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NEW DEVELOPMENT OF RADON PROGENY MEASUREMENT METHOD BASED ON ALPHA–BETA SPECTROMETRY

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Accurate measurement of radon progeny concentration is important for the dose assessment of radon exposure and the study of radon progeny behaviours. For measuring ^{218}Po, ^{214}Pb and ^{214}Bi concentration as well as EEC with high sensitivity, an alpha–beta spectrometry method was developed and applied in a step-advanced filter radon progeny monitor. The derivation details of this method is given in this paper and the uncertainty is discussed. The comparison experiments are carried out in radon chamber and in field. Results show that the alpha–beta spectrometry method can give ^{218}Po, ^{214}Pb and ^{214}Bi concentration as well as EEC with high sensitivity either for 60 min or for 30 min cycle, which leads to low uncertainty. This method can be used as a reference method for radon chamber and is suitable for portable radon progeny monitor.

INTRODUCTION

Radon exposure is one of the most important contributors of naturally occurring radiation to the public, which may lead to lung cancer(1). Lung cancer is caused by the deposition on respiratory tract of inhaled short-lived radon progeny rather than the radon gas(2). Therefore, precise measurement on radon progeny is quite important for both dose assessment of environmental radon exposure and the research of radon progeny behaviours.

For accurate measurement on ^{218}Po, ^{214}Pb and ^{214}Bi concentrations as well as EEC with high sensitivity, a radon progeny measurement method based on alpha–beta spectrometry was developed and applied in a step-advanced filter radon progeny monitor, which is quite different from traditional working-level monitor use fixed-filter(3). Compared with traditional radon progeny measurement methods such as alpha-counting methods(4, 5) and alpha-spectrum methods(6, 7), the alpha–beta spectrometry method can record the alpha spectrometry and beta spectrometry at the same time with sampling, and the ^{218}Po, ^{214}Pb, ^{214}Bi concentrations as well as EEC can be given analytically using only one spectrometry in each cycle, while three counts is needed for alpha-counting methods and two spectrum is needed for alpha-spectrum methods in each cycle.

In this paper, the assumption and the derivation details of alpha–beta spectrometry method are given. The sensitivity and the uncertainty are also analysed. Results of comparison experiments carried out in both national radon chamber of Chinese National Institute of Metrology and in field are also reported.

MATERIALS AND METHODS

The alpha–beta spectrometry method

For traditional radon progeny measurement methods in portable radon progeny monitor, alpha spectrum was recorded by PIPS detector and multi-channel analyser (MCA). To get each concentration of ^{218}Po, ^{214}Pb and ^{214}Bi, two or more counts/spectrums need to be recorded after sampling in one cycle, just like Thomas method(4) and Kerr method(6). Actually, when we carried out the detection using PIPS detector and MCA, the beta particles emitted from ^{214}Pb/^{214}Bi can also be detected. Traditionally measurement alpha–beta spectrometry for radon progeny could be seen in Figure 1, where the beta particles and alphas from ^{218}Po and ^{214}Po could be seen clearly and the crosstalk between two alphas could be ignored. Adjusting the lower threshold of MCA, the background in region of interesting (ROI) of beta particles could be controlled to a small constant value. So in fact, we could get three counts in β-ROI (100 keV–3 MeV), α_1-ROI1 (3–6.3 MeV) and α_2-ROI2 (6.3–8.0 MeV)(3).

If we could build the relationship between the counts of β-ROI, α_1-ROI1, α_2-ROI2 and ^{218}Po, ^{214}Pb, ^{214}Bi concentrations, then the ^{218}Po, ^{214}Pb, ^{214}Bi concentrations as well as EEC could be given. And if the sampling and detection are carried out at the same time in each cycle, we could get more integral counts which lead to higher sensitivity of radon progeny measurement.

