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PAPER

A Precise Determination of the ^{222}Rn Half-Life by 4π-αβ Liquid Scintillation Measurements

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The half-life for ^{222}Rn radioactive decay was determined by 4π-αβ liquid scintillation (LS) measurements using gravimetrically-determined aliquants from the National Institute of Standards and Technology (NIST) radon-in-water standard generator. The decay of ^{222}Rn in six matched samples, contained in flame-sealed glass LS vials, was followed for approximately 15 days (about four half-lives) with normalizations to a common similarly-prepared ^{226}Ra reference source. With the ^{218}Po-^{214}Pb-^{214}Bi-^{214}Po subseries following the ^{222}Rn in transient equilibrium, the overall LS detection efficiency for ^{222}Rn was very nearly 5 counts · s^{-1} · Bq^{-1}. Instrument nonlinearity tests over the entire range of measured activities (roughly 100 to 5 Bq) were also performed with a series of ^{226}Ra reference LS samples. All ^{226}Ra reference sources contained ^{222}Rn in secular equilibrium. The LS measurement results for the six decay curves, each having 120 data values, were fitted by both weighted and unweighted least-squares exponential regressions. The relative uncertainties in the weighted regressions for the six half-life determinations ranged from 0.049 to 0.060 percent. The six determinations had values ranging from T = 3.8200 to 3.8259 d, with a weighted mean (weighted by the reciprocal variance) of T = 3.82248 ± 0.00086 d (±0.022 percent uncertainty on a relative basis); and with an unweighted mean of T = 3.82252, having a standard deviation of 0.00247 d (±0.065 percent) and a standard deviation of the mean of 0.00086 d (±0.026 percent). Based on a detailed uncertainty analyses, an adopted value of T = 3.8224 ± 0.0018 d is justified.

Introduction

The record of previous determinations of the half-life for radioactive decay of ^{222}Rn is long and curious, virtually extending over the entire historical course of the science of radioactivity. This historical record has recently been extensively reviewed in a companion paper.[1] Following is a capsulized chronological summary of the mean values of the past determinations:[a]

P. Curie[3]	1902	T = 3.987 days
Rutherford and Soddy[4]	1903	3.71
Bumstead and Wheeler[5]	1904	3.896
Sackur[6]	1905	3.863
Rümelin[7]	1907	3.747
Bronson[7]	1907	3.72
M. Curie[8,9]	1910	3.85
Rutherford[10,11]	1911	3.847
Bothe and Lechner[12]	1921	3.811
Bothe[13]	1923	3.824
I. Curie and Chamié[14]	1924	3.823
Tobailen[15]	1955	3.8235

a For comparative purposes, all literature values reported in terms of other decay parameters, such as a decay constant $\lambda = \ln 2/T$ or average lifetime $\tau = 1/\lambda$, were uniformly transformed into a common half-life parameter T. All reported half-life or transformed half-life values in other time units were converted to units of days. To avoid possible rounding errors, one additional significant figure was included here in all citations, tabulations, and calculational analyses whenever parameter transformations or time unit conversions were made. Precision estimators for these mean values are not reported here in order to avoid confusions and numerous justifications of underlying conversion assumptions and provisoes. They are discussed and treated at length elsewhere.[1,2]

Marin[16]	1956	3.8231
Robert[17]	1956	3.825
Shimanskaia[18]	1956	3.83
Butt and Wilson[19]	1972	3.82351

The vast majority of these reported determinations were based on ionization chamber, including early electroscope, measurements. The only exceptions are the adiabatic microcalorimetry determinations by Robert[17] and by Shimanskaia,[18] and the precise NaI(Tl)-scintillation-detector measurements of Butt and Wilson[19] that extended over approximately 20 half-lives of ^{222}Rn. The methods used to derive the determined half-life varied considerably over the years. Initially, circa 1902-1924, the half-lives were determined by nonredundant two-datum point solutions, e.g.,

$$T = -\ln 2\ (t_2 - t_1) / \ln(N_2/N_1)$$

for a decay measurement pair of N_1 at time t_1 and N_2 at t_2, or various combinations of these nonredundant solutions on one or more ^{222}Rn samples. Later determinations made in the mid-1950s were obtained from linear least-squares regressions of logarithmic transformations of the measurement decay data to the exponential decay law, i.e., in which n pairs of (N_i, t_i) data ($i = 1,2,3\ldots,n$) were simultaneously χ^2 minimized and fitted to linear logarithmic equations in a form such as $\ln N_i = a - b t_i$ where the slope b is the decay constant equal to $\ln 2/T$. The advent of modern computer-based calculational methods,[20] based on matrix manipulations for the simultaneous solution of multiple equations, allowed Butt and Wilson[19] to perform more rigorous χ^2-minimizations to the exponential form $N_i = a\, e^{-(\ln 2/T)t_i}$ itself.

The most evident feature of this chronological record is that the reported values of T have, in general, progressively decreased. This is perhaps unsurprising since almost any type of activity-level-dependent nonlinearity in instrument response, e.g., ion-pair recombination effects, ionization current saturation, dead-time or pulse pileup losses, has the effect of lengthening the apparent half-life. Other features, not readily apparent in the summarized results presented here, but quite evident and addressed at length elsewhere,[1,2] are the progressive improvement in reported measurement precisions and that the central values of T for the various precise determinations barely overlap the uncertainty intervals of other precisely reported results. The estimated relative precision in these determinations decreased from about 10^{-2} in 1910, to about 10^{-3} in 1923-1924, to less than 10^{-4} in 1956-1972. In fact, the most recent five-to-six significant figure value reported by Butt and Wilson[19] often is considered to dominate the data set of previous determinations.

Most compilations and tabulations of nuclear and radioactive data (see for example references 21-23), provide a ^{222}Rn half-life of $T = 3.8235 \pm 0.0003$ d. This value appears and is sometimes cited to be entirely based on the reportedly precise Butt and Wilson[19] determination, and seemingly ignores a substantial data base of other determinations. The situation of a compiled physical datum being either exclusively based on one reportedly precise determination of that datum or dominated by that single determination, if conventional statistical weighting with other values are made, is hardly unique to this case and is somewhat well known but regrettable. The result, of course, is that the compiled data sets are often left to the mercy of experimental optimists. This has sometimes been remedied by using methods of limitation on the relative weights, such as by not allowing a relative maximum statistical weight of greater than 50 percent for any one value within an inconsistent data set.[24-26] Such critical evaluations, however, are increasingly rare in modern data compilations.

An exception is the recent critically-evaluated compilation by an International Union of Pure and Applied Chemistry (IUPAC) commission that provides a ^{222}Rn half-life value of $T = 3.823 \pm 0.004$ d.[27] For this IUPAC study, the experiments of the last seven reported determinations[13-19] were reviewed. The reported uncertainties for nearly every one of these determinations were considered to have been underestimated and were then "revised" either because of the absence of detailed information on the uncertainties, or because discussions of possible "systematic errors" were inadequate, or because the "total error" was extremely small and thereby presumed to be unbelievable. A weighted mean of the seven determinations (with revised uncertainties) results in $T = 3.823 \pm 0.001$ d, however the IUPAC study further invoked an "uncertainty rule" that virtually dictated that no recommended half-life value have a relative uncertainty of less than 0.1 percent. This rule evidently follows a similar general recommendation of an International Atomic Energy Agency (IAEA) review[28] that seriously questioned the validity of any stated relative uncertainty of less than 0.1 percent for a half-life. This very conservative approach (and the revision of reported uncertainties) may appear justifiable when applied to a single measurement result or even to a few determinations, but the general application of an arbitrary "≥0.1 percent uncertainty rule," particularly when there are a considerable number of independent determinations, does not necessarily seem well founded.

