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A new determination of the ^{209}Po half-life

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Abstract

A substantial 25% error in the then-known and accepted (102 ± 5) year half-life of ^{209}Po was reported on in 2007. This error was detected from decay data from two separate primary standardizations of a ^{209}Po solution standard, which were performed approximately 12 years apart. Despite author claims that this observation was not a new half-life determination, it was nevertheless included in subsequent nuclear data evaluations and compilations to obtain a currently tabulated value of (115 ± 13) a, computed from the median and range of the two half-life reports. A third primary standardization on the identical ^{209}Po solution has since been performed to derive a new half-life value of (125.2 ± 3.3) a. This half-life determination was obtained from 30 distinct data sets over a period of 20.7 years, encompassing over 700 liquid scintillation measurements with nearly 50 counting sources all prepared from the same solution, and as obtained over a very broad range of measurement conditions (composition of cocktails, characteristics of counters, time sequencing) during five periods in 1993, 1994, 2005, and 2013.

Keywords: 209-polonium, half-life, radioactivity, decay, liquid scintillation, measurement

1. Introduction and background

The nuclide ^{209}Po is the longest-lived isotope of polonium, and is widely used as a principal radiochemical tracer by the worldwide environmental measurement community [1]. Although knowledge of this radionuclide's half-life is critically important, it has become apparent over the past half-decade that its value is highly doubtful.

Prior to 2005, the only known measurement of the half-life of ^{209}Po was reported on by André *et al* [2] in 1956 as part of a much larger study of proton reactions on bismuth. This half-life determination was made relative to that of ^{208}Po on an irradiated source that contained both radionuclides. It is difficult to evaluate or critique the quality of this result because

of the absence of sufficient information on the experimental details of the measurements. Both the $^{208}\text{Po}/^{209}\text{Po}$ activity ratio and mass ratio were said to be obtained from ‘private communications’, with stated relative uncertainties of 5% and 1.14%, respectively. The value reported by André *et al* [2] was 103 a, obtained relative to an assumed ^{208}Po half-life of (2.93 ± 0.02) a. Correcting for the presently accepted value of (2.898 ± 0.002) a [3], the ^{209}Po half-life was taken to be (102 ± 5) a. This was the sole value given in all nuclear data evaluations and compilations up to 2009 (see Firestone *et al* [4]).

In 2005, Collé *et al* [5] uncovered a substantial 25% error in the reported ^{209}Po half-life, as established by two sets of measurements made approximately 12 years apart. The first set in 1993–94 was made for the standardization of the ^{209}Po massic alpha emission rate for Standard Reference Material SRM 4326 [6]. The National Institute of Standards and Technology (NIST), the national measurements laboratory of the USA, disseminated this radio-activity standard from 1995 through 2007. Details on the preparation and standardization of SRM 4326 were reported on *in extenso* by Collé *et al* [7]. The second set in 2005 consisted of a re-standardization of SRM 4326. This work [5] led to the discovery of the ^{209}Po half-life discrepancy. The two primary standardizations revealed an apparent two-point half-life of 128.3 a, with an estimated relative uncertainty of $\pm 5.5\%$ (or ± 7 a). Although the total decay difference in the two determinations was relatively small (about 6.5% over the 12 years), it translated into a large difference in half-life. The discrepancy was therefore surprising, but was nevertheless convincing. Solution instability was discounted because of previous polonium solution stability investigations [8]. The observed half-life was also longer, suggesting that the ^{209}Po was in excess (and not a loss) compared to a predicted decayed amount. Furthermore, the measurements were sufficiently precise, with minimal probability of inaccuracy due to between-determination bias, to provide confidence in the finding. The authors, however, were insistent that this did not represent a new half-life determination given that it was only based on two points. It was apparent, however, that the 1956 André result could not withstand critical scrutiny.

Despite the above assertion and recognition that the result was not a direct determination, recent data compilers nevertheless chose to incorporate the value in an evaluation of the ^{209}Po half-life. Chisté and Bé [9] quoted a personal communication by Collé: ‘... *the value 128 a was not a new determination. The whole point was to show that there was evidence to suggest and support that the extant 102 a value is very wrong, perhaps by 25%*’ [9]. In deciding ‘*to take into account all scarce information available*’ [9], they took the expedient of adopting ‘*the simple mean of the two existing values ... with an uncertainty covering them*’; viz., (115 ± 13) a [9]. One must wonder why the data evaluators didn’t also include the lower uncertainty bound for the 102 a value and an upper bound on the 128 a value in their uncertainty estimate!

