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Activity standardisation of ^{210}Pb by 4πα liquid scintillation counting method

Miroslav Havelka

Czech Metrology Institute, Radiová 1, 102 00 Prague, Czech Republic

HIGHLIGHTS

- ^{210}Pb was standardised through its decay product ^{210}Po after chemical separation.
- The procedures for ^{210}Pb-^{210}Po and ^{210}Bi-^{210}Po separation were designed.
- Precipitation of $Pb(NO_3)_2$ and KCl from organic solvents solution.
- Result of ^{210}Pb activity with standard uncertainty of 0.45% was obtained.

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ABSTRACT

The activity of ^{210}Pb in a solution with ^{210}Pb-^{210}Po in the radioactive equilibrium was determined through its decay product ^{210}Po by liquid scintillation counting (LSC), which has, after separation, as a pure α emitting nuclide, detection efficiency practically equal to 1. For the separation of ^{210}Po from ^{210}Pb solution, two methods based on precipitation of Pb, and Pb with Bi, leaving Po in the solution, were introduced. The first one was precipitation of Pb in the form of $Pb(NO_3)_2$ from a mixture of acetic acid, toluene, water and HNO_3. The second one was based on co-precipitation of Bi and Pb with KCl from a mixture of organic solvents, where ^{210}Bi with ^{210}Pb was fixed to the precipitate. The relative standard uncertainty of ^{210}Pb activity concentration was estimated to be lower than 0.45%.

1. Introduction

The ^{210}Pb is a member of ^{226}Ra decay chain (Bé at al, 2008) (Fig. 1); it is accumulated in "old" ^{226}Ra solutions. A ^{210}Pb solution usually contains a variable amount of its daughter products, ^{210}Bi and ^{210}Po. If the activity of ^{210}Pb is in the equilibrium with the activity of ^{210}Bi and ^{210}Po ($A(^{210}Pb) = 0.9824A(^{210}Po)$), it is possible to utilise the activity determination of ^{210}Po for the standardisation of the mother nuclide ^{210}Pb. The ^{210}Po, as a pure α particle emitting nuclide, can be easily standardised after its separation from the ^{210}Pb solution by 4πα − liquid scintillation counting (LSC), where the detection efficiency for alpha decay is expected to be 1. For the separation of ^{210}Po from the ^{210}Pb solution, two methods based on precipitation of Pb, eventually Pb with Bi, leaving Po in the solution, were introduced.

The first method utilised precipitation of Pb in the form of $Pb(NO_3)_2$ from acetic acid- toluene-water-HNO_3 mixture, where a major part of ^{210}Po and ^{210}Bi remained in the solution. The second separation method was based on co-precipitation of Bi and Pb with KCl from a mixture of organic solvents, where ^{210}Bi with ^{210}Pb was fixed to precipitate, while ^{210}Po remained in the solution. Practically, the first method was applied to remove ^{210}Pb from the ^{210}Pb-^{210}Bi-^{210}Po solution and then the second method was used for the separation of ^{210}Po from ^{210}Bi. Beside the ^{210}Po-^{210}Bi chemical separation, ^{210}Bi decay in ^{210}Bi-^{210}Po samples can be used as an alternative procedure although it requires longer time to reach the complete ^{210}Bi disintegration. The procedure using both chemical separations (Pb from Bi-Po and Bi from Po) is described

The proposed standardisation method can be seen as complicated in chemical preparation of ^{210}Po LSC sources. However, for the standardisation of ^{210}Pb, both methods, with or without chemical separation of daughters, were used – Woods et al. (2000), (4π(LS)- γ coincidence), Arinc at al., 2011, (Čerenkov counting method), Laureano-Pérez et al. (2007), (4παβ- (LS) CIEMAT/NIST method), Fitzgerald and Schultz (2008), (4πβ- γ anticoincidence counting (in equilibrium with its daughters)).

2. The principle of the method

The crucial part of the ^{210}Pb LSC standardisation through its decay product ^{210}Po is its separation from ^{210}Pb solution. For this purpose, two simple separation methods were newly introduced. They were

E-mail address: mhavelka@cmi.cz.