Further, in regard to the Butt and Wilson[19] determination, one might argue and reasonably suspect that no other ^{222}Rn half-life determinations have been reported over the past intervening 20 years because most other researchers would find it difficult, if not impossible, to design experiments that could compete with their reported precision. This last stated supposition, if true, is regrettable. Considering that few single determinations of a radioactive half-life to a relative precision of better than 10^{-4} can withstand critical scrutiny,[b] there is tre-

b The contention that few—very few—experimental determinations can support a precision of five significant figures is based on the author's experience, as well as the IUPAC and IAEA considerations cited previously.[27,28] The routinely tabulated half-life value for ^{127}Xe decay is a perfect example. Despite subsequent determinations and critique by Collé and Kishore,[29] the compiled ^{127}Xe half-life is still dominated by a single reportedly precise measurement that in reality is not likely to be known any more precisely than other measurements.

mendous merit and statistical power in having multiple independent determinations. It should be self-evident that the reliability in any physical datum is vastly improved by replicated determinations made by different experimentalists, at different times, under different conditions, with different sources, and with widely different instruments and measurement methods.

The work of this study is, in part, reported here on the basis of this motivation. It provides the first precise determination of the ^{222}Rn half-life by liquid scintillation (LS) measurements. The work was performed in the period December, 1989 to January, 1990 for other purposes and has only recently been reanalyzed in this light. The results of this study are based on LS measurements of six sources over a period of 15 days (about four ^{222}Rn half-lives).

Experimental aspects

Sources: preparation and considerations

The counting sources were prepared by dispensing aliquants of a ^{222}Rn aqueous solution from the NIST radon-in-water standard generator.[30,31] The standard generator consists of a polyethylene-encapsulated ^{226}Ra solution source in a small-volume accumulation chamber and an ancillary mixing and dispensing system which is partially automated with motor-driven syringes. It can be used to generate and accurately dispense radium-free ^{222}Rn solutions of known activity concentration.

The ^{222}Rn solution aliquants were dispensed beneath the surface and at the bottom of nominal 20-mL, glass, flame-sealable, LS vials which previously had been filled with an LS cocktail fluid. The scintillator fluid was "PCS," a xylene-surfactant-based scintillator.[c] Each sample vial contained approximately 7.5 g of ^{222}Rn solution and 13.0 g of PCS, and was subsequently flame-sealed with an especially-designed sealing cryostat maintained during sealing at a temperature of about 200°K (-77 to -72°C) that was obtained with a solid CO_2 (dry ice)/isopropanol slurry. The masses of the sample components in each sample were gravimetrically determined to a relative standard uncertainty of about 0.05 percent.[d]

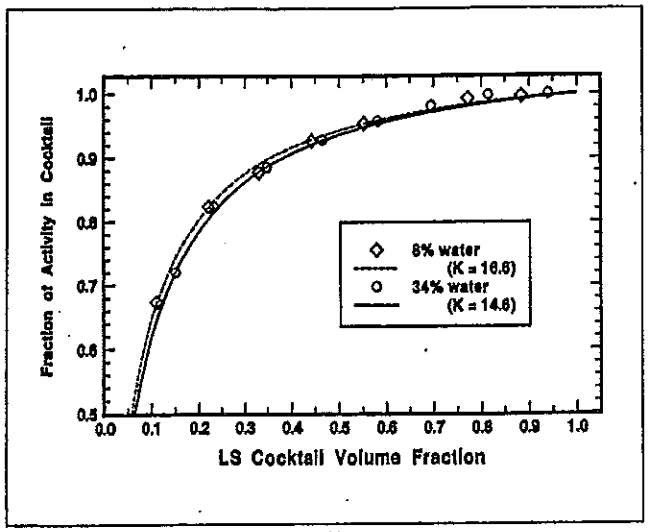

Figure 1 Fraction of the total ^{222}Rn activity in a LS sample (for two water/PCS scintillator cocktail mixes) as a function of the volume fraction of the cocktail in the sample demonstrating the partitioning of radon between the cocktail and air volumes.

The use of flame-sealed vials eliminated the possibility of radon losses from the sample even over long measurement intervals. Conventional screw-capped LS vials, with either vee-shaped polyethylene liners or cork-backed aluminum foil liners, exhibit evaporation losses of volatile scintillator fluids, and therefore were not considered to be sufficiently adequate for insuring the containment of the readily diffusive radon gas over long times.[e] Prior to filling and sealing, the total internal volumes of the vials up to scribed fiducial marks at their sealing points were gravimetrically determined by fillings with high-purity water having a known density. Estimated air-space volumes in the vials after filling with the PCS fluid and ^{222}Rn solution and after sealing ranged from 0.04 to 0.09 cm^3 in the six samples. Based on independent measurements of the partitioning of radon between the cocktail and air space (Figure 1),

c Amersham Corp., Arlington Heights, IL. The mention of commercial products throughout this paper does not imply recommendation or endorsement by the National Institute of Standards and Technology, nor does it imply that the products identified are necessarily the best available for the purpose.

d Unless otherwise noted, all uncertainties or individual uncertainty components provided throughout this paper are expressed in terms of "standard uncertainties" which are estimated (experimental) standard deviations or uncertainty quantities assumed to correspond to standard deviations, irrespective of the method used to evaluate their magnitude. Similarly, a combined or propagated uncertainty is expressed as an estimated standard deviation which is equal to the positive square root of the total variance obtained by summing all variance and covariance components, however evaluated, using the law of uncertainty propagation for the specific mathematical function given by the model of the measurement procedure. These practices are consistent with policy and conventions uniformly adopted by NIST,[32] and are compatible with those adopted by the principal international metrology standardization bodies.[33]

e Evaporation losses of the xylene-based PCS scintillator in conventional LS vials with cork-backed aluminum foil liners is considerable and highly variable. Based on mass loss measurements with 32 vials over 30 to 40 days, the mean evaporation loss rate was found to be 1.9 mg · d^{-1} with a relative standard deviation of the mean of 20 percent. The scintillator evaporation rate for vials with vee-shaped polyethylene liners is substantially less. However, their use introduces a different complication. Radon is readily soluble in polyethylene and it partitions between the air space and polyethylene liner in LS samples.

the fraction of radon contained in the air space was estimated to be in the range from 0.0001 to 0.0003.[f]

Two matched aqueous blanks of nearly identical sample composition were also prepared to perform background subtractions. These blank samples (along with the water used in the standard generator) were prepared with aged, ion-exchanged, "radon-free," distilled water having a total α- or β-emitting radionuclidic impurity, in terms of an equivalent ^{222}Rn activity concentration, of less than 0.003 ± 0.001 Bq · g^{-1}.

A ^{226}Ra reference source of similar composition, containing 4.5617 g of a solution having 19.30 ± 0.16 Bq · g^{-1} ^{226}Ra in 14.5173 g PCS, was also prepared and used for between measurement normalizations.[g]

LS measurement system

The measurements were performed with a Beckman LS7800 model LS counter equipped with two Hamamatsu R331-05 photomultiplier tubes operating in a coincidence mode, a logarithmic pulse amplifier coupled to an analog-to-digital converter (ADC) for spectral pulse-height analyses, and an external ^{137}Cs source for Compton-edge (Horrocks number) quench monitoring.[35] The Horrocks number ($H^{\#}$) is a quench indicating parameter that is based on the downward spectrum shift of the Compton edge of the external ^{137}Cs standard with increasing sample quenching. The parameter corresponds to the spectral channel number shift between the sample and an unquenched blank sample.

