Journal of Environmental Radioactivity xxx (2014) 1–3

Contents lists available at ScienceDirect

Journal of Environmental Radioactivity

journal homepage: www.elsevier.com/locate/jenvrad

Lifetimes of ^{214}Po and ^{212}Po measured with Counting Test Facility at Gran Sasso National Laboratory

L. Miramonti [a], G. Bellini [a], J. Benziger [b], D. Bick [c], G. Bonfini [d], D. Bravo [e], M. Buizza Avanzini [a], B. Caccianiga [a], L. Cadonati [f], F. Calaprice [g], C. Carraro [h], P. Cavalcante [d], A. Chavarria [g], V. Chubakov [i], D. D'Angelo [a], S. Davini [h], A. Derbin [j], A. Etenko [k], K. Fomenko [d,l], D. Franco [m], C. Galbiati [g], S. Gazzana [d], C. Ghiano [d], M. Giammarchi [a], M. Göger-Neff [n], A. Goretti [g], L. Grandi [g], E. Guardincerri [h], S. Hardy [e], Aldo Ianni [d], Andrea Ianni [g], V. Kobychev [o], D. Korablev [l], G. Korga [d], Y. Koshio [d], D. Kryn [m], M. Laubenstein [d], T. Lewke [n], M. Lissia [p], E. Litvinovich [k], B. Loer [g], F. Lombardi [d], P. Lombardi [a], L. Ludhova [a], I. Machulin [k], S. Manecki [e], W. Maneschg [q], F. Mantovani [i,*], G. Manuzio [h], Q. Meindl [n], E. Meroni [a], M. Misiaszek [r], D. Montanari [d,g], P. Mosteiro [g], V. Muratova [j], S. Nisi [d], L. Oberauer [n], M. Obolensky [m], F. Ortica [s], K. Otis [f], M. Pallavicini [h], L. Papp [d,e], L. Perasso [a], S. Perasso [h], A. Pocar [f], G. Ranucci [a], A. Razeto [d], A. Re [a], A. Romani [s], N. Rossi [d], A. Sabelnikov [k], R. Saldanha [g], C. Salvo [h], S. Schönert [n,q], H. Simgen [q], M. Skorokhvatov [k], O. Smirnov [l], A. Sotnikov [l], S. Sukhotin [k], Y. Suvorov [d,k], R. Tartaglia [d], G. Testera [h], D. Vignaud [m], R.B. Vogelaar [e], F. von Feilitzsch [n], J. Winter [n], M. Wojcik [r], A. Wright [g], M. Wurm [n], G. Xhixha [i], J. Xu [g], O. Zaimidoroga [l], S. Zavatarelli [h], G. Zuzel [q,r], Borexino Collaboration

[a] Dipartimento di Fisica, Università degli Studi e INFN, Milano 20133, Italy
[b] Chemical Engineering Department, Princeton University, Princeton, NJ 08544, USA
[c] Institut für Experimentalphysik, Universität Hamburg, Germany
[d] INFN Laboratori Nazionali del Gran Sasso, Assergi 67010, Italy
[e] Physics Department, Virginia Polytechnic Institute and State University, Blacksburg, VA 24061, USA
[f] Physics Department, University of Massachusetts, Amherst, MA 01003, USA
[g] Physics Department, Princeton University, Princeton, NJ 08544, USA
[h] Dipartimento di Fisica, Università e INFN, Genova 16146, Italy
[i] Dipartimento di Fisica, Università di Ferrara and INFN Ferrara, 44100 Ferrara, Italy
[j] St. Petersburg Nuclear Physics Institute, Gatchina 188350, Russia
[k] NRC Kurchatov Institute, Moscow 123182, Russia
[l] Joint Institute for Nuclear Research, Dubna 141980, Russia
[m] Laboratoire AstroParticule et Cosmologie, 75231 Paris Cedex 13, France
[n] Physik Department, Technische Universität München, Garching 85747, Germany
[o] Kiev Institute for Nuclear Research, Kiev 06380, Ukraine
[p] Istituto Nazionale di Fisica Nucleare, Sezione di Cagliari, I-09042 Monserrato, Italy
[q] Max-Plank-Institut für Kernphysik, Heidelberg 69029, Germany
[r] M. Smoluchowski Institute of Physics, Jagellonian University, Krakow, 30059, Poland
[s] Dipartimento di Chimica, Università e INFN, Perugia 06123, Italy

