The Physical Properties of Polonium. III. The HalfLife of Polonium

William H. Beamer and William E. Easton

It has been suggested that the lead always present in polonium samples may exert a great effect on the structure. These samples contain from 1.0 percent to 50 percent lead. At the higher concentrations of lead the NaCl type structure is likely. The atomic number of lead and polonium are 82 and 84 respectively and so intensities of lines from Pb-containing planes and from Po-containing planes would be very nearly equal. At low lead concentrations the interpenetrating lattices of Pb and Po might be maintained with Po atoms occupying the sites not filled by Pb atoms. This explanation of the simple cubic and simple rhombohedral patterns would fit well with the observed solubility of lead in the polonium and with the fact that PbS, PbSe and PbTe all have the NaCl structure.

The Physical Properties of Polonium. III. The Half-Life of Polonium

William H. Beamer* and William E. Easton**
University of California, Los Alamos Scientific Laboratory, Los Alamos, New Mexico
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The half-life of polonium has been measured by a calorimetric method. The reproducibility and precision of the data is much greater than has been previously reported. The value is found to be 138.3±0.1 percent days.

INTRODUCTION

THE half-life of polonium has been measured many times and values in the literature vary from 126 to 148 days.[1] Counting, total ionization, and calorimetric methods have been used. Schweidler[2] measured the decrease of ionizing power of a sample of polonium mounted on a copper foil over 2200 days and found the half-life to be 136.5 days. This value was accepted for some time. M. Curie[3] found the half-life to be 140 days by electrical counting methods extending over 184 days. The polonium was deposited on quartz and covered with an aluminum foil 0.01 mm thick.

There are a number of errors inherent in the counting methods, such as coincidence corrections, change of counter characteristics, etc. Several of these tend to increase the apparent half-life, for example, incomplete separation from other radioactive substances such as radium D. However, most of the errors tend to decrease the half-life by effective decrease of the amount of polonium with time. (1) Atomic aggregates of polonium are known to be knocked off the sample by alphas leaving the material; (2) diffusion of polonium into the

support material; (3) covering of the surface of the polonium by a layer of oxide or foreign material. These effects are all known to occur.

The calorimetric method, which measures the decrease of heat evolution with time of a sealed polonium sample, is affected only by these errors: (1) radioactive impurities, (2) heat evolved by secondary chemical or nuclear reactions produced by the alphas. Microcalorimetric determinations of half-life have been made by Sanielevici[4] (138.7±0.4 percent and 139.6±1 percent) and A. Dorablaska[5] (137.6±0.4 percent) but the measurements of the heat evolved were not consistent. The value of the heat evolved by a curie of polonium can be calculated from the energy of the alphas and of the recoil lead atoms. The accepted value for the energy of the alpha-particles is 5.303 Mev as measured by Rutherford, Wynn-Williams, Lewis, and Boden.[6] The alpha-spectrum for polonium has been investigated by Chang[7] and found to contain less than 0.1 percent of alphas different in energy from the 5.303-Mev value. Polonium gives off about seven quanta of gamma-rays per million alphas;[8] this can be neglected. The energy of the recoil atoms, disregarding relativity effects, is

* Present address: Spectroscopy Laboratory, The Dow Chemical Company, Midland, Michigan.
** Present address: 1550 Stone Road, Rochester, New York.
[1] St. Meyer and E. Von Schweidler, *Radioaktivität* (B. G. Teubner, Leipzig, 1927), second edition.
[2] E. V. Schweidler, Verh. d. D. Phys. Ges. (2), **14**, 539 (1912).
[3] M. Curie, J. de phys. et rad. (6) **1**, 12 (1920).

[4] A. S. Sanielevici, J. de chimie phys. **33**, 779 (1936).
[5] A. Dorablaska, Roczniki Chem. (Poland) **11**, 475 (1931).
[6] Rutherford, Wynn-Williams, Lewis, and Boden, Proc. Roy. Soc. A139, 617 (1933).
[7] W. Y. Chang, Phys. Rev. **69**, 60 (1946).
[8] W. Bothe, Zeits. f. Physik **96**, 607 (1935).