Source and Instrument characteristics

Measurements were initiated only after the sealed LS samples contained ^{222}Rn in radioactive equilibrium with its short-lived ^{218}Po-^{214}Pb-^{214}Bi-^{214}Po subseries (i.e., after a minimum of 5 to 6 hours). A typical LS spectra of ^{222}Rn in secular equilibrium with its four decay-product progeny in these samples is shown in Figure 2. The overall LS detection efficiency is almost exactly 5 counts · s^{-1} · Bq^{-1} resulting from a very nearly 100 percent efficiency for the three α decays (^{222}Rn, ^{218}Po, and ^{214}Po) and an almost comparable 100 percent efficiency for the two energetic β$^-$ decays (^{214}Pb and ^{214}Bi). Based on the CIEMAT/NIST ^{3}H efficiency tracing method[36-38] using theoretical modelling

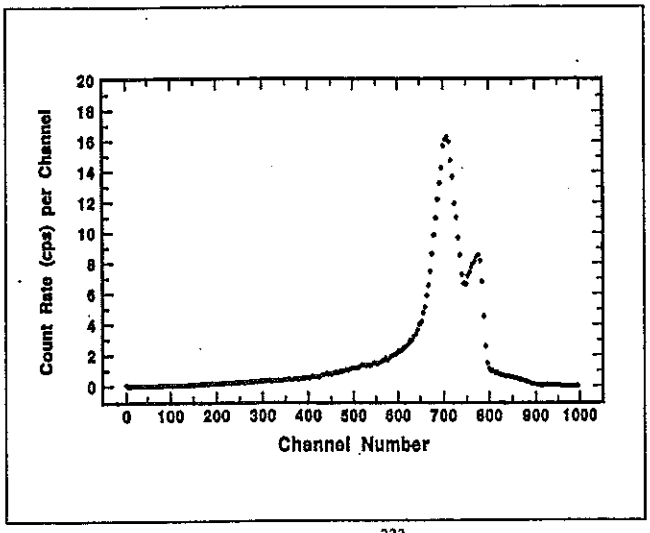

Figure 2 A typical LS spectrum for ^{222}Rn in radioactive equilibrium with its short-lived ^{218}Po-^{214}Pb-^{214}Bi-^{214}Po decay subseries.

calculations and measurements with matched sets of ^{3}H LS samples, the LS counting efficiency for the ^{222}Rn subseries in unquenched samples was found to be

$$4.977^{+0.008}_{-0.015} \text{ counts} \cdot s^{-1} \cdot \text{Bq}^{-1}.\ ^{h}$$

Average quenching summed over all six ^{222}Rn sources over all 120 measurements on each source was $H^{\#} = 141 \pm 6$, and remained constant with only random statistical variations throughout the measurements. The temporal variations in Horrocks number for one of the samples over the entire 15-day course of measurements is illustrated in Figure 3. The $H^{\#}$ values for this sample, which was typical of that for any other sample, ranged from 132 to 144 with a mean (solid line in Figure 3) of $H^{\#} = 138.7$ and a relative standard deviation interval (dotted lines) of ±1.5 percent. The results clearly demonstrate the

f The fraction of ^{222}Rn activity in the LS cocktail, A_f, as a function of the volume fraction of the cocktail in a LS sample, V_f, is given by $A_f = 1/[1+1/K(1/V_f - 1)]$ where K is the partition coefficient, or Ostwald coefficient, for the ratio of the solubility of radon in the cocktail mix to the solubility of radon in air. Least-squares regressions of data (typical to that given in Figure 1) to the above function to obtain fitted values of K are inherently imprecise. Comparable data sets for other water to PCS scintillator mixes yielded values of $K = 12.9$ (8 percent water); $K = 13.8$ (11 percent water); $K = 19.5$ (25 percent water); $K = 17.4$ (32 percent water); and $K = 10.5$ (51 percent water). Invariably, for any water-to-scintillator mix in the range of 10 percent to 50 percent water, the partition function was in the general range of $K = 10$ to $K = 20$.

g All ^{226}Ra reference sources used for this work were prepared from gravimetric dilutions of stock solutions that were used to prepare NIST Standard Reference Materials SRM 4965, SRM 4966, and SRM 4967. The "age" of the radium in the solutions, with accompanying ingrown of ^{210}Pb, is at least 44.3 years, as of September 1991. Refer to the original NIST Certificates for additional details on the calibration and characteristics of these solutions.[34] The large ingrowth of ^{210}Pb results in an apparent LS detection efficiency of about 7.87 counts · s^{-1} · Bq of ^{226}Ra (with the ^{222}Rn subseries in radioactive equilibrium with ^{226}Ra) for sources prepared from these solutions.

h The efficiency-tracing quench corrections include appropriate extrapolations for the scintillator-to-water ratio in the LS cocktail; for partitioning of radon between the scintillator cocktail mix and the air space in the samples; and for count-rate-versus-energy extrapolations to zero energy.

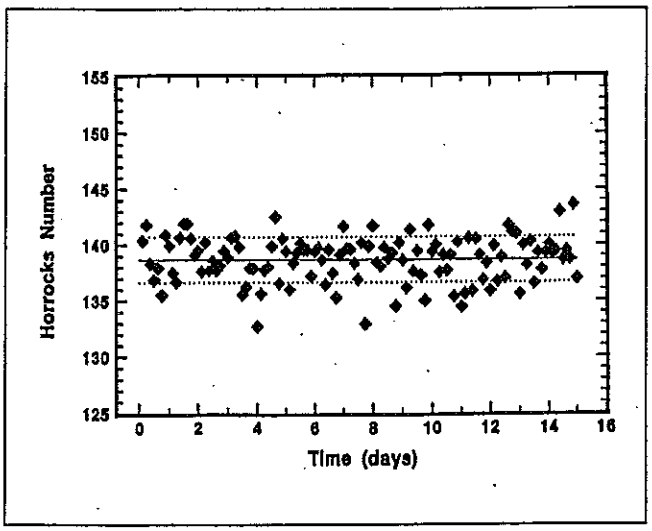

Figure 3 Variation of the quench indicating parameter, Horrocks number, for one of the ^{222}Rn LS samples over the course of the decay measurements.

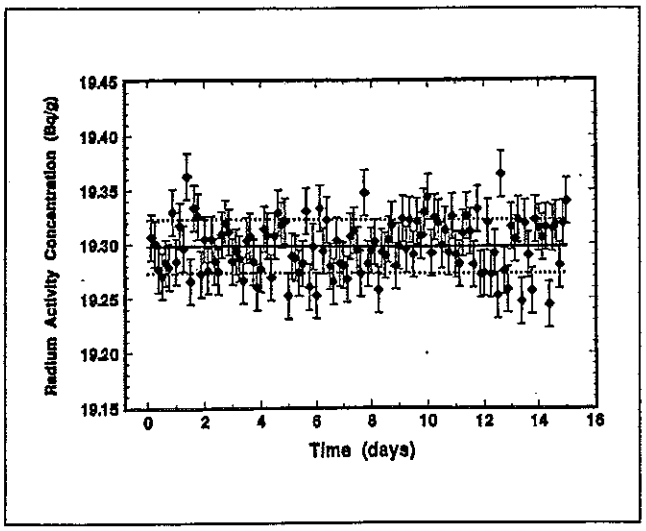

Figure 4 Variation of the ^{226}Ra reference source over the course of the decay measurements. The solid and dotted lines correspond to the mean and the upper and lower bounds of the one standard deviation uncertainty interval, respectively. Refer to text for details.

sample homogeneity and cocktail stability over the entire measurement course.