ARTICLE INFO

Article history:
Received 9 January 2013
Received in revised form
16 December 2013

ABSTRACT

The decays of ^{214}Po into ^{210}Pb and of ^{212}Po into ^{208}Pb tagged by the previous decays from ^{214}Bi and ^{212}Bi have been studied inserting quartz vials inside the Counting Test Facility (CTF) at the underground laboratory in Gran Sasso (LNGS). We find that the mean lifetime of ^{214}Po is $(236.00 \pm 0.42(\mathrm{stat}) \pm 0.15(\mathrm{syst}))$ µs and that of ^{212}Po is $(425.1 \pm 0.9(\mathrm{stat}) \pm 1.2(\mathrm{syst}))$ ns. Our results are

* Corresponding author. Tel.: +39 3200864636; fax: +39 532974210.
E-mail address: mantovani@fe.infn.it (F. Mantovani).

http://dx.doi.org/10.1016/j.jenvrad.2014.02.025
0265-931X/

Accepted 27 February 2014
Available online xxx

compatible with previous measurements, have a much better signal to background ratio, and reduce the overall uncertainties.

Keywords:
Po-214
Po-212
Alfa-decay lifetime
Natural radioactivity
Bi–Po coincidence

1. Introduction

Polonium isotopes ^{214}Po and ^{212}Po are part of ^{238}U and ^{232}Th decay chain, respectively: these radionuclides ^{214}Po and ^{212}Po have the shortest lifetimes and emit the most energetic particles of the respective chain ($E = 7.833$ MeV and $E = 8.954$ MeV). There exist only a few measurements of these two mean lifetimes with precision better than one or two percent. Present situation is summarized in Table 1.

Accurate mean lifetime measurements of ^{214}Po and ^{212}Po are useful not only for testing theoretical nuclear models, but also for tuning the Bi–Po coincidence technique, which is typically employed in underground experiments where very low contamination of U and Th is required. The β-decay of ^{214}Bi in uranium chain and ^{212}Bi in thorium chain followed by a delayed emission from ^{214}Po and ^{212}Po respectively provides an effective tag in measuring natural radioactivity background. Furthermore, mean lifetimes of ^{214}Po and ^{212}Po are of interest for geophysicists (Gat and Assaf, 1968), radio-biologists (Soto et al., 2002) and for nuclear physicists (Alexeyev et al., 2011).

Since we have been studying decay spectra of ^{214}Bi and ^{212}Bi with the purpose of experimentally constraining anti-neutrino spectral shape important for geoneutrino studies (Fiorentini et al., 2010), we have a large statistics of decays of ^{214}Po and ^{212}Po collected with the Counting Test Facility (CTF) (Alimonti et al., 1998), which was operational in the underground I.N.F.N. Gran Sasso National Laboratory. The high purity and low background in CTF allow a favorable signal to background ratio for these measurements. More specifically the ratio of signal to background of the present measurements is more than three orders of magnitude larger than the best existing measurements. In addition only in the case of ^{214}Po there exist a measurement (Von Dardel, 1950) with a number of decays comparable to the present one and that can follow the exponential decay for 7 mean lifetimes. All other measurements for ^{214}Po and all the ones for ^{212}Po have lower statistics and follow the decay for 2–4 lifetimes.

2. Materials and methods

The decay half-lives to be measured are very short: a few hundreds of microseconds that of ^{214}Po and a few hundreds of nanoseconds in the case of ^{212}Po. Therefore, their determination through the delayed coincidence technique is only marginally affected by the extremely low background in CTF. The CTF was equipped with a specific arrangement of the acquisition electronics tailored to register the time elapsed between the prompt β particle from the Bismuth decay and the delayed α particle signal associated with the Polonium decay (Bi–Po coincidences).

