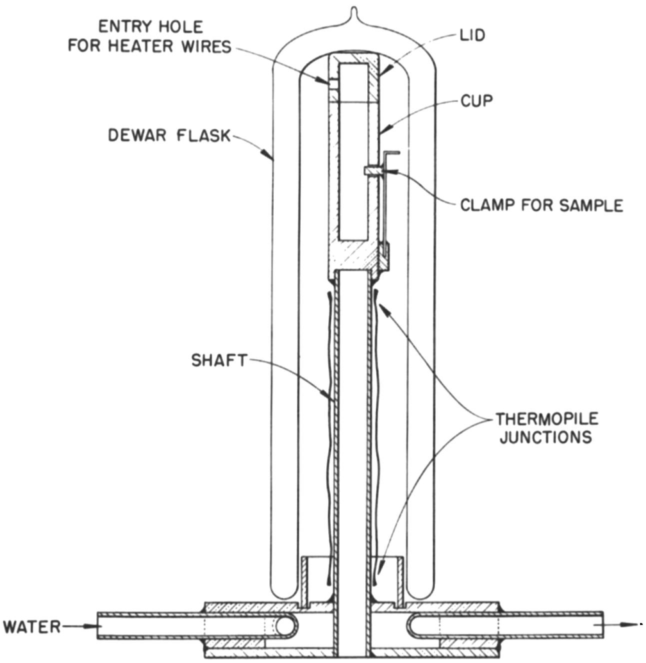

Fig. 1. Design of calorimeter. Overall height of cup, $3\frac{1}{4}$ in.; ID, $\frac{1}{2}$ in.; OD, $\frac{11}{16}$ in.; base $\frac{1}{2}$ in. thick; shaft, $\frac{1}{2}$-in. by 0.062-in. tubing of $4\frac{5}{8}$ in. effective length. This calorimeter was modified from one originally designed for assay of the thermal power of α-emitting samples in the Oak Ridge Research Reactor.

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[2] VAINO HOVI, "Radioactive β-Disintegration of Phosphorus-32, Sulfur-35, Promethium-147, Calcium-45, and Praseodymium-143," *Ann. Acad. Sci. Fennicae, Ser. A. VI*, **16**, (1959).

[3] VAINO HOVI and LASSE NIEMELA, "Measurements of the Mean Beta Disintegration Energies of ^{32}P and ^{147}Pm at Low Temperatures," *Ann. Acad. Sci. Fennicae, Ser. A. VI*, **103**, (1962).

[4] E. G. FUNK, I. W. MIHELICH and C. F. SCHWERDTFEGER, "Radioactive Decay of ^{143}Pm, ^{144}Pm, and ^{146}Pm," *Phys. Rev.* **120**, 1781-9 (1960).

[5] J. S. ELDRIDGE and W. S. LYON, "Promethium-148," *Nucl. Phys.* **23**, 1: 131-8 (1961).

[6] J. C. POSEY, T. A. BUTLER and P. S. BAKER, "Hot-Cell Calorimetry for Routine Determination of Thermal Power Generated by Kilocurie Sources," in *Proc. of the Tenth Conference on Hot Laboratories and Equipment*, (Washington, D. C., Nov. 26-28, 1962), pp. 263-68, American Nuclear Society, Chicago, (1962).

[7] J. C. POSEY, "Calorimetry for Measuring Radioactivity," *Isotopes and Radiation Technology* **1**, 1, 92-6 (1963).

by the radioactive source. Extraneous heat loss is minimized by enclosing the cup and shaft in an inverted Dewar flask.

The temperature difference between the ends of the shaft was measured by a five-couple iron-constantan thermopile with the hot junctions attached to the upper and the cold junctions to the lower extremities of the shaft. The junctions were evenly distributed about the circumference where they were held in place and electrically insulated by epoxy cement. The copper lead wires were connected at the hot junction end of the thermopile.

The calorimeter was calibrated by use of a battery-powered 50-ohm resistance heater substituted for the radioactive sample. The voltage drop across the heater and the heater current were measured by use of a voltage divider (Leeds and Northrup No. 7592) and shunt box (Leeds and Northrup No. 4385) in conjunction with a Rubicon double-range potentiometer. This potentiometer was used to measure the thermopile outputs for the calibration of the calorimeter and the radioactive source. Four calibrations were made at power levels close to those anticipated for the sample.

RESULTS AND CALCULATIONS

The results of calibration are shown in Table I. The decline of the calibration constant with increasing power levels is characteristic of calorimeters of this specific design. The calibration constants corresponding to the thermopile output observed in the calorimetric measurements were obtained by interpolation, using a linear, least mean square equation.

Two determinations of the thermal power of the sample were made on successive days, May 20 and 21, 1963 and the values, corrected to May 21, are given in Table II.

The fractions of the total energy escaping from the calorimeter as beta radiation, bremsstrahlung, gamma radiation, and promethium x-rays were negligible. The total activity of the sample, as determined by 4π-beta counting of aliquots from the dissolved oxide, is shown in Table III.

