# Under High Radon Environment α Energy Spectrum Analysis

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Abstract. Energy spectrum processing technology is one of the key technologies to realize the rapid monitoring of artificial radioactive aerosol. At present, most commercial aerosol continuous monitoring equipment are suitable for the measurement of artificial radioactive aerosol under natural radon background. The energy spectrum processor technology used is also aimed at the situation of low radon background, which is not suitable for the measurement of artificial radioactive aerosol in high radon environment. In this paper, the combination of energy spectrum fitting (fitting method) and variable tailing proportional coefficient method (ratio deduction method) is used, and the program written on MATLAB platform is used to analyze  $\alpha$  by deducting radon daughters in the energy spectrum, the energy spectrum measurement in high radon environment can be realized. By analyzing the energy spectrum of mixed samples of U and Ra with different concentrations and comparing with the measurement results after cooling, it shows good consistency, which verifies the feasibility of understanding the spectrum method and provides a reference for the energy spectrum analysis in high radon environment  $\alpha$  Energy spectrum analysis provides an important technical means.

Keywords: energy spectrum analysis; fitting method; proportional coefficient method.

# 1. Introduction

Artificial radionuclide monitoring in the environment is an important technical means to do a good job of internal exposure. At present, two main methods are sampling laboratory analysis and on-site continuous monitoring. The laboratory analysis method collects aerosol samples through a large flow air sampler, and the processed samples are measured with a mass spectrometer or an energy spectrometer. The lower detection limit of a mass spectrometer for uranium and plutonium aerosols reported in the literature can reach 130±21 nBq/m3 [1], and the lower detection limit of an energy spectrometer for plutonium aerosols can reach 10-5 Bq/m3 [2]. Both offline measurement methods show good measurement accuracy, but the measurement time is relatively long. When the on-site working environment is polluted, it cannot be detected and alarmed in time. On-site, continuous monitoring is an online monitoring method and also the most studied aerosol monitoring method at present. With the emergence of new ion-implanted silicon semiconductor detectors and the rapid development of multi-channel analyzers, a large number of aerosol continuous monitoring equipment based on energy spectrum analysis have emerged. At present, there are two main energy spectrum analysis methods, namely the fitting method and the proportional coefficient method, compared with  $\alpha/\beta$  for the ratio method and false coincidence method, the energy spectrum analysis method is more accurate in the deduction of radon daughters. Because the on-site continuous monitoring sampling time and measurement time are relatively short, combined with the interference of radon daughters, the measurement accuracy is not very high. The higher the radon daughter concentration, the greater the error caused by this interference.

The continuous aerosol monitoring equipment currently used is mostly suitable for the measurement of artificial radioactive aerosols under natural radon background conditions. In some nuclear facilities and radioactive workplaces, especially underground nuclear facilities located in granite zones, the radon daughter concentration can reach 103 Bq/m3~104Bq/m3 [3-5]  $\alpha$  energy spectrum, the aerosol formed by these high concentrations of radon and its daughters will cause serious interference to the measurement, which will reduce the measurement sensitivity of the instrument or even invalidate it. Therefore, in the high radon environment, more accurate energy spectrum processing technology is needed to effectively deduct the interference of radon daughters

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to the measurement, to realize the rapid measurement of artificial radioactive aerosols. Therefore, it is of great significance to study the energy spectrum analysis technology under high local conditions to ensure the safety of the nuclear environment and the health of radioactive workers.

# 2. Energy spectrum analysis of radon daughters

### 2.1 Analysis of radionuclide peaks in energy spectrum of radon daughters

As a natural radionuclide, radon mainly includes uranium, thorium and other radioactive substances  $\alpha \ \beta \ \gamma$  It is formed by decay. There are mainly three isotopes of radon in nature: 222Ra, 220Ra and 219Ra, which are decay daughters of 238U, 232Th and 235U natural radiation systems respectively [6]. The natural abundance of 235U in nature is low, only 0.711%, and the half-life is long. In addition, the half-life of 219Ra is short, only 3.96s, so the content of 219Ra in the environment is extremely low. As radon is an inert element, it exists in the form of gas at normal temperature and pressure, and will not be collected by the filter membrane during sample collection, so figure 1-1 of 222Ra and 220Ra is mainly the decay chain of 222Rn, 220Rn and their daughters [7].



Figure 1. 222Rn (a) and 220Rn (b) decay chains.

