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Calorimetric Determination of the Half-Life of Polonium-210

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Six determinations have been made of the half-life of polonium with four different steady-state, resistance-bridge calorimeters and five different samples of polonium. These six values of the half-life have been weighted and combined to give a grand-mean value of the half-life of 138.4005±0.0051 days (internal consistency) or ±0.0058 day (external consistency).

INTRODUCTION

MOUND Laboratory has been engaged in the development of calorimeters to be used in the measurement of radioactivity. The quality of the various calorimeter designs was judged by measurements on small localized sources of heat. Polonium-210 was used for the small sources of heat. Polonium is practically an ideal heat source for calorimetric measure because of its simple decay scheme. The polonium isotope decays to stable lead by the emission of essentially monoenergetic alpha particles and of one gamma ray per 10^5 alphas.[1]

During the course of our work, a large number of measurements were made with several samples of different over-all dimensions and with different amounts of polonium. The testing of a calorimeter of new design extended over a period of two to four months, and even in this length of time fairly precise values of the half-life of the samples were calculated from the accumulated data. It was decided to continue to obtain data over a longer period of time so that more precise values of the half-life would be obtained. The precisions of these determinations were used as one of the measures of the success of the calorimeter designs. Since our data and results were of unusually high precision, we believed it desirable to publish the information. The very precise value of the half-life of polonium allowed the use of these samples as reference heat sources in calorimetry research.

Since the identification of polonium by Curie[2] as a naturally occurring radioactive element, there have been many determinations of the half-life of polonium-210. Gmelin's Handbuch[3] lists 28 determinations of the half-life. Beamer and Easton[4] in two determinations obtained a value of the half-life of polonium-210 of 138.3 days±0.1 percent. More recently, Ginnings, Ball, and Vier[5] obtained a value of 138.39±0.14 days.

CALORIMETER DESIGN

The calorimeters used in this work were developed from the steady-state, resistance-bridge type used by Rutherford and Robinson[6] in the measurement of radioactivity by the calorimetric method. The Mound Laboratory development work was directed towards obtaining a knowledge of materials of construction and of dimensional dependence sufficient to permit the design of a calorimeter with arbitrary sensitivity, precision, stability, sample volume, sample-power range, and time to reach equilibrium. In the course of the work, the "heat-distribution error" of a calorimeter was used as a measure of the calorimeter's accuracy. The heat-distribution error of a calorimeter is defined as the maximum-percent difference among the measured values of a small constant-electric-power source placed at various positions in the available sample volume. Polonium properly enclosed can serve as such a source, if its half-life is accurately known.

A description of a resistance-bridge calorimeter and the method of operation is contained in Beamer and Easton's paper.[4] The operating parameters of the calorimeters used in the half-life measurements are summarized in Table I.

DESCRIPTION OF SAMPLES

Five different polonium-210 samples were used during a period of three years. The polonium was prepared by standard methods[4,7] but was sealed in containers fashioned to fit the calorimetric-research needs. The chemical purity of these samples was not accurately known. However, the precision of the half-life measurements indicated that over the period of the measure-

TABLE I. Operating parameters of the calorimeters.

Calorimeter	Electrical sensitivity (μv/mw)	Time to reach 0.01 percent of equilibrium (min)	Sample-container inside dimensions Diameter (in.)	Height (in.)	Heat distribution error (%)	Thermal resistance (°C/w)
A	59.5	360	$2\frac{1}{32}$	$2\frac{11}{16}$	0.15	26
B	54.4	75	$1\frac{1}{16}$	3	0.05	4.3
C	438.	75	$\frac{1}{4}$	$\frac{3}{4}$	not measured	80
D	204.	75	$\frac{3}{4}$	3	0.3	8

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[2] M. Curie, Compt. rend. **126**, 1101 (1898).

[3] L. Gmelin, *Handbuch der Anorganischen Chemie, System Number 12, Polonium und Isotope* (Verlag Chemie, g. m. b. H., Berlin, 1941).

[4] W. H. Beamer and W. E. Easton, J. Chem. Phys. **17**, 1298–1300 (1949).

[5] Ginnings, Ball, and Vier, J. Research Natl. Bur. Standards **50**, No. 2, 75–79 (1953).

[6] E. Rutherford and H. Robinson, Phil. Mag. **7**, 202 (1904).

[7] Isotopics **2**, No. 4, 1 (1952); **3**, No. 2, 6–14 (1953).

Table II. Description of samples.

