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STUDY ON PEAK SHAPE FITTING METHOD IN RADON PROGENY MEASUREMENT

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Alpha spectrum measurement is one of the most important methods to measure radon progeny concentration in environment. However, the accuracy of this method is affected by the peak tailing due to the energy losses of alpha particles. This article presents a peak shape fitting method that can overcome the peak tailing problem in most situations. On a typical measured alpha spectrum curve, consecutive peaks overlap even their energies are not close to each other, and it is difficult to calculate the exact count of each peak. The peak shape fitting method uses combination of Gaussian and exponential functions, which can depict features of those peaks, to fit the measured curve. It can provide net counts of each peak explicitly, which was used in the Kerr method of calculation procedure for radon progeny concentration measurement. The results show that the fitting curve fits well with the measured curve, and the influence of the peak tailing is reduced. The method was further validated by the agreement between radon equilibrium equivalent concentration based on this method and the measured values of some commercial radon monitors, such as EQF3220 and WLx. In addition, this method improves the accuracy of individual radon progeny concentration measurement. Especially for the ^{218}Po peak, after eliminating the peak tailing influence, the calculated result of ^{218}Po concentration has been reduced by 21 %.

INTRODUCTION

Rn/Tn and their progenies are the main resource of naturally occurring radioactive materials, which contribute nearly half of the total exposure dose of all natural resources[1]. They can be inhaled into human respiratory organs and contribute to the inner exposure. Numerous investigations have established that Rn/Tn is the second primary cause of lung cancer just behind smoking[2]. Consequently, Rn/Tn and their progeny have attracted considerable research interests in recent decades.

Exposure risk assessment of Rn/Tn is based on measurement of radon and thoron, especially their progeny. There are two methods for radon progeny concentration measurement: counting method and spectroscopy. In contrast, spectroscopy can provide information about alpha particles' energies, which will help to provide more accurate result within less time[3]. On account of the dramatic development of spectroscopy analysis technology, alpha spectroscopy has become one of the most popular methods for radon measurement in environment[4].

However, the peak tailing of alpha spectrum affects accuracy of this method. Alpha particles lose energy while passing through filter, air layer and detector death layer; therefore, the spectrum shows low-energy-peak tailing, which would bring about errors of peak count. For a long time, peak tailing is a big issue that affects accuracy of Rn/Tn progeny concentration measurement. Usually, traditional crosstalk factor correction method is used to alleviate this problem, but the crosstalk factors vary with the setting of alpha energy window and size of detector[5, 6]. Hence, its applications are limited.

To solve peak tailing problem and improve the alpha spectrum method, this article presents a shape fitting method. Experiments were carried out to compare the results before and after modification utilising shape fitting method. In addition, comparison was also done with two commercial continuous radon monitors to ensure the reliability of this method.

METHODOLOGY

Radon progeny concentration measurement based on shape fitting method

While measuring Rn/Tn progeny, contiguous peaks overlap each other due to peak tailing. Figure 1 shows a typical alpha spectrum. It is divided into three interest regions according to the three identified peaks: ROI1 covers the peak area consist of RaA (6.00 MeV) and ThC (6.05 MeV), ROI2 covers the peak area of RaC' (7.68 MeV) and ROI3 covers the peak area of ThC' (8.78 MeV). In the spectrum, it is hard to distinguish each peak from adjacent peak, so it is not possible to get exact count of each region.

To calculate Rn/Tn progeny concentration, it is essential to know net area of peak in different interval counting time. Usually, a fixed crosstalk factor is applied to alleviate this condition. The net counts of three interest regions are calculated using the