This paper reports on a third set of ^{209}Po data for the massic alpha emission rate of SRM 4326, obtained after another eight years, that confirms the earlier 2005 findings of Collé *et al* [5].

2. Methodology and experimental results

It is important to recognize that although all of the determinations from 1993 through 2013 were largely independent primary standardizations on the same ^{209}Po solution standard, the measurement method and data analyses used the identical protocols in all cases. The standardizations were based on $4\pi\alpha$ liquid scintillation counting, with corrections for detection of the low-energy conversion electrons from the 2 keV isomeric transition in ^{205}Pb and for

Table 1. Tabulated summary of the LS measurement conditions for the five determinations of the massic alpha emission rate for ^{209}Po solution standard SRM 4326.

	1993 (a)	1993 (b)	1994	2005	2013
Number of different cocktail compositions used	4 Quench varied	1 Quench varied	2 Quench varied	2 Quench varied	2 (Two quench conditions for each)
Number of sources per composition	3	6	4	5	3
Scintillation fluids used (see note 1)	RS	RS	RS & UGAB	RS	UGAB
LS counters used (see note 2)	B & P	B & P	B & P	P & B2	B2 & W
Number of measurements of each source in each counter	10	5 in B 10 in P	5 to 18	5	3
Duration of measurements (days)	2.98	25.37	25.66	5.3	21.86
Total number of measurements	240	90	206	100	72
Number of data sets in determination	8	4	4	4	8

(1) RS: Ready Safe, Beckman Instruments, Fullerton, CA, USA.

UGAB: UltimaGold AB, Packard Instrument Co., Meriden, CT, USA.

(2) B: Beckman LS7800 model LS Counter.

P: Packard Tri-Carb 2500TR LS Analyzer.

B2: Beckman–Coulter LS6500 Multi-Purpose Scintillation Counter.

W: Wallac Guardian 1414 Liquid Scintillation Counter 2.

radiations accompanying the small 0.45% electron-capture branch to ^{209}Bi [9]. This methodology was developed specifically to determine the massic alpha emission rate of ^{209}Po for the standardization of SRM 4326 [6, 7]. For consistency, all subsequent results from 2005 and 2013 were obtained using largely the same measurement conditions, identical spectral analysis procedures, and applied corrections. Table 1 provides a summary of the various measurement conditions used for the various determinations.

The standard SRM 4326 that was used for all measurements consisted of a very pure, carrier-free solution of ^{209}Po in nominal 2 mol L^{-1} HCl, having a solution density of $(1.031 \pm 0.004)\text{ g mL}^{-1}$ at $22\text{ }^{\circ}\text{C}$. The certified ^{209}Po massic alpha-particle emission rate was $(85.42 \pm 0.36)\text{ s}^{-1}\text{ g}^{-1}$ as of the Reference Time of 1200 EST 15 March 1994 [6, 7]. The stock material was originally obtained from Oak Ridge National Laboratory in 1989, and had been chemically and mass separated. The ^{208}Po to ^{209}Po activity impurity ratio as of the 1994 Reference Time was $0.001\,24 \pm 0.000\,20$ [7]. With decay to November 2013, the $^{208}\text{Po}/^{209}\text{Po}$ ratio was $<10^{-5}$. The standard was originally prepared and put up in March 1993 as $(5.1597 \pm 0.0024)\text{ g}$ of solution in 5 mL flame-sealed borosilicate glass ampoules. The solution used for this work was stored in flame-sealed glass ampoules and under controlled

Table 2. The ^{209}Po massic alpha emission rate E_α for SRM 4326 as a function of measurement time T for 5 determinations, where T is the midpoint time in years for all measurements in the determination. The quantities ΔT , S , U , and n correspond, respectively, to half the time interval in days for the total length of measurements for that T ; the relative total propagated combined standard deviation of the mean for the LS measurement, in percent, considering all within and between components of variance (see text and figure 1 caption); the relative overall combined standard uncertainty, in percent, which includes the estimated uncertainty due to spectral interpretations and analyses; and the number of data sets in the determination.