In the decay chain of 222Rn, because the half-life of 210Pb is relatively long, and the subsequent daughter activity concentration is low, it is difficult to detect in the environment, so only the influence of 218Po, 214Bi, and 214Po tailing on the energy spectrum analysis is considered. It can be seen from the decay chain that 214Bi  $\alpha$  branching ratio of the decay is only 0.02%, which has little influence on the energy spectrum analysis and can be ignored. 214Po half-life (164.3µs) It is very short compared with its parent 214Bi (19.9min), so the two nuclides are in a long-term equilibrium state, that is, the radioactivity of daughter and parent is equal, so 214Po will generate counts in the energy spectrum; 222Rn decay chain, with  $\alpha$  radioactivity mainly includes 216Po, 212Bi and 212P, but the energy of 216Po is relatively low, 0.36MeV~0.5MeV, and the range in the air is usually less than 3mm. In addition, due to the self-absorption of the filter membrane  $\alpha$  particles are difficult to be collected and measured by the detector, and will not produce energy peaks in the energy spectrum. Similarly, 212Bi and 212Po in the 222Rn decay chain also have a long-term radioactive balance, and 212Po with a very short half-life will also generate counts in the

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energy spectrum. To sum up, the nuclides that produce energy peaks	in the energ	y spectrum of
radon daughters are 218Po, 214Po, 212Bi and 212Po.		

#### 2.2 Analysis of interfering nuclides in energy spectrum

The  $\alpha$  particle in the detection process will cause their energy spectrum to expand to a certain extent, and gradually soften to the low-energy region, forming the trailing phenomenon of the energy spectrum. The energy of radon daughters is usually 6.00MeV~8.78MeV, while the energy of artificial radionuclides is 4.15MeV~5.18MeV. Due to the influence of this tailing phenomenon, when using the energy spectrometer to measure artificial radionuclides, radon daughters will count in the energy area of artificial radionuclides, thus causing certain interference to the measurement of artificial radionuclides. The degree of interference will increase with the increase of radon concentration. Figure 2 shows the energy spectrum of radon daughters.



Figure 2. Radon daughters energy spectrum.

The energies of 212Bi and 218Po are about 6MeV. Due to the limited resolution of the detector, the energy peaks formed by the two nuclides are usually superimposed together, so only three independent peaks can be seen in the energy spectrum and there is an obvious phenomenon of low energy drag. 212Po has obvious high-energy tailing, which is mainly caused by the decay of 212Po  $\alpha$  particle and its parent 212Bi decay  $\beta$  particle matches the resulting count. In order to reduce the influence of tail counting on the measurement of artificial radionuclides, the distance between the detector and the sample is less than 3mm, and the working pressure is less than 800mtorr. The activity concentration of thorium daughters in the air is much lower than that of radon daughters, usually 1/10 of that of radon daughters. When sampling and measuring for a short time (the sampling time in this paper is 15min, and the measuring time is 10min), the 212Bi activity is low, and the impact on the analysis of artificial radionuclides can be ignored. In addition, the 212Po energy peak is not obvious in the energy spectrum, and the energy is relatively high compared with that of artificial radionuclides, The tail counts generated by the energy spectrum in the artificial radionuclide energy region are very small and can usually be ignored [8]. Therefore,  $\alpha$  energy spectrum analysis mainly uses the fitting method and the proportional coefficient method to deduct 218Po and 214Po in the energy spectrum, so as to realize the rapid measurement of artificial radionuclides.

# 3. Energy spectrum processing

#### 3.1 Analysis of radionuclide peaks in energy spectrum of radon daughters

Before energy spectrum smoothing, the background of the instrument in the low energy area, the radiation from the surrounding environment and  $\beta$  Count value generated by the ray in the low energy region. Figure 3 shows the original energy spectrum and the energy spectrum after deducting the background at a certain radon concentration.



Figure 3. Original energy spectrum (left) Energy spectrum after deducting background noise (right).

The function of energy spectrum smoothing is to remove the noise count in the energy spectrum and improve the signal-to-noise ratio. Figure 4 shows the energy spectrum curve after deducting background noise and the energy spectrum curve after smoothing under certain radon concentrations.



Figure 4. It refers to the energy spectrum curve (data1) and smoothed energy spectrum curve after deducting background noise under certain radon concentration (data2).

At present, the least squares method is the most widely used method to smooth the data. The basic method is to take k data points on the left and right sides of the first count to form 2k+1 continuous data points Use polynomials to fit these data points. The value of the fitting polynomial in channel i is the count of the smoothed energy spectrum in channel i [9]. Therefore, a relatively simple Savitzky-Golay first-kind filter is used to smooth the energy spectrum.

