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RECOMMENDED VALUE FOR THE HALF LIFE OF POLONIUM

(Information Report)

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ABSTRACT

A critical survey has been made of the various determinations of the half life of polonium. Fifteen measurements which have been made in 1920 and subsequent years have been considered reliable enough to warrant their inclusion in a weighted mean. Each weight has been taken as proportional to the inverse square of the corresponding probable error. An arbitrary weight has been assigned to any determination for which no probable error has been found. It has been shown that the data are consistent, and accordingly they have been combined by the appropriate formulas to give the grand mean and its probable error: 138.387 ± 0.022 days

INTRODUCTION

It is the purpose of this paper to present a most probable value of the half life of polonium based on the available data. The Project Handbook[1] now lists the value of 140 days which was recommended in 1931 by a committee on radioactive standards.[2] The available data include 28 values which are recorded in Gmelin[3] and four determinations which have been made during the programs of the Manhattan District and the Atomic Energy Commission. The values are:

Author	Half Life (days)	Year
St. Meyer and E. v. Schweidler[4]	135	1904
St. Meyer and E. v. Schweidler[5]	135.5	1905
W. Marckwald[6]	139.8	1905
E. Rutherford[7]	143.	1905
W. Marckwald, H. Greinacher, and K. Herrman[8]	139.6	1905
St. Meyer and E. v. Schweidler[9]	138.2	1906
M. Curie[10]	140	1906
St. Meyer and E. v. Schweidler[11]	137	1906
E. Regener[12]	129.6	1909
J. W. Waters[13]	148	1910
E. Regener[14]	136 ± 0.5	1911
E. v. Schweidler[15]	136.5 ± 0.3	1912
R. Girard[16]	136.6	1913
R. W. Lawson[17]	127.1 136.0 136.4	1919
M. Curie[18]	140	1920
S. Maracinéanu[19]	139.5 139 - 140	1923

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S. Maracineánu[20]	134.5 - 135 138	1923
M. A. da Silva[21]	140.2	1929
A. Dorabialska[22]	137.6 ± 0.4 per cent	1931
M. Mäder[23]	138 ± 4 per cent 138.83	1932
A. S. Saniélévici[24]	138.7 ± 0.4 per cent	1935
M. Hoffer[25]	138	1935
A. S. Saniélévici[26]	139.6 ± 1 per cent	
O. E. Myers, S. S. Jones, R. Thompson, and R. A. Staniforth[27]	136.7	1946
W. H. Beamer and W. E. Easton[28]	138.32 ± 0.05 per cent 138.26 ± 0.05 per cent	1946
D. Anger and J. F. Eichelberger[29]	138.391 ± 0.023	1948

Even a casual examination of the various determinations since 1920 of the half life of polonium indicates that a value of 140 days is too large, since 13 out of the subsequent 15 determinations are lower. More specifically, the four recent calorimetric determinations, which have the advantage of considerably larger samples, give values definitely smaller than 140 days. It is apparent then that there exists sufficient data to justify the estimation of a weighted mean value and its probable error.

Since M. Curie's 1920 value generally superseded all previous estimations of the half life of polonium, only her determination and the subsequent ones will be considered in this discussion.

METHOD

The calculation of the most probable value of the half life and of its probable error has been performed by standard methods as given in Worthing and Geffner[30]. Two methods are given there for the computation of the grand mean: one for combining inconsistent means and one for combining consistent means. In either case, it is necessary to weight each datum inversely as the square of its probable error.

This requirement that the data be weighted has led to the introduction of some arbitrary numbers. Where complete data have been available,[27,28,29] the probable error of each determination has been computed by the same method. Where complete data have not been available but only a quotation of the author's estimate of the probable error, that estimate has been used.[21,22,23,25] Where no estimate has been given by the author and no data have been available, a probable error of one per cent has been assigned arbitrarily.[18,19,20,22,24] This assignment seems generous, since the sample size was small in all these indefinite cases. In addition, all the arbitrary assignments have been made to noncalorimetric determinations. There has been only one noncalorimetric determination for which the author has given a probable error,[23] and in that case a probable error of four per cent has been quoted.