T/a	$\Delta T/\text{d}$	$E_\alpha/(\text{s}^{-1} \text{g}^{-1})$	$S/\%$	$U/\%$	n
1993.214 96	1.489	85.957	0.19	0.26	8
1993.413 93	12.687	85.869	0.20	0.21	4
1994.184 53	7.521	85.434	0.12	0.19	4
2005.864 86	3.172	80.210	0.10	0.14	4
2013.920 65	9.316	76.526	0.23	0.34	8

laboratory conditions throughout the entire 20.7 years from its initial preparation in 1993 to its final use in 2013. A new original ampoule of the standard solution was opened for each of the determinations. Aliquants of the solution from these ampoules were used to prepare LS counting sources by gravimetric addition using aspirating pycnometers. These mass determinations were made from pycnometer mass differences with calibrated electronic microbalances [7].

All uncertainties expressed in this paper correspond to estimated standard deviations (or standard deviations of the mean where appropriate) or propagated combined uncertainties with a ‘coverage factor’ of $k = 1$, and are wholly compatible with conventions adopted by the principal international metrology standardization bodies [10, 11].

2.1. 1993–1994 determinations

The measurements in 1993–94, as used for the standardization of SRM 4326, were performed over three distinct time periods: (i) in March 1993; (ii) in July 1993; and (iii) in March 1994. They occurred at the midpoint times T shown in table 2, and whose durations are given by $T \pm \Delta T$. The mantissa of T is the fractional part of a mean solar year, i.e., 365.2422 days. At the time in 1993–94, the counting results from all measurements on the three occasions were corrected to a common Reference Time of 1200 EST 15 March 1994 using the then-known (assumed) half-life of (102 ± 5) a. Specific measurement conditions and the counting results for the many trials have been described in great detail by Collé *et al* [7]. For the present work, all of the original data were re-examined to remove the decay corrections, and to ensure that the results were on a comparable basis with the latter 2005 and 2013 determinations. In particular, the overall, propagated LS measurement precision for each data set was recalculated to formulate an overall propagated standard deviation of the mean for the determination that was consistent with that employed for the 2013 data (see below). The revised massic alpha emission rates E_α at midpoint times T with precision statistic S for n data sets for each of the three 1993–94 determinations are given in table 2.

2.2. 2005 determination

The methods, measurements, and results for the 2005 determination were reported in Collé *et al* [5]. That work was used to discern the ^{209}Po half-life discrepancy, as noted previously.

The conditions for these measurements are summarized in table 1. Though these results were not decay corrected at the time, all of the original data in this case were also re-examined to find a more exacting midpoint time, and to verify that the analyses were wholly consistent with that used to obtain the 1993–94 data and with the more recent 2013 result. All of the LS measurement precision estimators were also recomputed. Summary values of mean E_α at T , and S for n data sets for this determination are given in table 2.

2.3. 2013 determination

The 2013 results were obtained with two cocktail series both prepared with UltimaGold AB scintillation fluid, which were measured in two different LS counters (refer to table 1). The cocktails were contained in 20 mL glass LS vials. Each series used two quench conditions to reveal possible electron and photon detection efficiency dependence. Three similar sources at each condition were prepared along with suitable counting blanks. For one of the quench conditions, the efficiency was varied by the addition of an ethanolic solution of nitromethane as an imposed quench agent. The other quench condition was obtained by preparing cocktails with an aqueous fraction of nominal 5% and 10% by the addition of 2 mol L^{-1} HCl, which is the solvent for SRM 4326. The counting sources were essentially identically prepared as those in 1993–94 [7] and 2005 [5], with aliquants in the range 0.3 g to 0.7 g and aqueous fractions of 4% to 10%, with typical LS vial counting rates of 50 s^{-1} .

More than one counter is typically used in our laboratory for any given measurement to ensure that results are independent of the operating characteristics of the counters (detection threshold, photomultiplier efficiency, deadtime, etc), which vary substantially between instruments. It may be of interest to note, however, that over the nearly 21-year span of these ^{209}Po measurements no one counter was used, but there was always an overlap between employed counters for adjacent determinations; i.e., in 1993–94 the counters designated B and P were used (refer to table 1), and P was used along with B2 in 2005, and finally B2 was used with W in 2013. Similarly, as provided in table 1, there was a consistent overlapping of the commercial scintillation fluids that were used. Some of the specific difficulties and issues involved in the liquid scintillation spectrometry of ^{209}Po were also addressed earlier by Collé *et al* [12].