The detailed description of the CTF and of its performance have been published in (Alimonti et al., 1998). In this report we recall only the main features of the detector which are important in the later description of the results. The apparatus, installed in the Hall C of the Gran Sasso Underground Laboratory, consisted of an external cylindrical water tank (diameter ≅11 m, high ≅10 m; ≅1000 tons of water) serving as passive shielding against external neutrons and γ's for 4.8 m^3 of liquid organic scintillator (LS) contained in an inner spherical vessel with a diameter of ≅2 m. The yield of emitted photons was ~ 10^4 per MeV of energy deposited, the maximum peak of the fluorescence was located at 365 nm and the principal scintillator decay time was ≅4.5–5.0 ns. The inner vessel containing the liquid scintillator was realized with a nylon membrane (≅500 μm thick), with excellent optical clarity, which allowed the effective transmission of the scintillation light to the 100 Photo Multiplier Tubes (PMTs) forming the optical read-out, anchored on a 7 m diameter support structure inside the water tank. The PMTs were 8 inches ETL 9351 tubes featuring a quantum efficiency of 26% at 420 nm, a limited transit time spread (σ≅1 ns) and a good pulse height resolution for single photoelectron pulses (Peak/Valley ≥2.5). Light concentrators (57 cm long and 50 cm diameter aperture) mounted on the PMTs enhanced the optical coverage to about 20%. The measured light yield corresponds on average to 3.8 photoelectrons (p.e.) per PMT for 1 MeV energy deposit.

A special insertion system installed on the top of the CTF allowed to place suitable radioactive sources inside the detector without contaminating the liquid scintillator. The positioning of the sources was done by means of steerable aluminum rods. The pipe of the insertion system featured an internal diameter of 50 mm. It was possible to place sources containing the isotopes under study (in our case ^{214}Po and ^{212}Po) at the center of the CTF through the

Table 1
Existing measurements of the ^{214}Po and ^{212}Po half-life and compilations compared to the present work.

^{214}Po		^{212}Po	
Reference	Half-life (μs)	Reference	Half-life (ns)
Von Dardel, 1950	163.7 ± 0.2	Bunyan et al., 1949	304 ± 4
Ballini, 1953	158.0 ± 2.0	Flack and Johnson, 1962	305 ± 25
Ovilvie, 1960	159.5 ± 3.0	Astner et al., 1963	305 ± 5
Dobrowolski and Young, 1961	164.3 ± 1.8	McBeth and Winyard, 1972	304 ± 8[a]
Erlik et al., 1971	165.5 ± 3.0	McBeth and Winyard, 1972	300 ± 8[b]
Zhou et al., 1993	160.0 ± 12.0	Sanyal et al., 1975	296 ± 2
Nuclear Data Sheet: Wu, 2009	164.3 ± 2.0[c]	Bohn et al., 1981	309 ± 11
Table de Radionucléides: Christé and Bé (2007)	162.3 ± 1.2[d]	Table of Isotopes: Browne, 2005	299 ± 2[e]
This work (2012)	**163.6 ± 0.3**	Table de Radionucleides: Nichols, 2004	300 ± 2[f]
		This work (2012)	**294.7 ± 1.0**

[a] Curve (A) of Fig. 4 in McBeth and Winyard (1972).
[b] Curve (B) of Fig. 4 in McBeth and Winyard (1972).
[c] Average of Von Darnel (1950), Dobrowolski and Young (1961) and Erlik et al. (1971); used in Table of Isotopes.
[d] Average of all six values, with the original uncertainty of Von Dardel (1950) increased to 1.8 μs.
[e] Average of Bunyan et al. (1949), Astner et al. (1963), McBeth and Winyard (1972) (curve (A) of Fig. 4), Sanyal et al. (1975), and Bohn et al. (1981).
[f] Average of Bunyan et al. (1949), Flack and Johnson (1962), Astner et al. (1963), McBeth and Winyard (1972) (both curves (A) and (B) of Fig. 4), Sanyal et al. (1975) (with the original uncertainty increased to 2.7 ns.), and Bohn et al. (1981).