Each of the six ^{222}Rn LS samples along with two matched blanks and a common ^{226}Ra reference source were measured 120 times over 15 days to derive the six decay curves. To obviate possible temporal variations in the LS instrument response over the 15-day measurement interval, each measurement on each ^{222}Rn sample was normalized by the response obtained from the ^{226}Ra reference source for that measurement cycle. That is, the background-corrected and normalized count rate for sample j (j = 1,2,...6) during measurement cycle i (i = 1,2,...120) at time t_i was obtained from

$$R_j(t_i) = \frac{(R_{Rn_{j,i}} - R_{B_i})}{(R_{Ra_i} - R_{B_i})} \overline{R}_{Ra}$$

where $R_{Rn_{j,i}}$ = the uncorrected count rate for ^{222}Rn sample j in measurement cycle i; R_{B_i} = the average count rate of the two blanks obtained during measurement cycle i; R_{Ra_i} = the count rate of the ^{226}Ra reference source in measurement cycle i; and $\overline{R}_{Ra}$ = the mean background-corrected count rate of the ^{226}Ra reference source over all 120 measurement cycles, i.e.,

$$\overline{R}_{Ra} = \sum_{i=1}^{120} (R_{Ra_i} - R_{B_i}) / 120 \ ,$$

and where t_i is the midpoint time of the measurement interval for $R_j(t_i)$. The starting time $t = 0$ for each sample j corresponds to a common reference time 6 hours after the end of the ^{222}Rn accumulation period in the standard generator.[31]

The relative statistical (Poisson) counting uncertainty obtained from propagating the variances on $R_{Rn_{j,i}}$, R_{B_i}, R_{Ra_i}, and $\overline{R}_{Ra}$ (i.e., the total number of counts obtained for each) ranged from about 0.15 percent for measurement cycle i = 1 to about 0.6 percent for i = 120 in all six samples.

Figure 4 demonstrates the variations in R_{Ra_i} over the course of the 120 measurement cycles. Each of the represented activity concentration values of Figure 4 were obtained from R_{Ra_i} - R_{B_i} after division by common factors of 4.5617 g of ^{226}Ra solution in the sample and an assumed ^{226}Ra subseries detection efficiency of 7.87 counts · s^{-1} · Bq^{-1}. The uncertainty interval bars shown on the plotted individual values correspond to the one standard deviation statistical (Poisson) counting uncertainties. The values have variations ranging from 19.24 to 19.36 Bq · g^{-1} with a mean value of 19.298 ± 0.025 Bq · g^{-1}. This mean value and one standard deviation uncertainty interval is represented in Figure 4 by the solid and dotted lines, respectively.

An independent study of possible activity-level-dependent instrument nonlinearities, such as that due to uncorrected dead-time losses, was also conducted. For this study, a set of seven matched ^{226}Ra reference sources were prepared. These LS sources contained varying masses of gravimetrically-determined aliquants of a ^{226}Ra solution standard (each with a relative mass measurement uncertainty of ±0.05 percent), and had counting rates that extended beyond the ranges of that obtained in the ^{222}Rn decay measurements. The resultant LS measurements were normalized to the mean apparent ^{226}Ra detection efficiency (with ^{222}Rn subseries in radioactive equilibrium) obtained from the seven sources. The relative detection efficiencies ε_R as a function of LS counting rate R are illustrated in Figure 5. The uncertainty intervals shown correspond to relative standard deviations of the mean for 6 replicate measurements of each source. The mean ε_R = 0.99975 ± 0.00044. In this case, in which systematic activity-level (or count rate) dependent nonlinearities are of interest, a linear

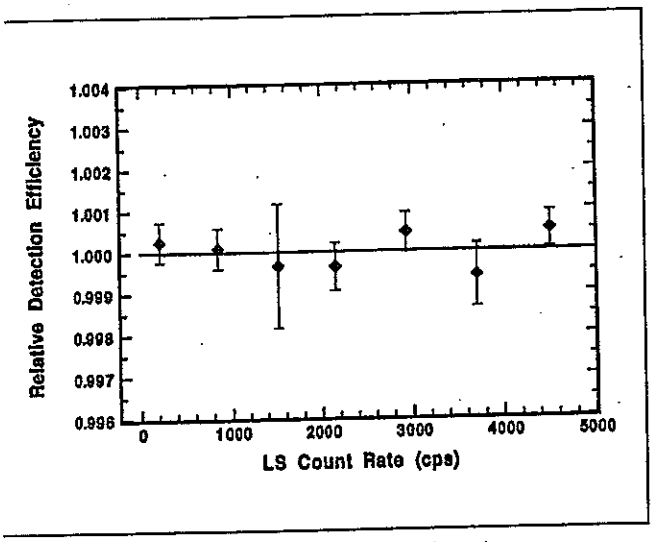

Figure 5 Invariance (linearity) of the LS detection response over the range of counting rates for the ^{222}Rn decay measurements, as reflected by the relative detection efficiency obtained from a set of matched ^{226}Ra reference sources. The uncertainty bars on the individual datum points correspond to standard deviations of the mean for 10 to 14 measurements of each source.

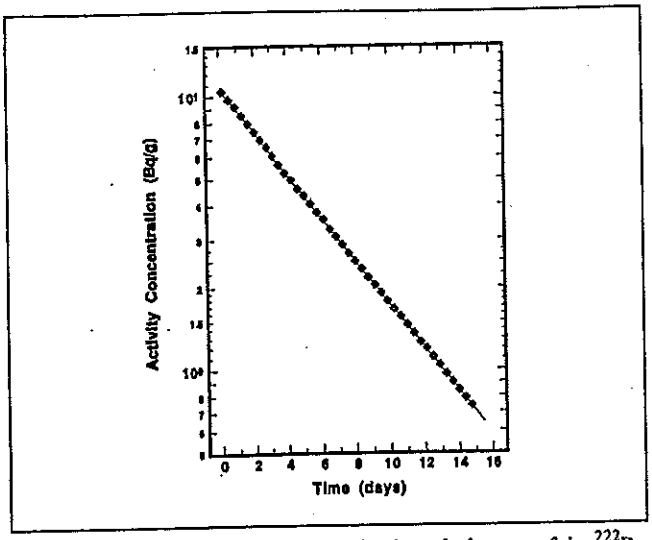

Figure 6 Decay curve on a logarithmic scale for one of the ^{222}Rn LS sources (labelled P8). Only every third datum of the 120 measurements of the decay curve is plotted.

regression of the data may be more informative than individual source deviations from the mean. The linear regression result

$$\varepsilon_R = 0.99996 \pm 0.00034 + (1.6 \pm 0.1)10^{-8}\, R$$

clearly demonstrates the relative invariance of the detection response over the range of counting rates. The regression slope $d\varepsilon_R/dR$ is of the order of 10^{-8} d^{-1}. Based on the uncertainty in the regression intercept and in the uncertainty in the mean ε_R, the measurement system nonlinearity is taken to have a corresponding relative uncertainty of about 0.03 to 0.04 percent.