As stated previously, the spectral interpretations and data analyses used for the 2013 result were identical to those developed for the 1993–94 measurements. In brief, the procedure consisted of dividing the spectrum into two regions with a cutoff just above the response due to the electrons from the ^{205}Pb 2 keV transition and the electronic noise. Typical spectra and analysis details are given in full in Collé *et al* [7]. On subtracting this lower energy region from the total spectrum, a reproducible and constant massic rate, attributed to the alpha emission, was obtained for each measurement trial. This rate was then corrected for the response due to radiations from the 0.45% electron-capture branch, taken as $k_{\text{EC}} = 0.9988$ with an estimated uncertainty of about 0.05%. The derivation of this k_{EC} correction is given in an appendix in Collé *et al* [7].

Measurements for the standardization of a new ^{209}Po solution standard (SRM 4326a) were performed concurrently with this 2013 determination. This provided a benefit in giving much more information on the structure of the ^{209}Po spectrum, which aided spectral analyses and on the systematics of the LS response to various conditions, such as quench variations. This work will be reported on separately [13], and resulted in the development of a revised ^{209}Po standardization protocol that employed a more robust spectral analysis procedure, which in turn required an order of magnitude smaller k_{EC} correction. The results for the 2013 data on SRM 4326 (as well as that for SRM 4326a) obtained with either analysis protocol

Table 3. Results for the 2013 measurements of the massic alpha emission rate for the ^{209}Po solution standard SRM 4326. Refer to table 2 and text for description of symbols.

Cocktail series		LS counter	T/a	$\Delta T/d$	$E_\alpha/$ ($\text{s}^{-1} \text{g}^{-1}$)	$s/\%$
Ia	Small quench	B2	2013.893 697	1.9107	76.546	0.304
Ib	Large quench	B2	2013.893 697	1.9107	76.478	0.140
Ia	Small quench	W	2013.917 122	1.8937	76.571	0.215
Ib	Large quench	W	2013.917 122	1.8937	76.493	0.333
IIa	5% Water	B2	2013.931 482	1.7430	76.631	0.149
IIb	10% Water	B2	2013.931 482	1.7430	76.574	0.051
IIa	5% Water	W	2013.944 707	1.3279	76.457	0.272
IIb	10% Water	W	2013.994 707	1.3279	76.462	0.220
Central T			2013.920 65	9.316		
Mean E_α					76.526	
Standard deviation (between-datum for $n=4 \times 2$)						0.083%
'Typical' s (within a datum for $n=3 \times 3$)						0.21%
Combined precision estimator, S						0.23%
Estimated spectral analysis uncertainty (type B assessment)						0.25%
Combined standard uncertainty, U						0.34%

were invariant and statistically equivalent to within the precision estimators of the measurements, a comforting finding. The preparation and standardization scheme for SRM 4326a obtained at this time also provided much more data on polonium solution stability in terms of demonstrating that there were no observable activity losses with serial dilutions, with LS source preparation, and with time.

Table 3 provides a summary of the E_α results for the eight sets of data obtained with four different sets of cocktails (designated as Ia, Ib, IIa, IIb in table 3), each having three similar sources, with each source counted three times on two different instruments. The rates for each source were averaged across the three repetitions (cycles) to obtain a mean and its 'within' standard deviation of the mean. Averaging the source means across the three sources for a trial of a given cocktail composition and counter yielded a grand mean and a 'between' standard deviation. These within and between precision estimators were added in quadrature to provide the propagated standard deviation of the mean (for $n=3$ cycles \times 3 sources) given as the value s in table 3. Figure 1 illustrates the precision in the eight grand mean values, which were combined to form the mean E_α at central time T .

Analysis of variance indicated that there were no statistically significant differences between high and low quench conditions or between LS counters. This approach used here yields a very conservative estimate of the imprecision in the measurements, but is believed to be an appropriate representation of the true experimental variance. The relative standard deviation computed from all 72 values is 0.248%, which gives a minimum-variance estimate of 0.029% for the relative standard deviation of the mean. Figure 2 shows the normal probability plot for all 72 individual measurements ($n=3$ cycles \times 3 sources \times 4 compositions \times 2 counters), along with a frequency histogram to illustrate the distribution. Figure 2 is based on the probability plot correlation coefficient (PPCC) test [14] that was calculated with