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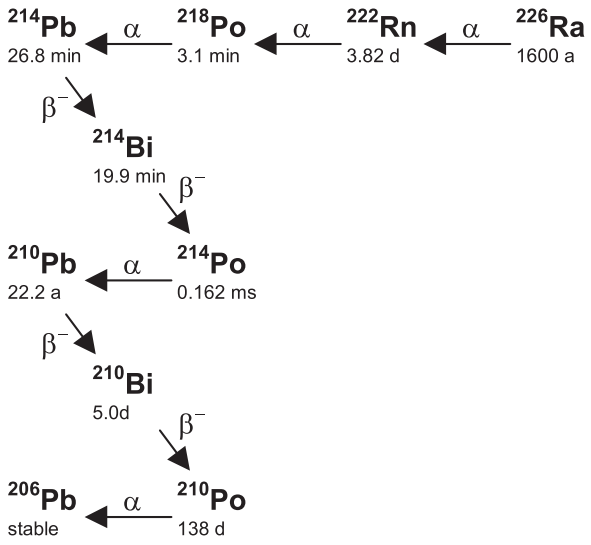

Fig. 1. Decay chain of ^{226}Ra.

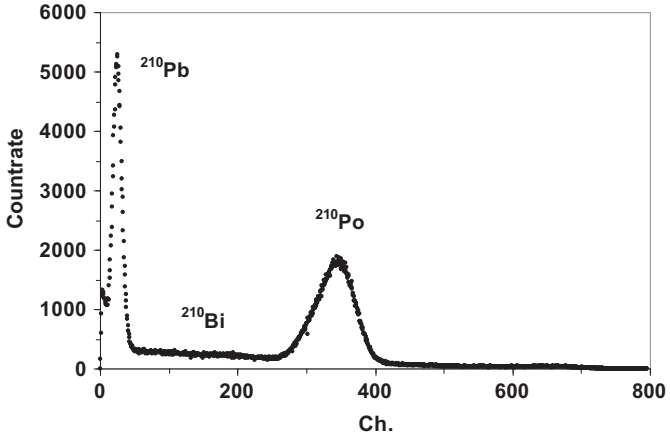

Fig. 2. Typical LSC spectrum of the sample containing approximately the same activity of ^{210}Pb, ^{210}Bi and ^{210}Po prepared into 15 mL Ultima Gold LLT cocktail in 20-mL plastic vial, displayed in linear (Tri-Carb 3100TR) energy scale. The peak at 350 Ch. corresponds to ^{210}Po 5.3 MeV alpha particles. Disintegrations of ^{210}Pb, beta emitter with maximum energy $E_{\beta-}$ = 65.3 keV, are recorded as the peak under channel 50. ^{210}Bi beta particles ($E_{\beta-}$ = 1162 keV) are recorded continuously up to 800 Ch.

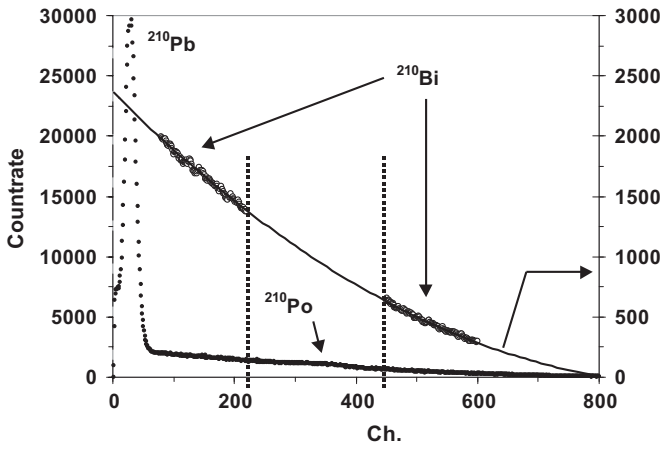

Fig. 3. LSC spectrum of the sample containing ^{210}Pb and ^{210}Bi approximately in radioactive equilibrium and ^{210}Po in low activity concentration (A_{Po} = ~ 2% A_{Pb}). ^{210}Po alpha particles are mainly recorded in the range from 221 to 450 Ch. For ^{210}Po activity determination, the ^{210}Bi beta spectrum in the range of the possible occurrence of ^{210}Po alpha peak (from 221 Ch. to 450 Ch.) was fitted with quadratic function, whereas the values from the spectrum range from 81 Ch. to 220 Ch. and from range from 451 Ch. to 600 Ch. were included into calculation. The fitting curve is displayed in a scale related to the right axis.