insertion system. We prepared three different sources (one for ^{214}Po and two for ^{212}Po) contained in quartz vials (transparent to UV light) with an external diameter of 50 mm, the maximum allowed by the insertion system, to contain LS with the necessary isotopes. The ^{214}Po source was realized by spiking the LS with ^{222}Rn obtained from a ^{226}Ra-based "Radon generator" and having the ^{214}Bi–^{214}Po sequence within its decay chain. The first ^{212}Po source was obtained by dissolving a stable hydrophilic complexes of Thorium in the LS contained in quartz vial. A second ^{212}Po source was prepared using a system able to flush ^{220}Rn (half-life of 55.6 s) gas directly into the spherical quartz vial. The preparation of the three sources is described in details in Bellini et al. (2013). The sources were inserted close to the detector's center with a mechanical positioning system, the position of the source was later determined within a few centimeters using the position reconstruction code.

3. Results and discussion

The total live-time of data taking for the ^{214}Po measurement is 10.2 d. For the two ^{212}Po sources, the livetimes are 6.3 d and 15.5 d, respectively. The correspondent overall statistics is $\sim 3.9 \times 10^5$ for ^{214}Po and $\sim (1.1 + 1.7) \times 10^5$ for ^{212}Po candidates. The ^{212}Po candidate samples from the two sources are analyzed independently.

The data analysis is presented in the details in Bellini et al. (2012) and it relies on a triple approach, in order to cross check the results, and to minimize the errors. Results for the mean lifetime are given with statistical and systematic errors. For the sake of comparison with previous measurements and compilations we also present in Table 1 the half-lives with statistical and systematic errors quadratically combined.

For ^{214}Po our result is:

$$\tau = 236.00 \pm 0.42(\mathrm{stat}) \pm 0.15(\mathrm{syst})\ \ \mu\mathrm{s}$$

the corresponding half-life $t_{1/2} = 163.58 \pm 0.31$ μs can be compared with the first six measurements in Refs. shown in Table 1. There exist also two compilations. The one published in Nucl. Data Sheet (Wu, 2009) and used in the Table of Isotopes includes in its average only the three measurements in Refs. (Von Dardel, 1950; Dobrowolski and Young, 1961; Erlik et al., 1971): $t_{1/2} = 164.3 \pm 2.0$ μs, while the compilation in LNE-LNHB/CEA – Table de Radionucléides (Christé and Bé, 2007) takes into account all six measurements: $t_{1/2} = 162.3 \pm 1.2$ μs. Both compilations have rules of avoiding that a single measurement to dominate in the average; this is the reason why the resulting average and error are not close to ones of Von Dardel (1950) measurement, which has by far the smallest error. Our result agrees with the two best existing measurements of Von Dardel (1950) and of Dobrowolski and Young (1961). All other measurements have low statistics, since they come as by products or tests of work aimed to something else.

In the case of ^{212}Po we have results taken with two different sources. The two measurements are statistically compatible and can be combined yielding:

$$\tau = 425.1 \pm 0.9(\mathrm{stat}) \pm 1.2(\mathrm{syst})\ \ \mathrm{ns}$$

The corresponding combined half-life $t_{1/2} = 294.9 \pm 1.0$ ns can be compared with the first seven measurements in Refs. and the two compilations shown in Table 1. The compilation in Nucl. Data Sheet (Browne, 2005) includes in its average only the five measurements in Bunyan et al. (1949), Astner et al. (1963), McBeth and Winyard (1972) (curve (A) of Fig. 4); Sanyal et al., 1975; Bohn et al., 1981): $t_{1/2} = 299 \pm 2$ ns, while the compilation in LNE-LNHB/CEA – Table de Radionucléides (Nichols, 2004) takes into account the six measurements (Bunyan et al., 1949; Flack and Johnson, 1962; Astner et al., 1963; McBeth and Winyard, 1972; Sanyal et al., 1975): $t_{1/2} = 300 \pm 2$ ns. As in the ^{214}Po case, the resulting average and error are not as close to the best values of Sanyal et al. (1975) as they would be, if measurements were weighted with their quoted errors.