TABLE I

Calibration of Calorimeter

Heater power (watts)	Thermopile output (millivolts)	Calibration constant (W/mV)
0.7724	0.9538	0.8108
0.7789	0.9650	0.8081
1.3823	1.721	0.8040
1.4438	1.800	0.8029

TABLE II

Thermal Power of Promethium-147 Pellet

Date of determination	Power corrected to 5/21 (W)
5/20	1.0166
5/21	1.0142
Average	1.0154

The precision of the calorimetric determinations was estimated by use of previous as well as present operating data. The standard deviation of individual measurements obtained in a previous study[7] involving 18 independent measurements was 0.0023 W. Because of the larger number of measurements involved, this value was accepted as a better estimation than the values obtained in the present work (0.0013 W for the standard deviation from the calibration data and 0.0024 W from the sample measurements). Using 0.0023 W as the true standard deviation of a single value, the standard deviation of the mean was calculated to be 0.0021 W, which is equivalent to 0.21%.

The standard deviation of the mean of the total activity measurements was calculated to be ± 26 C; this is equivalent to 1.1% of the average value. No systematic error is believed to be present.

The thermal power of Pm^{147} was determined from the data of this report to be 0.414 ± 0.005 W/kC. This value corresponds to an average beta energy of 0.070 ± 0.001 MeV. The standard deviations were calculated by normal methods of combining variances. The corresponding limits of error at the 95% confidence level were 0.012 W and 0.002 MeV.

By use of the above data and a half-life of 2.67 years, a power of 0.377 W/g of Pm^{147} was calcu-

TABLE III

Total Radioactivity of Sample

Date of determination	Activity (Curies)	Activity corrected to 5/21 (C)
8/13	2399	2546
8/13	2348	2492
8/27	2223	2385
8/27	2223	2385
9/13	2264	2458
9/13	2264	2458
Average		2454

lated. The average beta energy determined here agrees well with the value of 0.0704 ± 0.0040 MeV which Hovi and Niemela[3] determined by a calorimetric method.

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The Infinite Dilution Resonance Integral of Thorium-232

Reported values of the infinite dilution resonance integral of Th^{232} have covered an unusually wide range. Measurements have ranged from 67 to 106 barns, while values calculated from resonance parameters have varied from 76 to 97 barns. Most recently, Brose[1] has reported an experimental value of 82.7 ± 1.8 barns.

In connection with a study of U^{238}/Th^{232} mixtures[2], we have also measured and calculated the thorium infinite dilution resonance integral. The results, which afford an interesting comparison with previous work, are:

Measured: 81.2 ± 3.4 barns
Calculated: 82.3 barns.

The agreement with Brose's value is very good, especially in light of the fact that the experimental methods were different. Brose used an activation technique, while we used a static reactivity technique.

Measurements were made in the Advanced Reactivity Measurement Facility (ARMF-II) at the National Reactor Testing Station. The samples, containing a wide range of ThO_2 concentrations, were prepared by compacting powders in identical aluminum cylinders approximately one inch in diam and five inches long. To control absorber density in the different samples, lead dioxide powder was used as a diluent. The sample of lowest absorber density contained approximately 0.25 wt% thorium dioxide. Absorber particle size was sufficiently small to make particle self-shielding negligible.

Calibration was effected by performing both activation and reactivity measurements on gold samples; the standard used was a value of 1579 barns for the gold infinite dilution integral from a low-energy cutoff of 0.5 eV. This value was calculated from the recent Columbia data[3]. Experimentally based corrections to the observed reactivity data were made to account for the effects of the aluminum capsules, scattering by the absorber and the diluent, and absorption in the diluent. Theoretical corrections were applied to correct for the effect of the 20-mil cadmium filter, slight deviation of the ARMF flux from a $1/E$ spectrum throughout the resonance region, energy dependence of the adjoint flux, and the reactivity effects of fast fissions. The latter three corrections were based on a transport calculation of the real and adjoint fluxes in the cadmium-shielded measurement position.

The infinite dilution resonance integral was determined by extrapolating an analytical fit of the reactivity worths to yield a value of reactivity for each absorber atom at zero absorber concentration in the sample; this quantity is proportional to the infinite dilution absorption integral. An expression of the form

$$\rho_a = A(1 - e^{-BN})$$

where

ρ_a = reactivity due to absorption
N = absorber atom density
A and B = constants determined by the fit

gave an excellent fit to the data. The resulting infinite dilution resonance integral given above is adjusted to a cutoff energy of 0.5 eV and includes the $1/v$ contribution. One barn of the 3.4 barn uncertainty is due to a 3% uncertainty assigned to the gold infinite dilution value.

The calculated value given above is based on a 'best' set of resonance parameters compiled recently by the Cross Section Evaluation Center at Brookhaven National Laboratory[4]. The calculation

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