Assure the ^{218}Po, ^{214}Pb, ^{214}Bi concentration is unchanged in one cycle, and the sampling is carried out with stable flowrate. Ignore the difference of detection efficiency of different beta particles from ^{214}Pb/^{214}Bi and of different alpha particle from

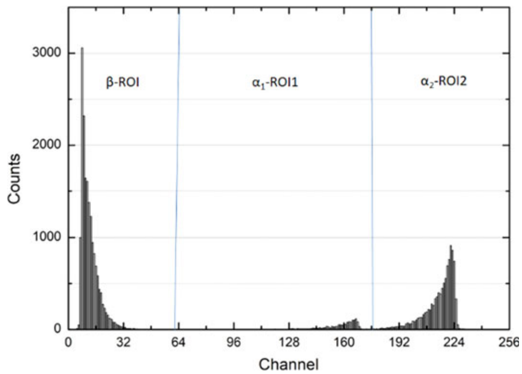

Figure 1. Traditionally alpha–beta spectrometry for radon progeny by PIPS detector (400 mm^2) and MCA (SPECTRA 5011).

$^{218}Po/^{214}Po$. Using the Bateman equation[8], the relationship between the counts of β-ROI, α_1-ROI1, α_2-ROI2 and ^{218}Po, ^{214}Pb, ^{214}Bi concentrations could be given as follows:

$$\begin{aligned} N_{\alpha 1} &= E_\alpha F \varepsilon_f H_{11} C_1 \\ N_{\alpha 2} &= E_\alpha F \varepsilon_f (\lambda_2 \lambda_3 H_{13} C_1 + \lambda_3 H_{23} C_2 + H_{33} C_3) \\ N_\beta &= E_\beta F \varepsilon_f [(\lambda_2 H_{12} + \lambda_2 \lambda_3 H_{13}) C_1 \\ &\quad + (H_{22} + \lambda_3 H_{23}) C_2 + H_{33} C_3] \end{aligned} \tag{1}$$

where N_β, $N_{\alpha 1}$ and $N_{\alpha 2}$ is the integral counts of β-ROI, α_1-ROI1 and α_2-ROI2. C_1, C_2, C_3 is the ^{218}Po, ^{214}Pb, ^{214}Bi concentrations in Bq m^{-3}. E_α is the detection efficiency of alpha particles, E_β is the detection efficiency of beta particles. F is the flowrate in L/min. ε_f is the collection efficiency. λ_i is the decay constant, where $i = 1,2,3$ represent for ^{218}Po, ^{214}Pb, ^{214}Bi. H_{ij} is the integrating function of T, just as follows:

$$\begin{aligned} H_{11} &= \int_0^T \frac{1 - e^{-\lambda_1 \cdot t}}{\lambda_1} dt = \frac{T}{\lambda_1} + \frac{\exp(-\lambda_1 T) - 1}{\lambda_1^2} \\ H_{22} &= \int_0^T \frac{1 - e^{-\lambda_2 \cdot t}}{\lambda_2} dt = \frac{T}{\lambda_2} + \frac{\exp(-\lambda_2 T) - 1}{\lambda_2^2} \\ H_{33} &= \int_0^T \frac{1 - e^{-\lambda_3 \cdot t}}{\lambda_3} dt = \frac{T}{\lambda_3} + \frac{\exp(-\lambda_3 T) - 1}{\lambda_3^2} \\ H_{12} &= \frac{H_{11} - H_{22}}{\lambda_2 - \lambda_1} = \frac{T}{\lambda_1 \lambda_2} + \frac{\exp(-\lambda_1 T) - 1}{\lambda_1^2 (\lambda_2 - \lambda_1)} \\ &\quad + \frac{\exp(-\lambda_2 T) - 1}{\lambda_2^2 (\lambda_1 - \lambda_2)} \\ H_{23} &= \frac{H_{22} - H_{33}}{\lambda_3 - \lambda_2} = \frac{T}{\lambda_2 \lambda_3} + \frac{\exp(-\lambda_2 T) - 1}{\lambda_2^2 (\lambda_3 - \lambda_2)} + \frac{\exp(-\lambda_3 T) - 1}{\lambda_3^2 (\lambda_2 - \lambda_3)} \\ H_{13} &= \frac{H_{12} - H_{23}}{\lambda_3 - \lambda_1} = \frac{T}{\lambda_1 \lambda_2 \lambda_3} + \frac{\exp(-\lambda_1 T) - 1}{\lambda_1^2 (\lambda_3 - \lambda_1)(\lambda_2 - \lambda_1)} \\ &\quad + \frac{\exp(-\lambda_2 T) - 1}{\lambda_2^2 (\lambda_3 - \lambda_2)(\lambda_1 - \lambda_2)} + \frac{\exp(-\lambda_3 T) - 1}{\lambda_3^2 (\lambda_2 - \lambda_3)(\lambda_1 - \lambda_3)} \end{aligned}$$