The uncertainty due to temporal variations in instrument response and of LS sample stability over the course of the 15-day decay measurement interval may be similarly evaluated by consideration of the variations in the ^{226}Ra reference source responses R_{Ra_i} (Figure 4). A linear regression of R_{Ra_i} normalized by the mean $\overline{R}_{Ra}$ gives a slope of $d(R_{Ra_i}/\overline{R}_{Ra})/dt = 0.000028 \pm 0.000027$ d^{-1} and an intercept of 0.99979 ± 0.00024. Taking the limits of the regression uncertainties at 15 days, the resulting relative uncertainty due to temporal variations may be considered to be 0.05 percent. The mean $\overline{R}_{Ra}$ over all 120 measurement cycles has relative uncertainties corresponding to 0.13 percent for the standard deviation and 0.012 percent for the standard deviation of the mean. The magnitude of these uncertainties are supported by the observed temporal variations in the quench parameter (Figure 3). A similar linear regression analysis of $H_i^{\#}/\overline{H}^{\#}$ as a function of t_i gives relative uncertainty limits of ±0.2 percent, and the mean $H^{\#}$ has a relative standard deviation of the mean of 0.12 percent. It must be emphasized however that the determinations of $H^{\#}$ are inherently imprecise and the precision estimators will of necessity overestimate the temporal instrument variability and LS sample stability. It should also be recalled that the $R_j(t_i)$ values were normalized by the responses of the ^{226}Ra reference source to minimize temporal instrument variations. Therefore, a relative uncertainty of about ±0.05 percent for possible instrument and LS sample variabilities and instabilities is certainly reasonable, and probably somewhat of a conservative estimate.

Measurement results and discussions

Decay curves and regressions

For convenience in comparing the six ^{222}Rn samples, the background-corrected and normalized values of $R_j(t_i)$ in units of counts · s^{-1} were converted to activity concentrations in Bq · g^{-1} using the gravimetrically-determined mass of each sample m_j (having a relative estimated standard uncertainty of ±0.05 percent) and the mean LS detection efficiency of

$$\varepsilon = 4.977^{+0.008}_{-0.015} \text{ counts} \cdot s^{-1} \cdot \text{Bq}^{-1};$$

e.g., $A_j(t_i) = R_j(t_i)/m_j\varepsilon$. Figure 6 shows a decay curve for one of sources (labelled P8) on a logarithmic scale expressed in terms of the midpoint times t_i. The straight line is a weighted χ^2-minimized (least squares) regression of an exponential to the data and corresponds to a fitted half-life of $T = 3.82116 \pm 0.00228$ d. Of the six decay curves, this illustrated regression had the poorest fit with a "goodness-of-fit" parameter of $\hat{s}_{fit} = 1.76$. The regressions were performed using a functional form $A(t_i) = A_0 e^{-(\ln 2/T)t_i}$ and the data were weighted by the reciprocal of the propagated statistical (Poisson) counting uncertainties in $R_j(t_i)$ (see above). Table 1 summarizes the results for all

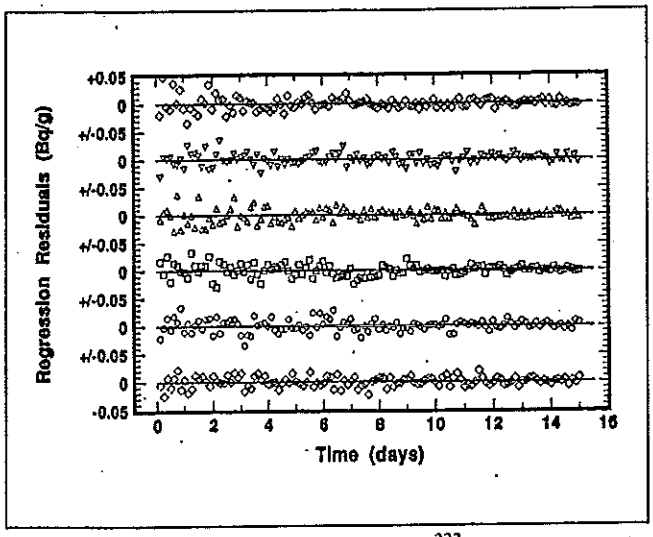

Figure 7 Regression residuals for the six ^{222}Rn LS sources. The residuals are given in terms of the activity concentration in units of $Bq \cdot g^{-1}$.

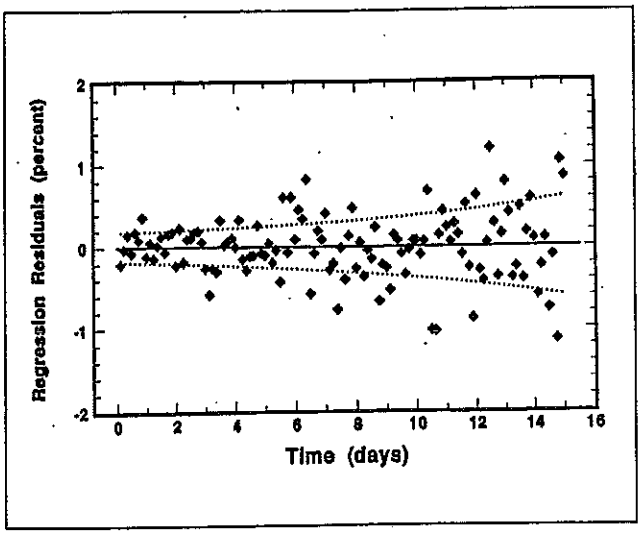

Figure 8 Relative regression residuals expressed in percent for one of the ^{222}Rn LS sources (P4). The dotted lines represent the upper and lower bounds of the relative one-standard-deviation uncertainty interval for the statistical (Poisson) counting uncertainty in each datum point about a residual of zero.

six decay curves. It contains the fitted half-life values T and corresponding standard uncertainties, the fitted intercepts which are the activity concentrations A_0 at time $t = 0$ and their standard uncertainties, and the $\hat{s}_{fit}$ parameters. The tabulated activity concentrations A_0 are nearly constant because the six LS samples were aliquants of an identical ^{222}Rn solution dispensed from the standard generator, and they correspond to the activity concentration of that solution.

The regression residuals for all six decay curves are illustrated in Figure 7. These residual results better illustrate the quality of the regressions and clearly are more informative than the decay presentation of Figure 6. In Figure 7, the residuals in absolute units of $Bq \cdot g^{-1}$ decrease with increasing time. However, when compared on a relative basis as is done in Figure 8 for sample P4, it is clear that the dispersion in the residuals is largely controlled by the underlying statistical counting precision.

Calculational considerations

For completeness, four calculational considerations should be mentioned. They follow.

First, the fitted half-life results reported here are, of course, independent of the additional uncertainties in m_j and ε since these terms are common to all $A_j(t_i)$ values used in the regressions. The $R_j(t_i)$ values were converted to $A_j(t_i)$ mainly to compare the fitted intercepts A_0 for the six samples. Regressions to $R_j(t_i)$ data would give identical results for the fitted T values.