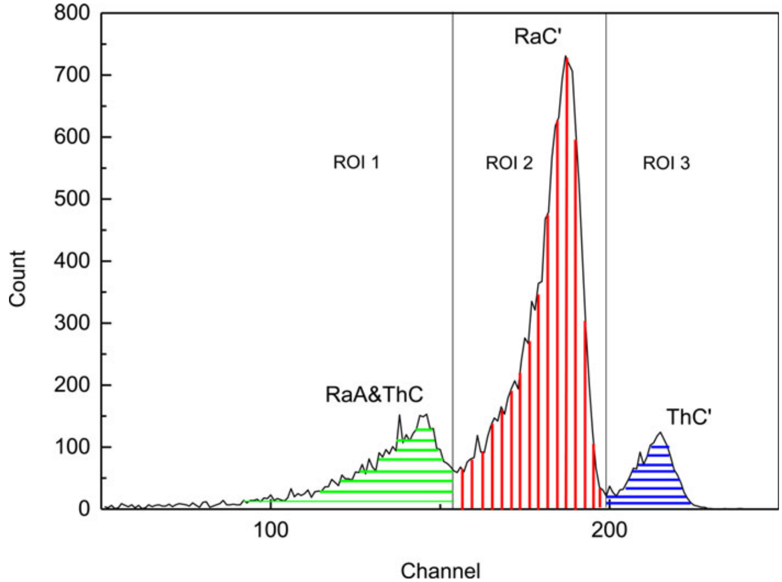

Figure 1. A typical α spectrum of field measurement.

following equations:

$$\text{ROI1: } N_1' = N_1 - K_{21}N_2 \quad (1)$$

$$\text{ROI2: } N_2' = N_2 + K_{21}N_2 - K_{32}N_3 \quad (2)$$

$$\text{ROI3: } N_3' = N_3 + K_{32}N_3 \quad (3)$$

K_{21} and K_{32} are crosstalk factors, which represent fraction of alpha particles that should have fallen in ROI2 and ROI3 but fall in ROI1 and ROI2, respectively. They are regarded as constant habitually and derived from pure Rn and pure Tn experiment[7]. In practice, crosstalk factors are quite sensitive to measurement conditions. Aerosol size and concentration as well as spectrum shift in aging multichannel spectrometer have influence on crosstalk factor[8, 9]. Therefore, the crosstalk factor needs to be calibrated regularly, and that will limit its application.

In this situation, an alternative approach is necessary. This article presents shape fitting method on the basis of thorough investigation of peak tailing problem in Rn/Tn measurement.

Alpha particles emitted by Rn/Tn progeny detected by the detector are supposed to form Gauss peaks in spectrum. It is expected that a Gauss function can feature the peak. However, those alpha particles lose energy while going through air between the filter and the detector. Their energy losses depend on the distance as well as the direction. On the other hands, the detector usually has 'dead layer' that forms an energy window for alpha particle. It also causes energy loss that depends on the alpha particle's direction. Considering all these factors, their energy losses rest with their effective distance. It can be described by an exponential function. At last, the real peak can be simulated by the function approximately as follows[10, 11]:

$$y(x) = G(x) + L(x) \quad (4)$$

$$\text{where } G(x) = Hg \times e^{-\frac{(x-xg)^2}{2\sigma^2}} \quad (5)$$

$$T(x) = Hs \times e^{\frac{x-xg}{\sigma \times ts}} \times erfc\left(\frac{x-xg}{\sqrt{2}\sigma} + \frac{1}{\sqrt{2}ts}\right) \quad (6)$$

$G(x)$ is a Gauss function indicating that radioactive counts are obeying Gaussian distribution, and it is used to fit the curve near the peak energy; $T(x)$ can be regarded as an exponential function approximately to characterise the low-energy-peak tailing, and it is mainly used to fit the curve below the peak energy. Hg, σ and xg stand for amplitude, the standard deviation and the peak channel of Gauss peak, respectively; Hs stands for amplitude of peak tailing; ts features the extent of peak tailing and x stands for channels in interest region.

In this algorithm, peak channel was fixed; this points to the likelihood that the influence of spectrum shift was reduced. Least square method was used to fit the rest four parameters (Hg, σ, Hs and ts) with initial parameter estimation values. MATLAB was utilised for the fitting. The main process is using MATLAB command 'fminsearch' to calculate optimum value of parameters, which can make the difference between measured curve and fitting curve minimum. These fitted parameters were unacted on spectrum shift, aerosol diameter and concentration. Consequently, this algorithm can prevent problem that happens in traditional crosstalk factor method effectively.

Net alpha count of each progeny can be gained validly with this alpha spectrum analysis method, and then Rn/Tn progeny concentrations are calculated via these count values. It provides an approach to solve peak tailing issue to some extent.