If cycle time T is determinated, then H_{ij} is constant. From Equation (1), the relationship between the integral counts of β-ROI, α_1-ROI1, α_2-ROI2 and ^{218}Po, ^{214}Pb, ^{214}Bi concentrations could be given analytically.

For $T = 60$ min, the solution could be as follows:

$$\begin{aligned} C_1 &= 1.1388 \times N'_{\alpha 1} \\ C_2 &= -0.1146 \times N'_{\alpha 1} - 0.2433 \times N'_{\alpha 2} \\ &\quad + 0.2433 \times N'_\beta \\ C_3 &= 0.0028 \times N'_{\alpha 1} + 0.4139 \times N'_{\alpha 2} \\ &\quad - 0.1349 \times N'_\beta \end{aligned} \tag{2}$$

For $T = 30$ min, the solution could be as follows:

$$\begin{aligned} C_1 &= 2.4722 \times N'_{\alpha 1} \\ C_2 &= -\ 0.2157 \times N'_{\alpha 1} - 0.7867 \times N'_{\alpha 2} \\ &\quad + 0.7867 \times N'_\beta \\ C_3 &= 0.0064 \times N'_{\alpha 1} + 1.0988 \\ &\quad \times N'_{\alpha 2} - 0.2468 \times N'_\beta \end{aligned} \tag{3}$$

where N'_x is the normalised count rate as follows:

$$N'_x = N_x / (E_x \times F \times \varepsilon_f \times 10^6) \quad x = \alpha_1, \alpha_2, \beta \tag{4}$$

The sensitivity and the uncertainty analysis

The sensitivity of radon progeny concentration can be expressed as the measurement count rate per radon progeny concentration (cph/Bqm^{-3}). The measurement sensitivity of ^{218}Po is some 8.4 cph/Bqm^{-3} for 60 min cycle with $F = 2.5$ lpm, $E_\alpha = 22.8\%$ and $\varepsilon_f = 100\%$, which is more than 18 times higher than that of Kerr method with same parameters.

Ignoring the uncertainty of detection efficiency, flowrate as well as the collection efficiency, the uncertainty of EEC (σ_{EEC}) could be expressed as follows:

For $T = 60$ min,

$$\sigma_{EEC}^2 = \left(\frac{1}{F \times \varepsilon_f \times 10^6} \right)^2 \left(\frac{0.0038 N_{\alpha 1}}{E_\alpha^2} + \frac{0.001 N_{\alpha 1}}{E_\alpha^2} + \frac{0.0055 N_{\alpha 1}}{E_\beta^2} \right)$$

For $T = 30$ min,

$$\sigma_{EEC}^2 = \left(\frac{1}{F \times \varepsilon_f \times 10^6}\right)^2 \left(\frac{0.0228 N_{\alpha 1}}{E_\alpha^2} + \frac{0.0002 N_{\alpha 1}}{E_\alpha^2} + \frac{0.0969 N_{\alpha 1}}{E_\beta^2}\right)$$

Comparison experiments

Comparison experiments were carried out in the radon chamber at NIM[9] and in a basement in Beijing. The detection efficiency of alpha particles and beta particle were calibrated using ^{241}Am and ^{90}Sr-^{90}Y electroplated sources, while the flowrate was calibrated by a Gilibrator2 flow calibrator (Gilian, USA).

In the radon chamber comparison experiment, the 400 mm^2 PIPS detector (SARAD, Germany) and SPECTRA 5011 MCA (SARAD, Germany) was used as the detection system. The MP-ΣN pump (SIBATA, Japan) was used with flowrate of 2.64 lpm. The Ø25 PTFE filter (Millipore, USA) was used as fixed filter with collection efficiency of 100%. The distance between the filter and the detector was nearly 3 mm.