Second, the decay curves were also evaluated in terms of unweighted exponential regressions (Table 2) to insure that there were no surprising differences with the results from weighted regressions. This was, in part, done because weighted regressions have the virtual effect of decreasing the lengths of the effective decay intervals for the half-life determination.[i]

For this work, there were no necessarily compelling reasons to justify the use of weighted over unweighted regressions. The

i It may be arguably demonstrated that a weighted regression reduces the effective time duration of a half life determination as follows: For n sequential measurements at times t_k (k = 1,2,3...n), the total time duration (unweighted) of the determination may be given by $t_u = \sum_{k=1}^{n} (t_k - t_{k-1})$ which is just $t_u = (t_n - t_0)$. Analogously, with normalized weighting factors of $w = s_k^2/s_t^2$, the effective time duration becomes $t_w = \sum_{k=1}^{n} w(t_k - t_{k-1})$. In the present work, t_u is approximately 15 d. With the reciprocal variance weighting factors used in the weighted regressions, t_w is 5.2 d.

j Differences in relative weights were even less if one incorporates the additional uncertainties due to instrument activity-level dependent linearities and the temporal instrument and source variabilities as demonstrated later. This in large part accounts for the insignificant differences in the weighted to unweighted regression results.

relative weights from the first (i = 1) to last (i = 120) measurement varied by only a factor of about 16;[j] and the measurement system was demonstrated to be substantially constant and linearly responsive over the entire decay intervals (Figures 3, 4, and 5). The effect of using weighted regressions over unweighted ones for the six decay curves was merely to slightly improve the uncertainties in the fitted parameters with no statistically significant changes in the values of the parameters themselves. This is clearly demonstrated in Table 3 where the differences in the fitted parameters are compared, and more vividly in Figure 9 for the fitted T values. In every case in Figure 9, the one standard deviation uncertainty intervals for fitted T values from the weighted and unweighted regressions overlap. This agreement between the weighted and unweighted regression results also supports the contention that there were no substantial systematic temporal irregularities in the $A_j(t_i)$ data base.

Sample Identity	A_0 (Bq · g^{-1})[a]	T (days)[a]	$\hat{s}_{fit}$[b]
P3	10.6728 ± 0.0056	3.82226 ± 0.00189	1.46
P4	10.6794 ± 0.0061	3.81995 ± 0.00205	1.59
P5	10.6770 ± 0.0064	3.82066 ± 0.00217	1.68
P7	10.6676 ± 0.0064	3.82591 ± 0.00218	1.68
P8	10.6753 ± 0.0067	3.82116 ± 0.00228	1.76
P9	10.6496 ± 0.0063	3.82519 ± 0.00215	1.66
unweighted mean	10.6703 ± 0.0109 (± 0.0045)[c]	3.82252 ± 0.00247 (± 0.00101)[c]	1.64
weighted mean	10.6704 ± 0.0025	3.82248 ± 0.00086	—

[a] All uncertainty intervals given in the table correspond to standard deviations unless otherwise noted.
[b] A reduced-χ^2 goodness-of-fit parameter given by the reduced root mean square of the weighted $A_j(t_i)$ residuals, d_i, about the fitted exponential, i.e., $\hat{s}_{fit} = [(\Sigma d_i^2/s_i^2)/\nu]^{1/2}$. ν = n-2 = 118.
[c] Standard deviation of the mean.

Table 1 Fitted parameters for weighted exponential regressions for the six ^{222}Rn samples.

Sample Identity	A_0 (Bq · g^{-1})[a]	T (days)[a]	$\hat{s}_{fit}$[b]
P3	10.6756 ± 0.0049	3.82078 ± 0.00259	0.0157
P4	10.6859 ± 0.0054	3.81632 ± 0.00281	0.0171
P5	10.6735 ± 0.0058	3.82278 ± 0.00305	0.0185
P7	10.6627 ± 0.0058	3.82881 ± 0.00307	0.0186
P8	10.6810 ± 0.0059	3.81806 ± 0.00310	0.0188
P9	10.6458 ± 0.0059	3.82740 ± 0.00308	0.0186
unweighted mean	10.6708 ± 0.0145 (± 0.0059)[c]	3.82252 ± 0.00499 (± 0.00204)[c]	0.0179
weighted mean	10.6714 ± 0.0023	3.82209 ± 0.00120	—

[a] All uncertainty intervals given in the table correspond to standard deviations unless otherwise noted.
[b] Standard deviation of the estimate, i.e., the reduced root mean square of the $A_j(t_i)$ residuals, d_i, about the fitted exponential, $\hat{s}_{fit} = (\Sigma d_i^2/\nu)^{1/2}$. ν = n-2 = 118.
[c] Standard deviation of the mean.

Table 2 Fitted parameters for unweighted exponential regressions for the six ^{222}Rn samples.

Third, any half-life determination must consider possible effects due to decay during the counting intervals. In this work, the regressions were performed using $A_j(t_i)$ values averaged over approximately equal Δt = 1200 s counting intervals and t_i values that were midpoint times for these intervals. In this regard, one may note that differences in counting rates between that at the midpoint times t_i and either that at the beginning (t_i - 1/2Δt) or end (t_i + 1/2Δt) of any of the counting intervals were less than 0.13 percent since the Δt intervals were substantially less than the ^{222}Rn half-life by roughly a factor of 275. Further, any systematic errors in the regressions due to the effect of decay during counting intervals is minimized when all of the counting intervals are of approximate equal time duration, as in these experiments. Nevertheless, the decay curves were analyzed by weighted nonlinear regressions of an exponential form (see below) that accounts for decay during the counting intervals assuming that each of the counting intervals were randomly distributed about Δt = 1200 ± 1 s.[k] These nonlinear regression results were identical (to six significant figures) to those given in Table 1 for reasons demonstrated below. Compared to the exponential form $A_j(t_i) = A_0 e^{-\lambda t_i}$ that uses

k The relative uncertainties on Δt were actually estimated to be less than ±0.05 percent on consideration of deadtime loss corrections and livetime determinations. These were in turn dependent on the source counting rates, the pulse resolving times for the LS counter, and the uncertainty in the timing oscillator frequency for the counter. The livetime was determined by counting pulses from a gated oscillator, and the combined uncertainty on the livetime is principally dependent on the oscillator precision.

Sample Identity	A_0(unweighted) / A_0(weighted) (percent)	T(unweighted) / T(weighted) (percent)
P3	+0.026	−0.039
P4	+0.061	−0.095
P5	−0.033	+0.055
P7	−0.046	+0.076
P8	+0.053	−0.081
P9	−0.035	+0.058
unweighted mean	$+0.004_6$	-0.004_3
weighted mean	$+0.009_7$	−0.010

Table 3 Percent differences in fitted parameters between the unweighted and weighted exponential regressions.

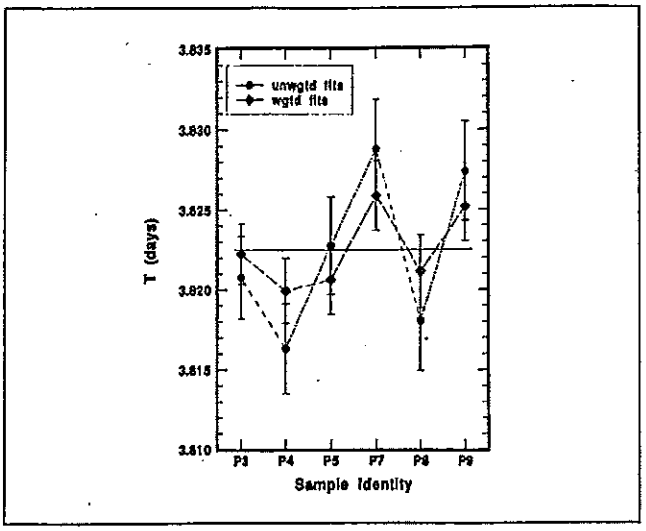

Figure 9 Comparison of the fitted half-life values T from unweighted and weighted exponential regressions for the six ^{222}Rn decay curves.

midpoint t_i values and assumed $A_j(t_i)$ values averaged over Δt, the "true" observed $\langle A_j(t_i)\rangle$ values, obtained by integrating