No.	Mount	Inner container			Outer container			Atmosphere	Initial watts	Method of deposition
		Material	Height (in.)	Diameter (in.)	Material	Height (in.)	Diameter (in.)			
I	platinum	platinum	$1\frac{3}{4}$	$\frac{1}{2}$	pyrex	$2\frac{1}{4}$	$\frac{5}{8}$	helium	0.47891 ±0.00005	electrodeposition
II	platinum	none	···	···	platinum	$\frac{7}{16}$	$\frac{1}{4}$	noncontrolled mixture	0.0165429±0.0000015	electrodeposition
III	platinum	platinum	$1\frac{3}{4}$	$\frac{1}{2}$	pyrex	$2\frac{1}{4}$	$\frac{5}{8}$	helium	0.25586 ±0.00001	volatilization
IV	platinum	platinum	$1\frac{3}{4}$	$\frac{1}{2}$	pyrex	$2\frac{1}{4}$	$\frac{5}{8}$	helium	0.28334 ±0.00002	volatilization
V	inside of hollow sphere	none	···	···	metal sphere	···	$\frac{13}{16}$	noncontrolled mixture	1.14697 ±0.00004	volatilization

ments only negligible quantities of radioactive impurities could have been present. Gamma counts and neutron counts of the samples did not indicate the presence of any radioactive impurities in measurable amounts.

A brief description of each sample, its preparation, and container is presented in Table II.

APPARATUS FOR MEASUREMENT AND CONTROL

All electrical equipment used in the measurements was of the highest accuracy and precision that was commercially obtainable. Potentials were measured with a Leeds and Northrup Wenner standardizing potentiometer whose accuracy was ±0.01 percent and whose precision was more exact. Currents were calculated from the measurement of the potential drop across Leeds and Northrup National Bureau of Standards type resistors by means of the potentiometer. The National Bureau of Standards type resistors had accuracies of ±0.01 percent and more exact precisions. The potentiometer was standardized by means of a Leeds and Northrup Eppley standard cell whose voltage was accurate to ±0.01 percent and was more precise. Bridge currents and heater currents were obtained from Willard low-discharge batteries.

The calorimeter bath contained a resistance bridge which served as the sensitive thermal element for a Leeds and Northrup type proportional-control system. The temperature of the bath was held constant to within 0.001°C during a measurement. Most of the measurements were made in a room controlled to ±0.2°F. Some of the measurements were made in a room controlled to ±1.0°F. The calorimeters did not show any drift or instability which could be attributed to variations in room temperature.

DISCUSSION OF MEASUREMENTS

The introduction of the sample into the calorimeter caused the internal temperature of the calorimeter to rise until the power loss to the bath was equal to the power supplied by the sample. The change in the potential difference across the bridge in the calorimeter was observed until equilibrium was reached. After the measurement of the equilibrium potential, the sample was removed, and current was then introduced into the calibrating heater in an amount sufficient to cause nearly the same bridge unbalance as was caused by the sample. The heater resistance was concurrently measured, and the power output which was computed for the heater was taken to be the power output of the sample after the heater power was linearly corrected for the difference in bridge unbalance between the sample and heater. After the performance of the calorimeter was well known, an alternative technique was used. A reference heater was compared to the calibrating heater. Power was then supplied by the reference heater in one side of the calorimeter at the same time that the sample was in the other side. Every 10 minutes, the bridge potential was measured, and the bridge current and heater current were readjusted if they had drifted slightly from their preselected values.

Each half-life in Table III was calculated from the respective decay constant which was determined by a least squares fit of the determinations of the power output of the sample and the times at which the equilibria were reached to the equation of logarithmic decay

$$\ln N = \ln N_0 - \lambda t, \tag{1}$$

where t = the time (days), λ = the decay constant (days^{-1}), N_0 = the sample size at time $t=0$ (atoms), and $T_{\frac{1}{2}} = \ln 2/\lambda$ = the half-life (days).

The determination of λ and N_0 was made by the standard least squares procedure which is used to fit a set of data (x_i, y_i) to a straight line,

$$y = a + bx, \tag{2}$$

where the set x_i (the times) are presumed to be accurately known, and the set y_i (the observed power outputs) are presumed to have random errors. Power

Table III. Values of the half-life of polonium-210.

Calorimeter	Sample	Time covered by measurements (days)	Number of observations	Half-life (days)	Probable error (days)
A	I	250	19	138.391	0.023
B	III	617[a]	32	138.401	0.012
B	IV	357	32	138.408	0.014
B	V	352	36	138.4059	0.0068
C	II	153	20	138.410	0.024
D	IV	105	24	138.314	0.024

[a] Only two measurements were taken after the 284th day.

output is directly proportional to the number of atoms. The appropriate equations for calculating a and b and their precisions may be derived or found in standard references on statistical treatment of data,[8] and they are tabulated here as Eqs. (3) through (9), inclusive.