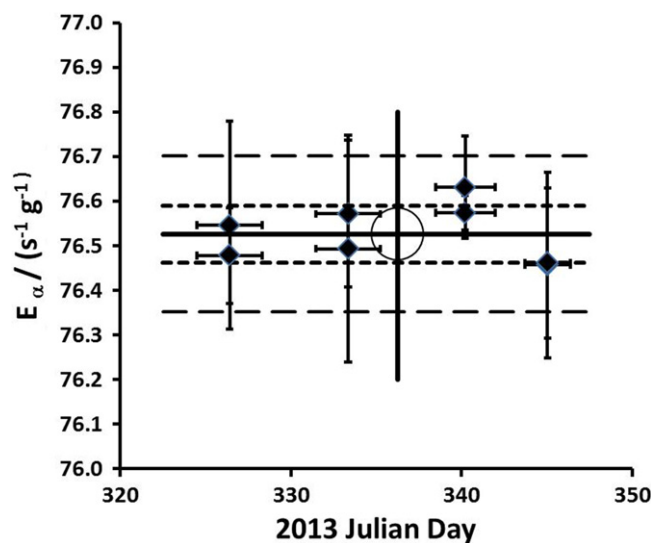


Figure 1. The eight E_α values given in table 3 as a function of measurement time T , illustrating the within- and between-measurement precision in the 2013 data. The vertical uncertainty bars are those for the $\pm s$ values given in table 3, which were derived from the addition in quadrature of the within-cycle component (standard deviation of the mean for $n=3$ measurement cycles on each source) and the between-source component (full standard deviation computed from the means for the $n=3$ sources). The horizontal bar on each datum corresponds to its $T \pm \Delta T$ measurement duration. The solid vertical line is taken as the central value of T for the data set. The mean E_α is given by the solid horizontal line. The inner dotted lines show the between-datum variation in terms of the full standard deviation obtained from the $n=8$ means. This was combined in quadrature with the ‘typical’ standard deviation of the mean (0.21%, as given by the average of the mean and median of the eight s values (i.e., vertical uncertainty bars)) to show the total propagated within- and between-components of variance given by the outer dashed lines. The large open circle represents the mean E_α at central time T .

DATAPLOT [15]. The data passes a Shapiro–Wilk test for normality at a 95% confidence level [16].

3. Half-life result and discussion

The results for the five separate determinations of E_α as a function of T were given in table 2, and are plotted in figure 3 as a decay curve to show the large discrepancy in the apparent ^{209}Po half-life with that obtained using the (102 ± 5) a value as reported by André *et al* [2]. A two-parameter exponential function was least-squares fit for half-life and scale to this data using DATAPLOT [15]. Resultant values for the fitted half-life for various cases are shown in table 4. The cases considered fitting with 5, 4, and 3 datum values, where the four-point fit was obtained by combining the two 1993 results, and the three-point fit combined all 1993–94 values. In addition, the cases considered two different weighting schemes: *viz.*, using weights of $w = U^{-1/2}$ (termed WU in table 4), where U is the relative uncertainty given in table 2; and using equal relative weights of $w = \bar{U}^{-1/2}$ (termed $W\bar{U}$), with $\bar{U} = 0.22\%$ that was obtained from averaging the mean and median values of U . The uncertainties $t_U U_T$ on the fitted half-

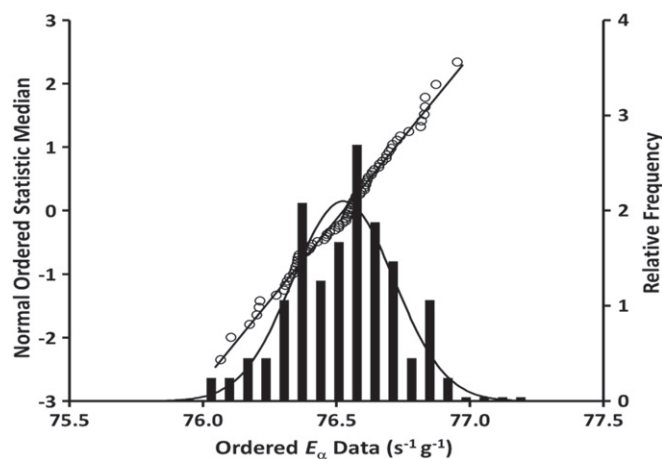


Figure 2. A Fitzgerald plot that overlays the probability plot correlation coefficient (PPCC) graph for all 72 individual LS measurements of the massic alpha emission rate E_α for the ^{209}Po solution standard SRM 4326 as obtained in 2013 and a frequency histogram of the same data. A normal distribution $N(76.52, 0.19)$ is superimposed on the histogram.