$\int A_j(t_i)dt / \int dt$ over the time interval Δt, are given by

$$\langle A_j(t_i)\rangle = A_0 \frac{e^{\lambda(t_i-1/2\Delta t)} - e^{\lambda(t_i+1/2\Delta t)}}{\lambda(\Delta t)}$$

$$= A_0\, e^{-\lambda t_i}\left[\frac{e^{\lambda\Delta t}-1}{\lambda\Delta t}\right]$$

where λ in the above expressions is the decay constant equal to $\ln 2/T$. The ratio of $\langle A_j(t_i)\rangle/A_j(t_i)$ is independent of t_i, its magnitude depends merely on the values of Δt and T. For exactly constant Δt, the ratio $\langle A_j(t_i)\rangle/A_j(t_i)$ remains constant. With $\Delta t = 1200 \pm 1$ s and assuming even a ±1 percent relative uncertainty in the ^{222}Rn half-life, the ratio $\langle A_j(t_i)\rangle/A_j(t_i)$ is 1.0012 ± 10^{-5}. For half-life determinations in which Δt varies considerably over the course of the measurements, the magnitude of the possible systematic error in a fitted T depends on the relative magnitudes of T and the various Δt values.[1]

Fourth, weighted nonlinear regressions of other functional forms to the $A_j(t_i)$ data were also performed. This was not done to investigate possible deviations from the exponential radioactive decay law, as in the study of Butt and Wilson,[19] but rather was done to evaluate possible systematic irregularities in the data themselves. Seven other three-parameter functions in comparison to the pure exponential were tested. In all cases, the functions contained A_0 and T, and one additional arbitrary parameter k. Table 4 summarizes the regression results obtained with the data from one of the samples (P3). The first case merely adds a constant term k, which one would expect to be zero since the regression procedure χ^2 minimizes a constant error term. In this case, $k = 0.0039 \pm 0.0037$ Bq · g^{-1} (which is virtually zero). The fitted T changed by roughly -0.14 percent accompanied by a considerably poorer uncertainty in T (changing from ±0.0019 d for the pure exponential case to ±0.0053 d) indicating the superiority of the exponential form over this function. The next case tested for a bias proportional to t_i using a proportionality constant k. Its results were almost analogous to that of the first case: a fitted k of near zero ($k = 0.002 \pm 0.002$ Bq · g^{-1} d^{-1}); fitted values of A_0 and T that barely changed; and less precision in these A_0 and T parameters. The next tested function added a second exponential term as would be the case for a multi-component decay curve due to the presence of a radioactive impurity. The regression results with this function gave fitted A_0 and T values and respective uncertainties that were unchanged to five significant figures. The fitted parameter $k = 0.01$ d, comparable to the half-life for

1 The failure to explicitly address whether corrections were made to account for possible systematic errors due to decay during the counting intervals is one of the principal shortcomings of the reportedly precise determination of Butt and Wilson.[19] Initial counting rates for both of their ^{222}Rn sources exceeded $2(10^5)$ counts per minute (cpm) and presumably would have required counting intervals of perhaps 10 m < Δt < 100 m to achieve reasonable statistical precision. Initial Δt durations were unreported but the authors implied that they were short compared to subsequent measurements. Decay of the two sources were followed to counting rates of less than 1 cpm, and the authors stated that "counting rates of up to one day were required." Their Δt counting rate intervals may have ranged over more than, or at least approached, an order of magnitude. Consider: the ratio $\langle A_j(t_i)\rangle/A_j(t_i)$ equals 1.004 with $\Delta t = 1$ h, and it equals 1.094 with $\Delta t = 1$ d.

Fitted form for $A(t_i)$	Fitted parameters: A_0 (Bq · g^{-1})	T (days)	k
$A_0e^{-(\ln 2/T)t_i}$	10.6727 ± 0.0056	3.8223 ± 0.0019	—
$A_0e^{-(\ln 2/T)t_i} + k$	10.6731 ± 0.0056	3.8171 ± 0.0053	0.0039 ± 0.0037 Bq · g^{-1}
$A_0e^{-(\ln 2/T)t_i} + k\,t_i$	10.6764 ± 0.0067	3.8188 ± 0.0040	0.0002 ± 0.0002 Bq · g^{-1} · d^{-1}
$A_0e^{-(\ln 2/T)t_i} + e^{-(\ln 2/k)t_i}$	10.6728 ± 0.0058	3.8222 ± 0.0019	0.01 ± 0.1 d
$A_0e^{-(\ln 2/T)t_i(1+kt_i)}$	10.6789 ± 0.0077	3.8155 ± 0.0061	-0.00014 ± 0.00012 d^{-1}
$A_0e^{-[(\ln 2/T)t_i + (\ln 2/k)t_i]}$	Regression did not converge		
$A_0e^{-[\ln 2/(T+k)]t_i}$	10.6728 ± 0.0056	3.77 ± 0.50	0.06 ± 0.05 d
$A_0e^{-[(\ln 2/T)t_i+k]}$	10.1 ± 6.8	3.8223 ± 0.0021	0.06 ± 0.7

Table 4 Results of weighted nonlinear regressions of other functional forms to the $A(t_i)$ data for sample P3.

an impurity, had a relative uncertainty of ±1000 percent! The next four cases consisted of increasingly complex exponential forms that considered possible slow temporal variations as well as various proportional and non-proportional bias effects in the exponential term. The most common feature of the results for these later four cases is the explosive increases in the uncertainties in the fitted parameters. It is indicative of the inferiority of these empirical fitting functions in describing the decay data. For every one of the above seven functional forms, F tests comparing the χ^2 for that case to the χ^2 for the pure exponential indicated that use of the additional parameter k did not improve the value of the reduced χ^2. Hence, the incorporation of the k parameter was not justified, and that the three-parameter function was inferior in every case to the two-parameter pure exponential form.

A mean half-life value

Unweighted and weighted means for the six half-life determinations from both the weighted exponential regressions and unweighted exponential regressions are provided in Tables 1 and 2. Selection of an appropriate mean T value is rather effortless. Examination of the four mean T values indicates agreement among them to four significant figures (T = 3.822) with a fifth significant figure ranging from a value of 1 to 5. However, selection of an appropriate corresponding uncertainty for this mean is a more daunting enterprise.

Uncertainty analyses and concluding commentaries

Following conventional practices, which are often acceptable to data compilers, and in which only the regression uncertainties in T are considered, a half-life of T = 3.82248 ± 0.00086 d, based on the weighted mean of the six weighted regression results (Table 1), could be reported. The precision in this value may be compared, or contrasted if you will, with the other reportedly precise determinations of the ^{222}Rn half-life. The oft-cited mean result of Butt and Wilson,[19] T = 3.82351 ± 0.00034 d, is based on NaI(Tl) measurements over nearly 20 half-lives on two sources with the decay curves analyzed by weighted exponential regressions. The reported uncertainty represents only that obtained from the regressions. No other possible uncertainty contributions were addressed by the authors. The almost equally precise, but frequently ignored result of Marin,[16] T = 3.82329 ± 0.00043 d,[m] is based on measurements over 5 half-lives on three sources. The three half-life values were computed from unweighted linear least squares fits of logarithmic transformations of the measured ionization currents. Marin performed instrument linearity tests, concluded that the "nonlinearity, if any, is extremely small," and based the uncertainty estimate on only that obtained from the linear regressions.