Based on shape fitting method, the Kerr method can calculate concentrations of all Rn/Tn progeny. The measurements of activity concentrations of Rn/Tn progeny can be divided into two processes: sampling and counting. At first, radon progenies in the air are sampled on the filer for 10 min. Then, count alpha particles emitted from the filter by the alpha spectrometer in three time intervals: (1) from the 2nd to 12th min, (2) from 15th to 30th min and (3) from 400th to 420th min. After that, three spectrums are obtained, which contain 3, 3 and 2 peaks, respectively. The last spectrum obtained from 400th to 420th min drops one peak, because the RaC' has nearly vanished during the third counting time. These measured spectrums are analysed by using shape fitting method, which fitted peaks from high energy to low energy in sequence and then gave net counts of each peak. Figure 2 shows the fitting curve result.

After these eight peaks fitted, their area counts were calculated through integration. Five of the eight peak area counts, denoted as N_{11}, N_{12}, N_{22}, N_{13} and N_{33}, were used for Rn/Tn progeny concentration calculation based on the Kerr method. N_{11}, N_{12}, N_{22}, N_{13} and N_{33} separately stand for alpha counts from Energy Region 1 in the first time interval, alpha counts from Energy Region 2 in the first and second time intervals, alpha counts from Energy Region 3 in the first and third time intervals. These five counts have already eliminated the influence of peak tailing.

Based on the decay law of Rn/Tn progeny during process of sampling and counting, it is easy to establish the relationship between concentrations of radon progeny and time interval counts. The following ordinary differential equations can be used to describe the time variation of the radon progeny atom number on filter:

$$\frac{\mathrm{d}n_i(t)}{\mathrm{d}t} = C_i v + \lambda_{i-1} n_{i-1}(t) - \lambda_i n_i(t) \qquad (7)$$

$$\frac{\mathrm{d}n_i(t')}{\mathrm{d}t} = \lambda_{i-1} n_{i-1}(t') - \lambda_i n_i(t') \qquad (8)$$

Equations (7) and (8) are for sampling period and post-sampling period, respectively. ***C*** stands for the concentration of Rn/Tn progeny in the sampled air; *n* stands for the radon progeny atom number; $i = 1, 2, 3$ stands for ^{218}Po, ^{214}Pb and ^{214}Bi for radon decay chain or $i = 1, 2$ stands for ^{212}Pb and ^{212}Bi for thoron decay chain. Solving these ordinary differential equations, the relationship between radon progeny atom numbers and concentrations can be figured out. Then the relationship between spectrum counts and radon progeny concentrations can be worked out.

In this article, according to the sampling and counting process as mentioned earlier, concentration of Rn/Tn progeny can be expressed as follows[12–14]:

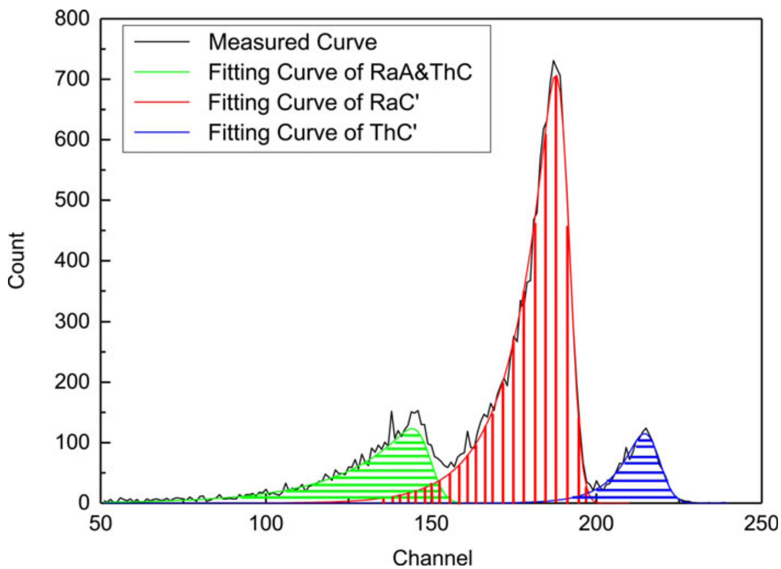

Figure 2. The fitting curve of α spectrum.