Calorimetric determinations of the half life are subject to errors arising from extra heat sources or sinks such as radioactive impurities and chemical reactions. The presence of such sources or sinks would almost certainly be detected by nonlinearity in the fit to the equation

$$\log_e N = \log_e N_o - \lambda \cdot t \tag{1}$$

where:

N_o = original activity at time $t = 0$

N = activity at time t

λ = decay constant

t = time

Noncalorimetric determinations such as counting and the measurement of the ionization of a gas are subject to a variety of errors: radioactive impurities, aggregate recoil, oxidation of the surface, absorption of oblique rays, and diffusion into the mount. Since the probable errors of the microcalorimetric measurements vary between 0.4 per cent and 1.0 per cent, the 1.0 per cent probable error assigned to the presumably less accurate noncalorimetric determinations does not seem too large

Once the probable errors of the individual measurements have been assigned; the grand mean and its probable error can be determined. It is first useful to test the data for consistency This test has been made in the following fashion. Let p_i denote the probable error of the i-th determination of the half life whose value is X_i Two different determinations are mutually consistent if

$$\frac{0.67\,\left[X_i - X_j\right]}{\sqrt{2\,\left[p_i^2 + p_j^2\right]}} < 1.83 \tag{2}$$

This requirement is equivalent to the statement that there is less than one chance in a 100 that two determinations X_i and X_j with probable errors p_i and p_j will differ by more than $\left[X_i - X_j\right]$.

Another criterion of consistency has been proposed by Birge[31] In this case, it is necessary to know the grand mean, $\overline{X}$,

$$\overline{X} = \frac{\Sigma\, w_i \cdot X_i}{\Sigma\, w_i} = \frac{\Sigma \left[\frac{k^2}{p_i^2} \cdot X_i\right]}{\Sigma \left[\frac{k^2}{p_i^2}\right]} \tag{3}$$

The weights, w_i, are inversely proportional to the squares of the probable errors, and k^2 is the constant of proportionality.

$$w_i = \frac{k^2}{p_i^2} \tag{4}$$

then, as before, the means are considered consistent if

$$\sqrt{n}\cdot\left\{\frac{0\ 67\sqrt{\frac{\sum w_i\left(X_i-\bar{X}\right)^2}{n}}}{k}-1\right\}<1\ 83 \qquad (5)$$

where n equals the number of determinations which are combined into the grand mean When the individual means are consistent, the probable error of the grand mean can be computed from the equation

$$p_{\bar{X}}=\frac{1}{\sqrt{\sum\frac{1}{p_i^2}}} \qquad (6)$$

EXAMINATION OF THE DATA

The data necessary in the use of Equations 2 and 3 are presented in Table I Columns 2 and 3 contain the data as they were originally given by the authors Columns 4 and 5 contain the data as they are used in this paper Changes in Rows 9, 12, 13, and 15 will be commented on separately Column 6 contains the results of the application of Equation 2 to Columns 4 and 5 to test the consistencies of the individual means with respect to the 14th mean This test is the most severe, and indicates that the means are consistent

TABLE I

TEST OF MUTUAL CONSISTENCY

ROW	VALUE *(days)*	PROBABLE ERROR *(per cent)*	VALUE *(days)*	PROBABLE ERROR *(per cent)*	HX = CONSISTENCY	REFERENCE
1	140		140	± 1.0	0.55	18
2	139.5	–	139.5	± 1.0	0.38	19
3	139.5		139.5	± 0.7	0.54	19
4	138		138	± 1.0	0.14	20
5	140.2		140.2	± 1.0	0 62	21
6	137.6	± 0 4	137.6	± 0.4	0.69	22
7	138	± 4.0	138	± 4.0	0.03	23
8	138.83		138.83	± 1.0	0.15	23
9	138.7	± 0.4	138.2	± 1.0	0.07	24
10	138		138	± 1.0	0.14	25
11	139.6	± 1.0	139.6	± 1.0	0.41	26
12	138.32	± 0.05	138.32	± 0.30	0 08	28
13	138.26	± 0.05	138.22	± 0.19	0.31	28
14	138.391	± 0.016	138.391	± 0.016	–	29
15	136 7		137.34	± 0.28	1.32	27

