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ScienceDirect
Applied Radiation and Isotopes 65 (2007) 728–730
Applied Radiation and Isotopes
www.elsevier.com/locate/apradiso

Technical note

A note on the half-life of ^{209}Po

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Received 10 October 2006; accepted 11 October 2006

Abstract

The widely adopted value of (102 ± 5) a for the ^{209}Po half-life, which is based on a single determination reported in 1956, appears to be in error by a large factor. Decay data from two separate primary standardizations of a ^{209}Po solution standard, conducted approximately 12 years apart, are inconsistent with the adopted value and its assigned uncertainty. An estimated half-life, larger than the adopted value by about 25%, is more consistent with the standardization data. A longer half-life is also supported by measurements on a recently standardized ^{210}Pb solution standard.
Published by Elsevier Ltd.

Keywords: Half-life; Polonium-209; Radioactivity

The half-life for the radioactive decay of ^{209}Po is tabulated as (102 ± 5) years in essentially every nuclear structure and decay data compilation (cf., Martin, 1991; Firestone et al., 1996; LBNL, 2004; ENSDF, 2006). Surprisingly, despite its importance, there has been only one known determination of this half-life, and it serves as the sole basis for the presently and widely adopted value. This laboratory recently developed evidence that strongly suggests that this adopted half-life is in error by as much as 25%. In as much as long-lived standards of ^{209}Po are used throughout the worldwide environmental radiation community as the principal radiochemical tracer for the measurement of polonium isotopes, a reasonably accurate value of its half-life is critically important.

Andre et al. (1956), as a small part of a much larger study on the proton reaction cross sections of ^{209}Bi, reported a ^{209}Po half-life of 103 a, which was obtained relative to an assumed half-life of (2.93 ± 0.03) a for ^{208}Po. The determination was based on mass spectrometric measurements of the ^{209}Po/^{208}Po atom ratios and on relative ^{209}Po/^{208}Po activity ratios in an irradiated polonium sample. The two ratios were reported to have relative uncertainties of 1.14% and 5%, respectively, although no experimental details on the methods used are available since both ratios were obtained from "private communications" (Andre et al., 1956).

There are no other known determinations or reports on the ^{209}Po half-life.

Using the presently accepted value of (2.898 ± 0.002) a for the ^{208}Po half-life (ENSDF, 2006), the ^{209}Po half-life reported by Andre et al. (1956) becomes 102 a, as presently given in most data evaluations (cf., Martin, 1991). The presently assigned uncertainty of 4.9% is undoubtedly optimistic.

The present work is based upon measurements and analyses of a standardized ^{209}Po solution that has been tracked since its preparation in March 1993 to September 2006.

This ^{209}Po solution standard was originally prepared, calibrated, and disseminated by the National Institute of Standards and Technology (NIST) as Standard Reference Material SRM 4326 (NIST, 1995). This work has been described in detail by Collé, et al. (1995). The solution was certified to have a massic alpha particle emission rate of $R_{1994} = (85.42 \pm 0.18)\,s^{-1}\,g^{-1}$ as of 1200 EST 15 March 1994. The uncertainty specified here is only the combined standard uncertainty u (used to approximate the square root of the variance u^2) which was calculated according to ISO (1995) and NIST (Taylor and Kuyatt, 1994)

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[1]The National Institute of Standards and Technology is an agency of the Technology Administration of the US Department of Commerce.

0969-8043/$ - see front matter Published by Elsevier Ltd.
doi:10.1016/j.apradiso.2006.10.007

guidelines; whereas that reported within the SRM 4326 Certificate is an expanded uncertainty obtained by multiplying u by a coverage factor of $k = 2$ and was chosen to obtain an approximate 95% level of confidence. The standardization was based on $4\pi\alpha$ liquid scintillation (LS) spectrometry using two different LS counting systems under a wide variety of cocktail composition and measurement conditions, with confirmatory measurements by $2\pi\alpha$ gas-flow proportional counting. The LS rates were appropriately corrected and extrapolated to account for the response due to a ^{208}Po impurity, for the ^{209}Po electron capture branch, and for the very low-energy 2.3-keV state in ^{205}Pb—fed by the alpha decay of ^{209}Po decay—(Collé et al., 1994). The corrected mean massic alpha emission rate was derived from 536 independent determinations of the LS counting rate, using 27 counting sources (of widely varying chemical quenching and sample composition) that were in turn obtained from seven randomly selected sealed ampoules of the ^{209}Po solution. The determinations were performed over a 1-year time interval. Refer to Collé et al. (1995) for the methodology and for the specification of the various measurement conditions.