$$\begin{pmatrix} C_{\text{RaA}} \\ C_{\text{RaB}} \\ C_{\text{RaC}} \\ C_{\text{ThB}} \\ C_{\text{ThC}} \end{pmatrix} = \frac{1}{fv} \times 10^{-6} \times \begin{pmatrix} 28.2701 & 0 & 0 & -15.9019 & 0 \\ -3.4731 & -6.5704 & 7.5058 & 1.9536 & 0 \\ 0.5004 & 6.9944 & -3.2009 & -0.2815 & 0 \\ 0 & 0 & 0 & -0.0635 & 3.1340 \\ 0 & 0 & 0 & 4.9803 & -0.4522 \end{pmatrix} \times \begin{pmatrix} N_{11} \\ N_{12} \\ N_{22} \\ N_{13} \\ N_{33} \end{pmatrix} \quad (9)$$

$$\begin{pmatrix} \sigma^2{}_{\text{RaA}} \\ \sigma^2{}_{\text{RaB}} \\ \sigma^2{}_{\text{RaC}} \\ \sigma^2{}_{\text{ThB}} \\ \sigma^2{}_{\text{ThC}} \end{pmatrix} = (\frac{1}{fv} \times 10^{-6})^2 \times \begin{pmatrix} 28.2701^2 & 0 & 0 & -15.9019^2 & 0 \\ -3.4731^2 & -6.5704^2 & 7.5058^2 & 1.9536^2 & 0 \\ 0.5004^2 & 6.9944^2 & -3.2009^2 & -0.2815^2 & 0 \\ 0 & 0 & 0 & -0.0635^2 & 3.1340^2 \\ 0 & 0 & 0 & 4.9803^2 & -0.4522^2 \end{pmatrix} \times \begin{pmatrix} N_{11} \\ N_{12} \\ N_{22} \\ N_{13} \\ N_{33} \end{pmatrix} \quad (10)$$

$$\sigma_{\text{EEC}}{}^2(\text{Rn}) = (0.105 \times \sigma_{\text{RaA}})^2 + (0.515 \times \sigma_{\text{RaB}})^2 + (0.38 \times \sigma_{\text{RaC}})^2 \quad (11)$$

$$\sigma_{\text{EEC}}{}^2(\text{Tn}) = (0.912 \times \sigma_{\text{ThB}})^2 + (0.088 \times \sigma_{\text{ThC}})^2 \quad (12)$$

where v (l min^{-1}) stands for the sampling flow rate, and f stands for the detection efficiency of alpha spectrometer. The uncertainty of Rn/Tn progeny only considers the error of count statistical.

Comparison experiment

To show the effect of shape fitting method utilised in radon progeny measurement, comparison experiment was carried out in a 20-m^3 radon chamber at China National Institute of Metrology.

The radon chamber was filled with radon and thoron gases. Radon and thoron concentrations inside were about 7500–8000 and 12 000–17 000Bq/m^3, respectively, and were assumed to be approximately stable. Aerosol concentration was ~10 000–18 000 cm^{-3} with 150 nm diameter. Keep the Rn/Tn and aerosol concentration stable for more than 24 h, so Rn/Tn progenies were assumed to be in a high-level concentration.

In the experiment, Whatman GF/F (GE Co., USA) with 100 % filter efficiency was used with a sampling flow rate of 2.9 l min^{-1}. Counting was carried out using Spectra5031 (SARAD) multichannel spectrometer with 400-mm^2 PIPS detector. The detection efficiency was calibrated to be 19 %.

In order to measure Rn/Tn concentrations in the radon chamber continuously, EQF3220 (SARAD, German) and WLx (Pylon, Canada) were used. Then, Rn/Tn and their progeny concentrations derived from the Kerr method with and without shape fitting modification were compared with EQF3220 and WLx measurement results.

RESULTS AND DISCUSSION

Peak area modification

The peak area modification results are shown in Table 1 as well as Figure 2. In the figure, the fitting curves are very similar to the measured curve, indicating that the function used is appropriate to describe the spectrum curve and the shape fitting algorithm is practicable. Moreover, the fitting curve distinguishes three peak regions clearly. Figure 3 shows the error between measured curve and fitting curve. The error is no more than 50. This is quite small compared with the channel data. This means that this method does not transform spectrum curve much, and it has little influence on spectrum data and cannot help to alleviate experimental error such as fluctuating count, but it does improve spectrum analysis.

Table 1. Correction of peak area.

	Measured peak area	Fitted peak area	Ratio of correction (%)[a]
N_{11}	3719	3269	−12.1
N_{12}	12 296	12 570	2.2
N_{22}	16 748	17 158	2.5
N_{13}	1699	1755	3.3
N_{33}	3646	3844	5.4

[a]Ratio = $(\text{Area}_{\text{fitted}} - \text{Area}_{\text{measured}})/\text{Area}_{\text{measured}}$.