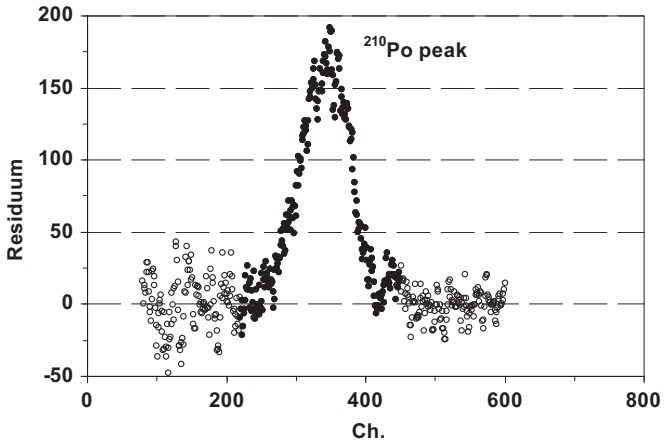

Fig. 4. Residua from quadratic function fitting, the same data as displayed in Fig. 3. The area of the ^{210}Po peak corresponds approximately to 1% of the total count of ^{210}Pb - ^{210}Bi sample.

derived from a very low solubility of inorganic salts $Pb(NO_3)_2$ and KCl in toluene-acetic acid solution. The precipitation of $Pb(NO_3)_2$ from an organic solution (acetic acid) was also used, for example for Th-Ra separation (Havelka, 2016). To reach the adequate separation yield of ^{210}Pb and ^{210}Bi, the precipitation of both $Pb(NO_3)_2$ and KCl was repeated twice, so beside the main Po fraction, two Pb and two Bi fractions were prepared. The separation of ^{210}Po from the ^{210}Pb-^{210}Bi-^{210}Po solution was accompanied by losses, therefore it was necessary to determine the activity of ^{210}Po in all separated fractions. In case of the proposed procedure, it was relatively simple. Both $Pb(NO_3)_2$ and KCl precipitates are very well soluble in water, so it was possible to transfer them easily to LSC samples. The sources originating from Pb precipitates were counted immediately after the chemical separation (was finished) when the concentration of ^{210}Bi was low and did not significantly influence the determination of ^{210}Po activity. The source without ^{210}Pb (prepared from the main Po fraction and ^{210}Bi fractions) were counted several times, at least twice (after partial ^{210}Bi decay), to make the evaluation of ^{210}Po and ^{210}Bi contributions to the LSC count rate from decay curves possible; the values were then used for the ^{210}Po activity calculation. The ^{210}Po activity in Pb fractions was determined from LSC spectra. A typical LSC pulse height spectrum of ^{210}Pb-^{210}Bi-^{210}Po sample is shown in Fig. 2. The spectra were analysed using the fitting of ^{210}Bi contribution in the range of ^{210}Po alpha peak and ^{210}Pb beta "peak" to estimate ^{210}Pb, ^{210}Bi and ^{210}Po concentrations (Fig. 3, Fig. 4). Because a small part of alpha particles was not recorded in the alpha peak area, the value of ^{210}Po activity obtained by this method was underestimated by 2–5%. However, the real samples prepared from Pb fractions contained approximately only ~ 1.3% ^{210}Po and 5% ^{210}Bi, so this procedure did not influence the final result significantly (0.07%).

3. Separation of ^{210}Po from ^{210}Pb, ^{210}Bi

For standardisation, a ^{210}Pb-^{210}Bi-^{210}Po solution (of ^{210}Pb activity concentration ~ 25 kBq g^{-1}, in radioactive equilibrium with ^{210}Bi and ^{210}Po) in HNO_3(65%)-acetic acid mixture (1:4, w/w) was used (Fig. 5). The solution aliquot of ~ 0.2 g (~ 5 kBq) in 6 mL plastic scintillation vial was supplemented with 10 μg Bi^{3+} (in HNO_3–acetic acid solution) and HNO_3–acetic acid mixture (1:4), so the total amount of 65% HNO_3 in the sample was 50 mg. The first $Pb(NO_3)_2$ precipitation was carried out with 0.03 mL of Pb solution in acetic acid (~ 2 mg Pb), 1.45 g of acetic acid-toluene mixture (weight ratio 2:1) and 0.53 g toluene. After