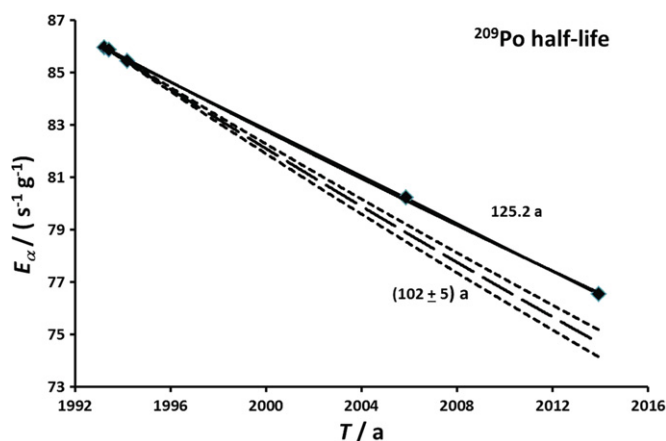


Figure 3 Decay data for the massic alpha emission rate E_α as a function of time T (in years) for the ^{209}Po solution standard SRM 4326. A comparison with the decay curve obtained on assuming a half-life of (102 ± 5) a that shows the large discrepancy with the presently observed half-life of 125.2 a.

lives in table 4 correspond to 68% confidence intervals that consider the number of values used in the fit by expanding the uncertainty in the fit U_T by use of the Student's t standardized variate for $\nu = (n - 2)$ degrees of freedom. One should note that for the equal weighting $W\bar{U}$ cases, the choice of \bar{U} only affects the fit's reduced chi-square χ^2/ν , and not the uncertainty. The relative residuals on the fits for all six cases are given in figure 4.

It is apparent from table 4 and figure 4 that the derived half-life values T for the various cases are largely comparable, which are satisfyingly almost invariant of the weighting scheme and number of values considered in the fit. The relative range in T values is $<2\%$, with a

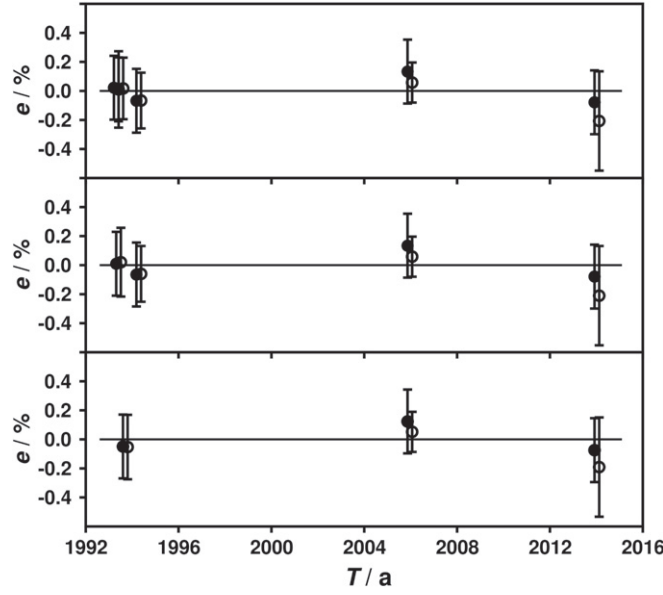


Figure 4. Relative residuals e in percent for the $n=3, 4$, and five-point fits (bottom to top trace) considering both the WU and $W\bar{U}$ weighting schemes as given in table 4 for the ^{209}Po half-life obtained from least squares fits of a two-parameter exponential function to the data. The closed circles correspond to that for the equal relative weight $W\bar{U}$ cases and the open circles for the WU cases. For clarity the e values for the WU cases were shifted slightly to the right on the T scale.

Table 4. Results for the derived half-life $T_{1/2}$ of ^{209}Po in years as obtained from least squares fits of the data in table 2 to a two-parameter exponential function for various cases. Refer to text for descriptions of the two weighting schemes. The value n refers to the number of datum points used in the fit, with $\nu=(n-2)$ degrees of freedom. The uncertainty $t_\nu U_T$ is a 68% confidence interval and χ^2/ν is the reduced chi-square for the fit.