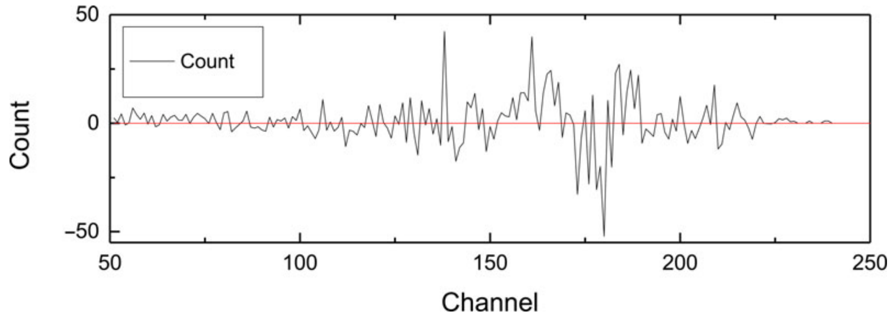

Figure 3. Error line of the fitting curve.

Table 2. Comparison of measurement results of the Kerr method with EQF3220 and WLx (Bq m^{-3}).

	RaA	RaB	RaC	ThB	ThC	EEC(Rn)	Uncertainty	EEC(Tn)	Uncertainty
Kerr's method without modification	8448	3819	3653	1224	737	4242	97	1181	20
Kerr's method with shape fitting modification	6976	4138	3692	1290	757	4267	98	1243	20
Ratio of correction (%)	−17.4	8.4	1.1	5.4	2.7	0.6		5.2	
EQF3220						3922	50	1157	28
WLx						4043	481	1042	124

EEC, equilibrium equivalent concentration.

In Table 1, the net peak counts are calculated with and without modification. The former one was calculated by integrating fitted peak curve, while the latter one was calculated by summing the interest region's counts up.

As evident in Table 1 that for high energy peaks (peak energy of 7.69 and 8.78 MeV), their peak area augments after correction. This is credible as the shape fitting method has compensated the counts loss due to low-energy tailing. On the contrary, low-energy-peak areas decrease after correction. This is ascribed to the fact that the alpha particles came from high energy have been subtracted.

Through the ratio of correction data, it is easy to see that the correction for N_1 is most noticeable, as much as 12.1 %. This accords with the fact that high-energy alpha particles (especially emitted from RaC) drop in the lowest energy region due to energy loss.

The comparison experiment results

In order to verify the feasibility of shape fitting method applied in radon progeny measurement, this study carried out Rn/Tn concentration measurement experiments in relative stable Rn/Tn concentration circumstance and calculated Rn/Tn concentration by the Kerr method using peak counts with and without shape fitting modification, meanwhile EQF3220 and WLx were used to monitor Rn/Tn concentration. Finally, these calculated results were compared with results provided by EQF3220 and WLx. Comparison results are given in Table 2.

It is indicated by the comparison results that concentrations of Rn/Tn progenies after the modification have not changed much the RaA concentration. EEC_{Rn} and EEC_{Tn} after correction are quite consistent with reference values provided by EQF3220 and WLx, which proves that the shape fitting method is reliable.

Particularly, concentration of RaA has decreased as much as 17.4 %, which mainly arises from the decrease of N_1 as discussed previously. This implies that the accuracy of RaA concentration is ameliorated markedly as the shape fitting method can give a more accurate peak area count of RaA.

CONCLUSION

When measuring ^{222}Rn/^{220}Rn concentration using alpha spectrum method, alpha particle usually loses energy when it goes through the air layer and 'dead layer', which results in the peak tailing from high-energy region to low-energy region. Peak tailing has been a holdback for a long time. In contrast to traditional crosstalk factor correction, shape fitting method is immune to spectrum shift and aerosol in environment; therefore, it helps to solve the peak tailing problem by providing more accurate net counts of each peak and finally improves the accuracy of Rn/Tn progeny concentration measurement. The theory analysis and experiment results in this study have confirmed the feasibility of this method.

It is expected that with a further simplification, the shape fitting method will gain a rosy application prospect in Rn/Tn progeny concentration measurement.

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