4. Conclusions

Thanks to extreme radio-purity of the CTF detector and to a long expertise in source preparation and insertion systems, new accurate measurements of the ^{214}Po and ^{212}Po lifetimes have been provided. We find that the mean lifetime of ^{214}Po is $236.00 \pm 0.42(\mathrm{stat}) \pm 0.15(\mathrm{syst})$ μs and that of ^{212}Po is $425.1 \pm 0.9(\mathrm{stat}) \pm 1.2(\mathrm{syst})$ ns. Our results, obtained from data with signal-to-background ratio larger than 1000, reduce the overall uncertainties and are compatible with previous measurements.

References

Alexeyev, E.N., et al., 2011. Experimental Test of the Time Stability of the Half-life of Alpha-decay Po-214 Nuclei. arXiv:1112.4362 [nucl-ex].

Alimonti, G., et al., 1998. A large-scale low-background liquid scintillation detector: the counting test facility at Gran Sasso. Nucl. Instrum. Meth. A 406, 411–426.

Astner, G., et al., 1963. A Hindered E2 Ground state transition in Po207. Nucl. Phys. 45, 49–53.

Ballini, R., 1953. Contribution a l'etude experimentale de l'isomerie nucleaire. Ann. Phys. 8, 441–488.

Bellini, G., et al., 2012. First evidence of pep solar neutrinos by direct detection in Borexino. Phys. Rev. Lett. 108, 051302.

Bellini, G., et al., 2013. Lifetime Measurements of ^{214}Po and ^{212}Po with the CTF Liquid Scintillator Detector at LNGS. arXiv:1212.1332 [nucl-ex].

Bohn, H., et al., 1981. Spectroscopy of excited states in 212Po,210Pb, and 213At employing 18O induced few-nucleon transfer reactions. Z. Phys. A302, 51–59.

Browne, E., 2005. Nuclear data sheets for A = 212. Nucl. Data Sheets 104, 427–496.

Bunyan, D.E., Lundby, A., Walker, D., 1949. Experiments with the delayed coincidence method, including a search for short-lived nuclear isomers. Proc. Phys. Soc. Lond. 62A, 253–263.

Christé, V., Bé, M.M., 2007. 214Po – Comments on evaluation of decay data. Table de Radionucléides. LNE – LNHB/CEA.

Dobrowolski, T., Young, J., 1961. The determination of the half life of RaC′. Proc. Phys. Soc. Lond. 77, 1219–1220.

Erlik, A., et al., 1971. Lifetime measurements of alpha emitters in the millisecond region. Nucl. Instrum. Methods 92, 45–49.

Fiorentini, G., et al., 2010. Nuclear physics for geo-neutrino studies. Phys. Rev. C 81, 034602.

Flack, F.C., Johnson, J.E., 1962. The Gamma radiation from 212°(ThC′). Proc. Phys. Soc. Lond. 79, 10–13.

Gat, J., Assaf, G., 1968. Atmospheric Bismuth-212: measurement and some geophysical applications. Science 622, 977–979.

McBeth, G.W., Winyard, R.A., 1972. Isotope identification and radio assay by time interval analysis. Int. J. Appl. Radiat. Isot. 23, 527–533.

Nichols, A.L., 2004. 212Po – Comments on evaluation of decay data. Table de Radionucléides. LNE – LNHB/CEA.

Ovilvie, K.W., 1960. The half-value period of radium C′. Proc. Phys. Soc. Lond. 76, 299–301.

Sanyal, S., et al., 1975. Half-life measurement of the 212Po Ground state. Phys. Rev. C12, 318–319.

Soto, J., et al., 2002. A simple method of alpha irradiation for experiments in radiobiology. Nucl. Instrum. Meth. B 197, 310–316.

Von Dardel, G., 1950. A precision determination of the half-life of Radium C′. Phys. Rev. 79, 734–735.

Wu, S.-C., 2009. Nuclear data sheets for A = 214. Nucl. Data Sheets 110, 681–748.

Zhou, J.W., et al., 1993. Advances towards fast thermal detectors of intermediate mass with high resolution and large dynamic range. Nucl. Instrum. Meth. Phys. Res. A335, 443–452.

Please cite this article in press as: Miramonti, L., et al., Lifetimes of ^{214}Po and ^{212}Po measured with Counting Test Facility at Gran Sasso National Laboratory, Journal of Environmental Radioactivity (2014), http://dx.doi.org/10.1016/j.jenvrad.2014.02.025