GRAND MEAN $T_{\frac{1}{2}}$ = 138 387 ± 0 022 DAYS = 138.387 DAYS ± 0.016 PER CENT

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Birge s test has also been applied to the data The use of Equation 5 indicates with a single number whether all the individual means and the grand mean are consistent Birge s test is slightly simpler to use than the repeated application of Equation 2, but it possesses the disadvantage of not making immediately clear which values are inconsistent. Birge's test also indicates that the data considered here are consistent

Birge's Test for Consistency $1.30 < 1.83$

Returning to Row 9 of Table I, Saniélévici[24] gives a value of 138.7 days ± 0 4 per cent for the half life This value represents the grand mean of two separate determinations. 139.28 days ± 3.4 per cent and 138 10 days ± 1.1 per cent The application of Equations 3 and 6 to the two determinations gives a weighted mean value of 138 2 days ± 1 0 per cent rather than 138 7 days ± 0 4 per cent, Saniélévici s value Since the grand mean is evaluated by Equations 3 and 6 in this paper, it seems proper to use the present re evaluation of Saniélévici's results, 138 2 days ± 1.0 per cent Now Saniélévici's data may be introduced on the same basis as the other data and thereby preserve consistency

Rows 12 and 13 concern the results of two determinations by Beamer and Easton[28] at Los Alamos They state· "The half life was obtained from the data by the least square method The probable error was calculated by the standard procedure 11 (see our reference 30) and found to be about ± 0.05 per cent for both samples "

The procedure in Worthing and Geffner to which Beamer and Easton refer is as follows. It is required to find the value of "a" and of "b" which gives the best fit of a set of data (x_i, y_{io}), $i > 2$, to the straight line

$$y = a + b \cdot x \tag{7}$$

It is assumed that the set x_i are accurately known and that the set y_{io} have random errors If the various y_{io} have weights w_i, then

$$a = A \Sigma w_i \cdot y_{io} - A \quad \Sigma w_i \cdot x_i \cdot y_{io} \tag{8}$$

$$b = B' \Sigma w_i \cdot x_i \cdot y_{io} - B \Sigma w_i \cdot y_{io} \tag{9}$$

$$A = \frac{\Sigma w_i \cdot x_i^2}{D} \tag{10}$$

$$A = \frac{\Sigma w_i \cdot x_i}{D} \tag{11}$$

$$B' = \frac{\Sigma w_i}{D} \tag{12}$$

$$B = A' \tag{13}$$

$$D = (\Sigma w_i) \cdot (\Sigma w_i \cdot x_i^2) - (\Sigma w_i \cdot x_i)^2 \tag{14}$$

Worthing and Geffner give the probable error of a single observation y_{io} and the probable errors of "a" and "b" as·

$$p_{y_{io}} = 0\ 675 \sqrt{\frac{\Sigma \left[w_i \,(y_{io} - a - b \cdot x_i)^2\right]}{n - 2}} \tag{15}$$

$$p_a = p_{y_{io}} \Sigma w_i (A - A' \cdot x_i) \tag{16}$$

$$p_b = p_{y_{io}} \Sigma w_i (B' \cdot x_i - B) \tag{17}$$

where n is the number of observations

For the case of equal weights, which appears to be the case of Beamer and Easton, equations *16* and *17* give

$$p_a = p_{y_{io}} \tag{18}$$

$$p_b = 0 \tag{19}$$

Now "b" is analogous to λ, the decay constant, in Equation *1* and thus the formulas of Worthing and Geffner should give a probable error of zero for the half life.

It was therefore decided to apply the computational methods of Birge[31] to the data of Beamer and Easton [28]

Birge gives the following formula for finding the probable errors of "a" and "b" in Equation *7* The probable error of a single observation, y_{io}, remains the same as in Equation *15*.

$$p_a = p_{y_{io}} \cdot \sqrt{\frac{\Sigma x_i^2}{D}} \tag{20}$$

$$p_b = p_{y_{io}} \cdot \sqrt{\frac{n}{D}} \tag{21}$$

The probable errors appearing in Rows 14 and 15 of Table I have been computed by the formula of Birge so it should add consistency to the weights to have the probable errors of Beamer and Easton also computed by this method Since the weights are taken as inversely proportional to the square of the probable errors, it is important to have as much of the data as possible weighted in the same fashion.