Field comparison measurement was carried out using three RPM-SF01 radon progeny monitors (Sairatec, China) in a basement at 60 min cycle, which worked in three different modes with the alpha–beta spectrometry method, the Kerr method and the Bigu method separately[10]. The lower limitation of detection of this monitor for the alpha–beta spectrometry method, the Kerr method and the Bigu method is 0.08 Bq/m^3, 0.89 Bq/m^3 and 0.12 Bq/m^3, respectively.

Table 1. Comparison results in radon chamber.

Cycle	Alpha–beta spectrometry method (Bq/m^3)				EQF3220 (Bq/m^3)
	^{218}Po	^{214}Pb	^{214}Bi	EEC	EEC
60 min	3459	3874	2241	3210 ± 23	3292 ± 211
	2779	2717	2006	2455 ± 20	2613 ± 121
	2838	3010	1768	2520 ± 20	2695 ± 218
	2319	2779	1236	2144 ± 19	2346 ± 203
30 min	4058	4614	2947	3922 ± 15	4083 ± 147
	3457	4487	2305	3549 ± 14	3866 ± 143
	2143	1813	1642	1783 ± 10	1738 ± 89
	1853	1781	1050	1511 ± 9	1664 ± 95

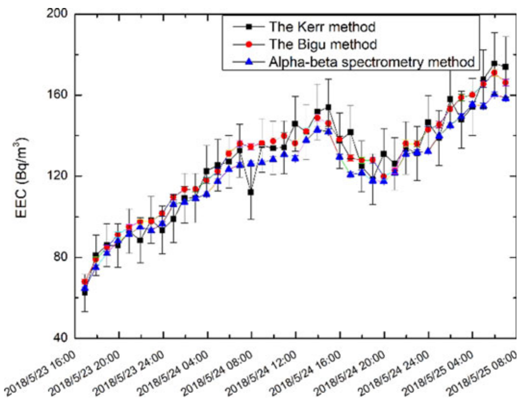

Figure 2. Field comparison of three different measurement methods.

RESULTS AND DISCUSSION

Comparison experiments in radon chamber

The reference value of radon progeny in the radon chamber of NIM was gotten from EQF3220 (SARAD, Germany), which could be traced back to the Chinese radon progeny standard. The comparison results are shown in Table 1, where the result of 60 min cycle is from last paper[3] and the result of 30 min cycle is recently measured.

Results show that the ^{218}Po, ^{214}Pb and ^{214}Bi concentrations as well as EEC could be given in each cycle using the alpha–beta spectrometry method. EEC given by alpha–beta spectrometry method is quite consistent with that of EQF3220 either for 60 min or for 30 min cycle. The uncertainty of alpha–beta spectrometry method is much lower than that of EQF3220 due to its high sensitivity.

Comparison measurement in field

Field comparison result in basement is shown in Figure 2. Results show that three different measurement methods agree with each other very well within 10% uncertainty. The ratio of ^{218}Po:^{214}Pb:^{214}Bi for alpha–beta spectrometry method is 1:1.06:0.87, while the ratio for Kerr method is 1:0.98:0.92, which have a little difference. But considering the uncertainty of ^{218}Po concentration and the difference of detection efficiency between beta particles emitted from ^{214}Pb/^{214}Bi and from ^{90}Sr-^{90}Y, the results can be accepted.

CONCLUSIONS

For the purpose of accurate measurement on radon progeny concentration, a newly developed radon progeny measurement method based on alpha–beta spectrometry was analysed and a series of comparison experiments were carried out in this study. This method can give the ^{218}Po, ^{214}Pb and ^{214}Bi concentrations as well as EEC in one measurement cycle with high sensitivity, which leads to lower uncertainty than other methods. This method can be used as reference method for the radon chamber. Most importantly, this method was derived from theoretical equations and has no limitation on measurement cycle time, which make it quite suitable for portable radon progeny monitor. Actually, this method is already realised in RPM-SF01 radon progeny monitor.

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