As part of a re-certification of SRM 4326, the ^{209}Po solution was re-standardized in late 2005 using procedures that were as nearly identical as possible to those performed for the original standardization. The LS measurements were performed using three different systems: Packard Tri-Carb A2500TR (Perkin-Elmer, Wesley, MA), Beckman LS 6500 (Beckman Coulter, Fullerton, CA); and Wallac 1414 Winspectral (Perkin-Elmer, Wesley, MA).[2] The first of these was also used in the 1994 measurements. All three spectrometers have different operating characteristics (e.g., low-energy threshold, deadtime, linear vs. logarithmic amplification and ADC, etc.), which helps to ensure that any standardization results are independent of the instrumental conditions. The LS cocktails in both glass and plastic vials were also prepared using the same alkylated-benzene-based scintillant ("Ready Safe", Beckman Coulter, Fullerton, CA) and with the same cocktail compositions (water fractions, volume, etc.). The corrected mean massic alpha emission rate, derived from 25 independent determinations of the LS counting rate with 10 counting sources, was found to be $R_{2005} = (80.20 \pm 0.22)\,s^{-1}\,g^{-1}$ as of reference time of 1200 EST 15 November 2005. The uncertainty interval given here corresponds to a combined standard uncertainty whose assessment and components are essentially identical to those given previously (Collé, 1995; NIST, 1995). The within-measurement precision, in terms of the relative standard deviation of the mean, for a single set of 5 sources measured five times within a given LS counter was in the range of 0.02–0.04% in all cases. The between-measurement standard deviation for a single set of samples measured five times across the three spectrometers was also typically 0.02%.

The decay-corrected value of R_{1994} as of 15 November 2005, using the adopted half-life of (102 ± 5) a, is $(78.91 \pm 0.34)\,s^{-1}\,g^{-1}$, which differs from R_{2005} by 1.6%. The difference, although small, is statistically significant and cannot be accounted for by the known metrology. Excepting for a possibility that the half-life is in error, the discrepancy is inexplicable. Polonium solution instability can be discounted because studies of similar polonium solutions exhibited long-term stability (Collé, 1993) and because the R_{2005} result compared to the decay-corrected R_{1994} suggests an excess of ^{209}Po, not a loss. The apparent half-life obtained with the two R_{1994} and R_{2005} values is $T_{1/2} = 128.3$ a with a propagated uncertainty of 5.5% (or ± 7 a). This uncertainty may be an underestimate because of other unknown but possible effects and temporal dependencies between the two measurements. This value makes the data consistent, but is not intended to represent a new half-life determination. Fig. 1 illustrates graphically the discrepant inconsistency that exists between the decay-corrected standardization that was performed in 1994 and the re-standardization in 2005. Although seeming small,