The calorimeter used by Beamer and Easton was constructed according to a design by the Calorimetry Group at Mound Laboratory of the Monsanto Chemical Company, Dayton, Ohio, and was quite similar to the one used in Reference (27), constructed at the Monsanto Chemical Company. It was believed on the basis of studies of heat distribution errors, that the calorimeter used in Reference (29) was of the order of 10 to 15 times more accurate than the calorimeters of References (27) and (28)

It should be expected then that the probable errors of References (27) and (28) should be about the same magnitude and about 10 to 15 times larger than the probable error of reference (29). It is apparent in Table I, Rows 12, 13, 14, and 15, that applying Birge's method to the data of Beamer and Easton give probable errors of the expected order of magnitude with respect to Rows 14 and 15

Myers, Jones, Thompson, and Staniforth in Reference (27) reported a value of 136 7 days. Row 15 of Table I Since no probable error was calculated and since their paper contains the required data, it was decided to extend their calculation to give a probable error of the half life. A perusal of their paper made it apparent that certain changes should be made in their calculation.

Myers, Jones, Thompson, and Staniforth made 16 determinations of the sample size during a period of 138 days at fairly regular intervals of about 9 days. After a lapse of 109 days, they made 3 more determinations in the next 59 days. During the 109 days of no measurements, a new calibrating heater was constructed. As a result of tests with the second heater, the first 16 measurements were assigned various corrections.

It is apparent from Equation 1 that a constant percent error in N and N_o cannot affect the determination of the decay constant or, therefore, the half life. A constant percent error in N and N_o merely shifts the straight line vertically. However, there is considerable danger of getting an erroneous value of the half life by combining data from two different calibrating heaters even though using corrections. Thus, if the first 16 measurements had a constant percent error, E_1, and gave a slope, λ_1, and the last three measurements had a constant percent error, E_2, and gave a slope, λ_2, and if $E_1 \neq E_2$ and if $\lambda_1 = \lambda_2$, then the slope λ for all 19 measurements is not equal to λ_1 but varies from λ_1 by an amount depending on $(E_1 - E_2)$. It is extremely important not to make any changes in the apparatus during the course of the measurements because the error in the slope arises from varying percent errors and not from a constant percent error. Apparatus changes during the experiment that increase the accuracy of a measurement of N will decrease the accuracy of λ; and if such changes are made, the half life measurements should be started anew.

Therefore, it was decided to omit the last 3 measurements of Myers, Jones, Thompson, and Staniforth, and for the first 16 measurements to consider their original rather than their corrected data, since the nature of the correction is not such as to add accuracy to the determination of λ.

Another comment can be made on their calculations. The values of N are recorded only to three figures, all of which are truly significant. These values of N should have been computed to five figures, as the residuals are of the order of zero in the differences between three figures in the observed N's and three figures in the calculated N's.

Some comment should be made on the values entered in Row 3 of Table I. S. Maracinéanu[19] gives a value of 139 140 days for the half life. It was decided arbitrarily to enter a value of 139.5 days ± 0.7 per cent. This probably overweights the determination, but not to a serious extent. Another determination of S. Maracinéanu[20] of 134.5 to 135 days has been neglected. In this determination, Maracinéanu used a sample of polonium deposited on lead. It is known that polonium diffuses rapidly into lead and that the diffusion would lead to a low determination of the half life.

CONCLUSION

Equations 3 and 6 have been applied to Columns 4 and 5 of Table I to obtain a grand mean and its probable error for the half life of polonium

$$T_{\frac{1}{2}} = 138.387 \pm 0.022 \text{ days}$$

In terms of per cent, the probable error takes the value 0.016.

It is felt that a further study is warranted if we are successful in constructing a more accurate calorimeter. It would also be desirable to make a more comprehensive study of the papers that are quoted herein to enable a more accurate assignment of weights. In addition, some of the measurements made prior to 1920 might be included.

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