The previously cited IUPAC critical compilation[27] rationalized that no half-life value with an "accuracy" of better than 0.1 percent could be recommended since that would require that all potential experimental uncertainty contributions to a precision level of 10^{-4} to 10^{-5} be "investigated, documented and their effect on the result taken into account." The IUPAC authors further contended that "an experiment in which such a thorough study has been both performed and documented has yet to be reported..."[27]

With the above considerations (and challenge) in mind, the following uncertainty analyses are made and offered.

A realistic uncertainty assessment

The uncertainty in this ^{222}Rn half-life determination, and in other determinations, can be considered to be comprised of the following possible component uncertainties: (1) inherent statistical (Poisson) counting imprecision; (2) timing; (3) background corrections; (4) losses of radon or detection efficiency geometry effects due to positional changes or redistributions of the radon in the counting sources; (5) activity-level-dependent instrument nonlinearities, such as uncorrected dead-time effects; (6) temporal variations in the instrument response or

m Marin's[16] originally reported mean value was 0.011 percent less than the value given here. Examination of the original results for the three decay curves indicates that a calculational or transcription error was made in deriving the weighted mean. None of the data compilations cited previously,[21-23] including the careful IUPAC study,[27] appear to have noticed this error. The originally reported precision estimator was also given by Marin in terms of a "probable error" and was converted to the standard deviation given here. These recalculations and conversions are treated more fully in Collé and Maroufi-Collé.[1]

detection efficiency over the course of the measurements; (7) other source-dependent temporal changes, such as that due to LS sample instabilities; and lastly, (8) regression precision and uncertainty in the fitted T parameter. Others, not applicable here but possibly present in other experimental determinations, might include the inappropriateness of the error measurement model (e.g., the use of linear regressions on logarithmically transformed data) or neglect to account for systematic effects (e.g., that due to decay during counting intervals of differing duration).

Each of the above uncertainty components were addressed in one form or another previously. Component (1), the statistical counting precision, was used to provide the regression weights and is embodied in the weighted regression residuals as reflected in the component (8) uncertainty term. The timing uncertainties (2) were considered and demonstrated to be negligible. Independent background corrections (3) were made for each of the 120 measurement cycles and were further accounted for by normalizations to the responses from a common ^{226}Ra reference source used for each measurement. The uncertainty in the background corrections would also be at least partially incorporated in the component (8) uncertainty. Leakage losses of radon (4) were obviated by the use of flame-sealed glass LS vials. Effects due to changing distributions of radon or its decay products in the sources (4) would be manifest in the component (7) uncertainty. Component (5) due to instrument nonlinearities was estimated, primarily from the data of Figure 5, to have a relative uncertainty of about ±0.035 percent. Other temporal variabilities due to both that of the instrument (6) and sources (7) over the course of the measurements were estimated from the data of Figures 4 and 3 to have a relative uncertainty of about ±0.05 percent. The relative uncertainties in the fitted half-life parameters from the regressions were summarized in Tables 1 and 2 and ranged from ±0.068 percent to ±0.081 percent for the unweighted regressions and from ±0.049 percent to ±0.060 percent for the weighted regressions. Other possible unjustified or insupportable assumptions, which are sometimes obviously present in other experimental studies, were also addressed previously.

With the above, a realistic combined uncertainty for any one of the four aforementioned mean T values may be derived from three distinctly different approaches. The relative uncertainty in the mean T (s_T) may be taken to be a combination of the relative uncertainty in the instrument linearity (s_l = ±0.03 to ±0.04 percent), the relative uncertainty in other temporal variations for the instrument and sources (s_t = ±0.05 percent), and the relative uncertainty in the fitted T values (s_f).

For Approach I, using the results for the weighted regressions, the uncertainties s_l and s_t are directly combined with any one of several precision estimators for s_f:

Ia. With s_f obtained from the uncertainty in the fitted T from any one regression, s_f = ±0.049 percent to ±0.060 percent, and $s_T = (s_l^2 + s_t^2 + s_f^2/6)^{1/2}$ = ±0.062 percent to ±0.069 percent where s_f was reduced by $\sqrt{6}$ for the six determinations.

Ib. With s_f obtained from the weighted mean of the six fitted T values, s_f = ±0.022 percent, and $s_T = (s_l^2 + s_t^2 + s_f^2)^{1/2}$ = ±0.062 percent to ±0.068 percent.

Ic. With s_f obtained from the unweighted mean of the six fitted T values, s_f = ±0.026 percent, and $s_T = (s_l^2 + s_t^2 + s_f^2)^{1/2}$ = ±0.064 percent to ±0.069 percent where s_f is the standard deviation of the mean.

The results for the unweighted regressions may be treated analogously:

Id. With s_f obtained from any one regression, s_f = ±0.068 percent to ±0.081 percent, and s_T = ±0.065 percent to ±0.072 percent.

Ie. With s_f obtained from the weighted mean, s_f = ±0.031 percent, and s_T = ±0.066 percent to ±0.071 percent.

If. With s_f obtained from the unweighted mean, s_f = ±0.053 percent, and s_T = ±0.079 percent to ±0.083 percent.

For Approach II, the uncertainties s_l and s_t are reduced by $\sqrt{6}$ prior to propagation to account for their independent presence in each of the six T determinations. Using the same s_f estimators given above, a comparable set of s_T values are derived by this approach, first for the weighted regression results:

IIa. $s_T = (s_l^2 + s_t^2 + s_f^2/6)^{1/2}$ = ±0.031 percent to ±0.036 percent.

IIb. $s_T = [(s_l^2 + s_t^2)/6 + s_f^2]^{1/2}$ = ±0.032 percent to ±0.034 percent.

IIc. $s_T = [(s_l^2 + s_t^2)/6 + s_f^2]^{1/2}$ = ±0.035 percent to ±0.037 percent.

And then for the unweighted regression results:

IId. s_T = ±0.037 percent to ±0.038 percent.

IIe. s_T = ±0.039 percent to ±0.041 percent.

IIf. s_T = ±0.058 percent to ±0.059 percent.

For Approach III, the uncertainties in s_l and s_t were incorporated into regression weighting factors by combining them in a root sum square with the statistical counting precisions described previously. The resulting weighting factors (in terms of the reciprocal total variance) on each $A_j(t_i)$ value were then used to perform weighted exponential regressions whose results could be compared to that obtained from the previous weighted regressions. The relative uncertainties in s_f from these regressions ranged from ±0.076 percent to ±0.102 percent for the six decay curves. Therefore, the result of Approach III may be given as:

III. With s_f = ±0.0.76 to ±0.102 percent, $s_T = s_f/\sqrt{6}$ = ±0.031 percent to ±0.042 percent.

Any one of the three approaches are statistically justifiable. They merely differ in terms of their underlying error model for the measurement process, i.e., in terms of how the estimated s_l and s_t uncertainty terms enter into it. The three approaches result in a relative combined standard uncertainty ranging from ±0.062 percent to ±0.083 percent with a median and mean of s_T = ±0.069 percent for Approach I; from ±0.031 percent to ±0.059 percent with median of ±0.037 percent and mean of s_T = ±0.040 percent for Approach II; and from ±0.031 percent to ±0.042 percent with a median and mean of s_T = ±0.036 percent for Approach III. Combining the various four mean T values (Tables 1 and 2) with all appropriate combinations of

the above s_T estimates results in central values of $T = 3.8224$ d and $s_T = \pm 0.048$ percent.

A parting shot

An adopted value of $T = 3.8224 \pm 0.0018$ d is therefore justified, but is offered here with some trepidation in fear that it will be subsequently and blindly compiled with other determinations whose uncertainties are superficially treated and underestimated.

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