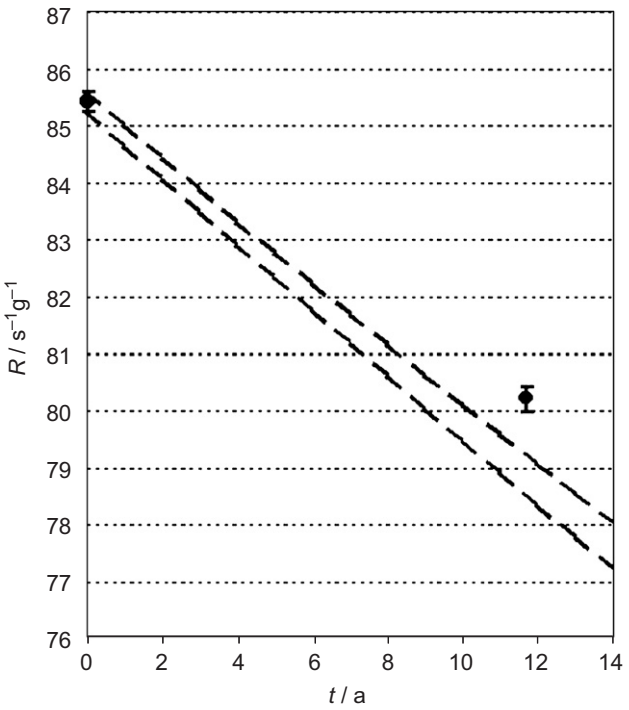

Fig. 1. The massic alpha particle emission rate R (in units of alphas per second per gram) for the standardized ^{209}Po solution standard as a function of time t (in years). The two data points are from the primary standardizations conducted 11.7 years apart and exhibit an apparent half-life of about 128 years. The error bars show the combined standard uncertainty intervals on the two measurements. The broken curves correspond to the upper and lower uncertainty bounds (for the combined standard uncertainty) on the decay of the 1994 value of R using a half-life of (102 ± 5) a. Refer to the text for additional detail.

[2]Certain commercial equipment, instruments, and materials are identified in this paper to foster understanding. Such identification does not imply recommendation or endorsement by NIST, nor does it imply that the materials are the best available for the purpose.

the discrepancy manifests into a huge change in the half-life due to the short time interval for decay.

In early 2006, Schultz (2006) conducted several experiments that utilized SRM 4326 solution as a polonium radiochemical tracer for the measurement of ^{210}Po. These measurements were used to support research on developing new alpha-emitting radiotherapy drugs. The methodology employed was similar to that previously described (Schultz et al., 2006). On learning of the present concerns of the authors, Schultz indicated that he too had suspected that the decay-corrected ^{209}Po content of SRM 4326 appeared to be too low, discerning that the adopted ^{209}Po half-life was inconsistent.

More recently, the SRM 4326 solution was used as a tracer for ^{210}Po in another series of experiments at this laboratory. To confirm a primary standardization of ^{210}Pb for new SRM 4337 by application of an alternative measurement method, ^{210}Po, in radioactive equilibrium, was separated from the ^{210}Pb stock solution by spontaneous electrodeposition on silver (Figgins, 1961) and assayed by $2\pi\alpha$ spectrometry with a surface barrier detector. The chemical yield of the separation was monitored with the ^{209}Po SRM 4326 solution. Employing this type of independent confirmatory measurement, even if it is substantially less accurate, is a vital aspect of our standardization work at NIST. Its intent is to ensure that the standardization result is confirmed by supplementary measurements that are completely independent of the primary method. The primary ^{210}Pb standardization was based on LS efficiency tracing with a ^{3}H standard [cf. (Collé, 2000) and references therein for details of the tracing method as applied to other beta emitting nuclides]. The standardized massic activity of ^{210}Pb for the SRM 4337 solution is $A = 9.037\,\mathrm{kBq\,g^{-1}}$ as of a reference time of 1200 EST 15 June 2006. Although the final uncertainty assessment on A is incomplete (awaiting additional confirmatory measurements), the relative combined standard uncertainty (for $k = 1$) is presently estimated to be of the order of $\pm 1\%$. The confirmatory measurement, based on tracing with ^{209}Po and using the adopted (102 ± 5) a half-life, yielded a result of $A_{c(102)} = 8.77\,\mathrm{kBq\,g^{-1}}$, which differs from A by about 3%. The result for $A_{c(102)}$ was based on duplicate samples (for the Po separation and α counting), whose individual results agreed to be better than 0.2%. These A and $A_{c(102)}$ results are not considered to be in agreement, and again support the apparent inconsistency in the adopted ^{209}Po half-life. Upon re-analysis of the data on assuming the longer and previously estimated ^{209}Po half-life of 128 a, the confirmation value was found to be $A_{c(128)} = 8.92\,\mathrm{kBq\,g^{-1}}$. This value differs from A by about 1.3% and can be considered to be in agreement for confirmatory purposes.

The conclusions of this work are threefold: (1) the widely adopted half-life value of ^{209}Po, based on a single report in 1956, cannot withstand critical scrutiny, (2) the evidence given here strongly supports that this adopted half-life may be underestimated and in error by as much as 25%, and (3) that a new precise determination of this half-life is urgently needed to support the continued use of this long-lived polonium isotope standard for radiochemical tracing. It is hoped that this work will stimulate and encourage other workers to do a more definitive determination.

Our former colleague, Dr. Michael Schultz, is appreciated for relating the results of his tracing experiments. Our colleague, Dr. Ryan Fitzgerald, is heartily thanked for his thoughtful contributions and discussions on this work.

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