Weighting scheme	Statistic	$n=5$	$n=4$	$n=3$
WU	$T_{1/2}/a$	125.8	126.0	125.2
	$t_\nu U_T/a$	1.7	2.5	5.3
	χ^2/ν	0.47	0.58	0.71
	$T_{1/2}/a$	124.4	124.4	124.0
$W\bar{U}$	$t_\nu U_T/a$	1.4	2.1	4.3
	χ^2/ν	0.44	0.54	0.69

minimum of 124.0 a and maximum of 126.0 a. One might almost be tempted to assign a value of 125 a with an uncertainty of ± 1 a, though this is likely to seriously underestimate the uncertainty. The choice of a specific value for T of those given in table 4 is somewhat arbitrary. Based on the systematics, we have chosen to adopt a value that averages the WU and $W\bar{U}$ cases for the fits of four points. There seemed scant justification for ruling out equal weighting of the values. Use of WU weighting over $W\bar{U}$ was not compellingly better given that the assignments of U values had a large subjective component in estimating the uncertainty due to spectral interpretations and analyses. The four-point fit was chosen because the

Table 5. Uncertainty evaluation for the $T_{1/2} = 125.2$ a ^{209}Po half-life, given in terms of both a standard uncertainty interval in years and as a relative value in percent. The A and B designators refer to the type of assessment used to evaluate the component [10, 11]. The assignment for each component is delineated in the text.

Uncertainty component	Type	u/a	$u/\%$
Least-squares fit	A	2.3	1.8
Weighting model choice	B	0.3	0.2
Long-term effects	B	2.4	1.8
Quadrature combination		3.3	2.6

five-point fit clearly overweights the 1993–94 values, whereas the three-point fit undervalues the wealth of 1993–94 data compared to that in 2005 and 2013.

With this, the ^{209}Po half-life is given as

$$T_{1/2} = (125.2 \pm 3.3)\text{a},$$

where the uncertainty bounds correspond to a standard uncertainty for $k=1$ [10, 11] or an assumed 68% confidence interval. The uncertainty assignment is based on that given in table 5. The ‘least squares fit’ component was taken as the median of $t_{\nu}U_T$ in table 4 for the $n=4$ WU and W \bar{U} cases. The ‘weighting model choice’ component was estimated from the differences in the central values of the half-lives T for the two models (table 4) that were in excess of the least squares fitting uncertainty. The component for ‘long-term effects’ includes consideration of the uncertainty due to differences in examining and treating the LS spectra under varying conditions of cocktail composition, quenching, and instrument characteristics. This component was evaluated through a sensitivity analysis, *viz.* by calculating the magnitude of the change in $T_{1/2}$ that would result from systematically warping the E_{α} data by a factor of $f=(1+\delta T)$ over the 20.7 a duration of the measurements, where δ is given by $\delta=(0.0022/20.7)\text{a}^{-1}$ for the relative uncertainty $\bar{U}=0.22\%$. This examination of possible long-term effects is consistent with the protocol given by Pommé *et al* [17].

One must appreciate that there is a large uncertainty in any half-life determined by decay when the decay period is a short compared to the half-life, as well noted by Pommé *et al* [17]. The half-life uncertainty varies inversely with the time duration of the decay. For illustrative purposes, consider that the relative uncertainty on a 125-year half-life based on just two values separated by 21 years is approximately 12 times the uncertainty on the values. For example, if each of two measurement values separated by 21 years has a relative uncertainty of 0.2%, then the uncertainty in the 125-year half-life would be 2.4%.

4. Conclusion

A ^{209}Po half-life value of $(125.2 \pm 3.3)\text{a}$ is supported by precise decay data obtained on five measurement occasions over a period of 20.7 years. The data were acquired from primary standardization measurements of the massic alpha emission rate for a pure, carrier-free solution standard of ^{209}Po (SRM 4326). The present result is in agreement with and confirms the original finding of a 25% error in the ^{209}Po half-life [5] that was extant for over 50 years. It is hoped that future nuclear data evaluations and compilations will not incorporate the previous value of André *et al* [2] with the present result to obtain a new reference value for databases. Given the absence of any evidence on the origin or quality of the value, or any documentation on the experimental methods used to obtain it in the mid 1950s, the 102 a value remains wholly unsubstantiated.

5. Disclaimer

Certain commercial equipment, instruments, or materials are identified in this paper to foster understanding. Such identification does not imply recommendation by the National Institute of Standards and Technology, nor does it imply that the materials or equipment identified are necessarily the best available for the purpose.

Acknowledgements

This culminates nearly 25 years of work on ^{209}Po by this laboratory, and has had many collaborators and coauthors over the years whose contributions have been noted in previous publications. Contribution of NIST.

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