The residuals d_i are

$$d_i = y_i - y(x_i) = y_i - a - bx_i. \tag{3}$$

The values of a and b are found from the equations

$$a = [(\sum x_i^2)(\sum y_i) - (\sum x_i)(\sum x_i y_i)]/D, \tag{4}$$

$$b = [n(\sum x_i y_i) - (\sum x_i)(\sum y_i)]/D, \tag{5}$$

$$D = n(\sum x_i^2) - (\sum x_i)^2, \tag{6}$$

(where n is the number of measurements for each half-life determination).

The probable errors of y_i, a, and b have been determined from equations given by Birge.[9] Birge's formulas can be derived by the application to Eqs. (4), (5), and (6) of the usual equations for the determination of the probable error of a function whose variables have known probable errors.[10] The probable error of a single power measurement $p(y_i)$ is found directly from the residuals

$$p(y_i) = 0.6745[\sum d_i^2/(n-2)]^{\frac{1}{2}}. \tag{7}$$

The probable errors, $p(a)$ of a and $p(b)$ of b, are:

$$p(a) = p(y_i)[\sum x_i^2/D]^{\frac{1}{2}}, \tag{8}$$

$$p(b) = p(y_i)[n/D]^{\frac{1}{2}}. \tag{9}$$

An examination of Eq. (1) discloses that a constant-percent error in the values of the sample power N cannot influence the value of the decay constant λ. The effect of such an error is to produce a parallel displacement of the line causing an error in the determination of $\ln N_0$ only. This situation is important to the determination of the half-life, because it means that, as long as experimental conditions are duplicated for every power determination, the half-life value is more accurate than the power determinations and should approach the precision of the power determinations.

Six values of the half-life of polonium-210 have been calculated from the data on five samples taken with four calorimeters. The results are given in Table III.

A grand-mean value of the half-life can be computed from the individual values which were determined. In this procedure, each value of the half-life X_i is given a weight inversely proportional to the square of its probable error p_i.

$$\bar{X} = \sum\left[\frac{k^2}{p_i^2}\cdot X_i\right] \Big/ \sum\left[\frac{k^2}{p_i^2}\right]. \tag{10}$$

Birge[9] has given a test for the consistency of the individual means which enter into the calculation of the grand mean. The means are considered consistent if

$$hx = m^{\frac{1}{2}}\left\{0.67\left[\frac{\sum[(X_i-\bar{X})/p_i]^2}{m-1}\right]^{\frac{1}{2}} - 1\right\} < 1.83, \tag{11}$$

where m is the number of means combined into the grand mean. The data of Table III when substituted into Eq. (11) give a value of 0.15, and therefore the means are consistent. The probable error $p(\bar{X})$ of the grand mean $\bar{X}$ is

$$p(\bar{X}) = (\sum 1/p_i^2)^{-\frac{1}{2}}. \tag{12}$$

Since the six half-lives of Table III are compatible, they have been combined by Eq. (10) to give a grand mean of the half-life $\bar{T}_{\frac{1}{2}}$ with a probable error determined from Eq. (12).

$$\bar{T}_{\frac{1}{2}} = 138.4005 \pm 0.0051 \text{ days.}$$

The probable error of the grand mean can also be calculated with the equation for combining means possessing external consistency:

$$p(\bar{X}) = 0.6745\left\{\frac{\sum[(X_i-\bar{X})/p_i]^2}{(m-1)\sum(1/p_i)^2}\right\}^{\frac{1}{2}}. \tag{13}$$

With $X = 138.4005$ and the data of Table III, Eq. (13) gives

$$\bar{T}_{\frac{1}{2}} = 138.4005 \pm 0.0058 \text{ days.}$$

The calorimeters, samples, and conditions of measurement were very similar throughout the measurements, so that there should be a high degree of internal consistency in the data. The application of Eq. (13) to these data appears rather meaningless in terms of evaluating the effects on the half-life of any constant errors of the method. Equation (12) determines precision only since it involves only probable errors and in no way includes deviations from the grand mean. Thus the quoted probable errors are primarily a measure of the precision of the method. Accuracy of the method rests both on the precision of the measurements and on the fact that the slope of a straight line is being determined rather than the absolute location of the line.

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