$A(\alpha) \div A$ (recoil) or 4/206 of that of the alpha. Thus

$$-Q = \frac{(1+4/206)\, 5.303 \text{ Mev/alpha} \times 1.6020 \times 10^{-6} \text{ erg/Mev} \times 2.208 \times 10^{12} \text{ alpha/min./curie}}{10^7 \text{ erg/joule (abs.)}},$$

$-Q = 114.7$ joules (abs.)/curie/hour.

EXPERIMENTAL

Preparation and purity of samples.—In this investigation two samples were studied. The pure polonium was electroplated from a dilute nitric acid solution onto a platinum foil. This foil was placed inside a platinum capsule in a nitrogen atmosphere and then this was sealed off in a glass ampule after evacuation and filling with pure helium. By the use of a noble metal and a helium atmosphere the possibility of secondary reactions was minimized. The neutrons produced by the (α, n) reaction are an indication of the purity of the deposit. A count of the neutrons emitted by the samples was very low and showed no rise over the period of time the samples were measured, thus indicating that the samples were free of significant quantities of light elements. A count of the gammas from the samples agreed very well with the value of pure polonium thus indicating the absence of other gamma-emitters or of impurities giving off gammas in secondary reactions.

Apparatus.—The calorimeter used in these experiments was a modification of that described by Watson and Henderson[9] and Rutherford and Robinson.[10] This modification was developed by Dr. L. V. Coulter. It consists of two aluminum cylinders on each of which are wound the two opposite arms of a copper wire (No. 38 double silk) resistance bridge. The aluminum cylinders are mounted rigidly by wooden pegs in separate brass tubes which are immersed in a constant-level water bath which is held to a constant temperature at about 30°C ±0.001°C. The sample, centered in a cylindrical brass container, is placed in one of the aluminum tubes and the unbalance of the d.c. bridge is determined by a potentiometer when equilibrium is reached. At this point the heat evolved is equal to the heat leaking out to the bath and the temperature of the aluminum bobbin is constant. This heat effect is calibrated by passing a known current through a manganin heater of known resistance which replaces the sample in the sample tube. The other tube is used as a dummy and contains a dummy sample holder during the runs but stays at the constant temperature of the bath. The current which passes through the heater passes through shorted leads in the dummy to reproduce heat effects of the leads. The value of the temperature rise of the sample container and therefore the magnitude of the voltage across the bridge and the time elapsed in obtaining equilibrium depends upon the amount of heat generated by the sample and the heat leak away from the aluminum tube.

[9] Watson and Henderson, Proc. Roy. Soc. **A118**, 318 (1928).
[10] Rutherford and Robinson, Phil. Mag. **25**, 312 (1913).

Figure 1 gives the calorimeter bridge circuit. Figure 2 gives the heater circuit. A Wenner thermocouple potentiometer is used across the resistance bridge. The lowest dial of the lower of the two ranges reads in 0.1-microvolt increments. This potentiometer is also used across the standard resistance shown in Fig. 2 to set the bridge current at a constant value. The potentiometer for the energy circuit is a Leeds & Northrup Type K-1 and the current and voltage across the heater are measured to five significant figures.

A typical measurement of polonium sample for the half-life determination follows these steps:

(1) The bridge current is adjusted to its constant value and this adjustment is checked frequently during the whole series of operations.

(2) Since all the resistances of the bridge are not exactly the same, readings of the unbalance of the bridge are made with the empty sample holder in place. The bridge drop is always measured by taking readings until three or four values taken at five-minute intervals agree within 0.1 or 0.2 microvolt and no trend is observable. This reading is called the zero value of the bridge.

(3) The sample is placed in the sample holder and when equilibrium has been established the voltage across the bridge is read as before.

(4) The sample is replaced by the heater and a zero value obtained. This differs slightly from the zero without the heater.

(5) The current is passed through the heater to duplicate the voltage of the bridge obtained from the sample. The current through the heater and the IR drop across the heater are measured. Since it is often not possible to duplicate exactly the potential drop caused by the sample a straight line interpolation is made to find the value of the sample. This interpolation is taken
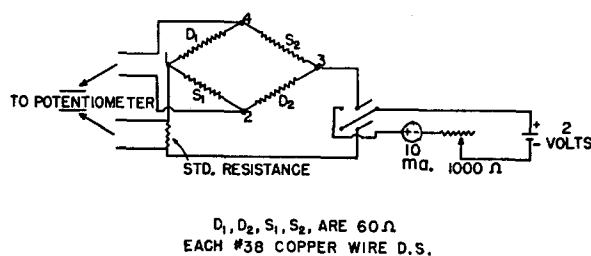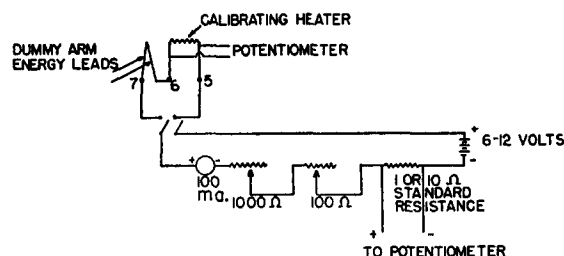

D_1, D_2, S_1, S_2, ARE 60 Ω
EACH #38 COPPER WIRE D.S.

FIG. 1. Calorimeter circuit.

FIG. 2. Energy circuit.

TABLE I. Sample 1.

Point No.	Days	Value of sample in curies
1	0	1.093
2	7.03	1.051
3	9.03	1.043
4	10.01	1.036
5	15.01	1.009
6	21.00	0.9826
7	23.01	0.9674
8	26.95	0.9526
9	28.97	0.9424
10	31.03	0.9305
11	34.08	0.9191
12	35.94	0.9091
13	38.06	0.8986
14	40.98	0.8805
15	43.01	0.8761
16	47.96	0.8576
17	49.93	0.8490
18	51.97	0.8396
19	54.97	0.8272
20	57.03	0.8186
21	58.94	0.8132
22	61.96	0.7987
23	63.96	0.7890
24	65.97	0.7846
25	68.96	0.7706
26	73.52	0.7547
27	75.95	0.7460
28	77.92	0.7361
29	83.23	0.7174
30	85.00	0.7090
31	93.12	0.6905
32	97.00	0.6648

Least squares equations: $\log y = 0.03710 - 0.0021763 \times$ half-life $= 138.32$ days.

TABLE II. Sample 2.

Point No.	Days	Value of sample in curies
1	0	1.435
2	8.87	1.371
3	9.75	1.366
4	10.88	1.356
5	13.86	1.338
6	20.76	1.291
7	23.04	1.278
8	29.82	1.238
9	31.83	1.224
10	34.84	1.202
11	36.82	1.190
12	38.81	1.179
13	41.83	1.161
14	46.01	1.140
15	48.83	1.121
16	50.74	1.115
17	55.74	1.085
18	57.65	1.072
19	64.51	1.036
20	66.55	1.027
21	71.91	1.001
22	73.49	0.9922

Least squares equation: $\log y = 0.15644 - 0.0021773 \times$ half-life $= 138.26$ days.

three thermoregulator-controlled 250-watt knife heaters. The temperature of the room is maintained constant within $\pm 1°F$ by special air-conditioning apparatus.

RESULTS

The results of the experiments are shown in Tables I and II for the two samples studied. Column 1 gives time in days; column 2 gives the value of the sample.

The half-life was obtained from the data by the least squares method. The probable error of the least squares constants was calculated by the standard procedure[11] and found to be about ± 0.05 percent for both samples.

ACKNOWLEDGMENT

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[11] A. G. Worthing and J. Geffner, *Treatment of Experimental Data* (John Wiley and Sons, Inc., New York, 1943), p. 249.

from a calibration curve for the instrument and never has to be made over a range of more than one percent of the value of the sample. The calibration curve is a straight line over the range of heat input used. For very small samples a current is passed through shorted leads in both the dummy and sample holders when the sample is being measured to duplicate any heat effects produced when the heater is in place.

The water bath is maintained at a constant temperature near 30°C to within $\pm 0.001°C$ by a mercury-in-glass thermoregulator which operates a thyratron relay circuit. The circular bath is about three feet in diameter, holds about 100 gallons of water and is stirred by a centrifugal pump which circulates 110 gallons of water a minute. The water is heated by three constant heat and