The final energy spectrum smoothing formula is:

$$\overline{y}_{i} = \frac{1}{35} \left( -3y_{i-2} + 12y_{i-1} + 17y_{i} + 12y_{i+1} - 3y_{i+2} \right)$$
(1)

#### 3.2 Peak finding methods

The main purpose of peak searching is to reposition the energy peak after smooth noise reduction. There are many methods for peak finding, such as derivative peak finding, function fitting peak finding, Kalman peak finding, second-order difference peak finding, symmetric zero area peak finding, etc. because  $\alpha$  The tailing phenomenon of the energy spectrum, the energy peak is not a Gaussian function and symmetrical form, so the most ideal peak seeking method is derivative peak

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seeking method [10]. In the analyzed energy spectrum, there are only two peak positions of 241Po and 218Po, so it is relatively simple to use the second derivative to find the peak. In the energy region, the minimum point of the second derivative can be considered the peak position.

Therefore, the second derivative of the 5-point smooth energy spectrum is:

$$\overline{y}_{i}'' = \frac{1}{7} (2y_{i-2} - y_{i-1} - 2y_{i} - y_{i+1} + 2y_{i+2})$$
<sup>(2)</sup>

#### 3.3 Energy calibration

Energy calibration is a process of one-to-one correspondence between channel address and energy. The channel address and particle energy shall meet the following formula:

$$Ch = aE_{\alpha} + b \tag{3}$$

Where Ch is the channel address and  $E_{\alpha}$  is  $\alpha$  Particle energy, a, b are energy scale coefficients, usually a certain value. In the actual measurement process, due to the change in the measurement environment and the change of the resistance and capacitance values in electronic devices, the energy peak will drift. Therefore, before energy spectrum analysis, energy calibration must be carried out to convert the channel address to the corresponding energy. The essence of the energy calibration process is to solve the new energy calibration coefficient.

Assume that the peak position channel address of 218Po after peak searching is P1,214Po, the peak position channel address is P2, and the energy scale coefficient is a', b'. At this time, the relationship between energy and channel address still satisfies Formula (11), and the value of a', b' is obtained as follows:

$$a' = \frac{P_2 - P_1}{1.69}, b' = \frac{7.69P_1 - 6.00P_2}{1.69}$$
(4)

The relationship between the energy scaled channel address Ch and energy is as follows:

$$Ch' = \frac{P_2 - P_1}{1.69} E_{\alpha} + \frac{7.69P_1 - 6.00P_2}{1.69}$$
(5)

The energy spectrum after energy processing can be used for further energy spectrum analysis.

### 4. α Energy Spectrometry

#### 4.1 fitting method

The energy spectrum fitting deduction method is to use an appropriate function to fit the energy spectrum of radon daughters, and deduct the counts of radon daughters in the energy spectrum from high to low, so as to obtain the energy spectrum of artificial radionuclides, calculate the net count P of artificial radionuclides in the energy spectrum, and then calculate the activity concentration of artificial radionuclides by using Formula (13).

$$C = \frac{N}{\varepsilon Q t} \tag{6}$$

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Among C is the activity concentration;  $\varepsilon$  is the instrument detection efficiency; Q is the sampling flow; t is the measurement time.

The spectrum solution process of fitting method is shown in Figure 5:



Figure 5. Flow Chart of Spectrum Solution by Fitting Method

The second part of the article has discussed the pretreatment of the energy spectrum in detail, so the method of background deduction of energy spectrum fitting is mainly introduced here. Bland and García-Torano et al  $\alpha$  Energy spectrum fitting model [11-13]:

$$\begin{cases} Y(x) = \sum_{i=1}^{NPI} A_i \left\{ (1 - \varepsilon_i) \cdot G + \varepsilon_i [\eta_i \cdot T_{1j} + (1 - \eta_i) \cdot T_{2j}] \right\} \\ G_i = \frac{1}{\sigma_i \cdot \sqrt{2\pi}} \cdot \exp\left\{ \frac{(x - \mu)^2}{2\sigma^2} \right\} \\ T_{n,j} = \frac{1}{2\tau_{i,n}} \cdot \exp\left\{ \frac{(x - \mu)^2}{\tau_{i,n}} + \frac{2\sigma_i^2}{2\tau_{i,n}^2} \right\} \cdot erfc\left\{ \frac{1}{\sqrt{2}} \left( \frac{x - \mu}{\sigma} + \frac{\sigma_i}{\tau_{i,n}} \right) \right\} \end{cases}$$
(7)

Where A is the peak area,  $\mu$  is the peak position,  $\sigma$  is the Gaussian parameter,  $\varepsilon$  is the asymmetric weight of the peak shape,  $\eta$  is the tailing parameter, A is the contribution fraction of each exponential function tailing, and erfc is the error complementary function. The function fitting is realized based on the nonlinear least square method through MATLAB. For the processed energy spectrum, first fit 214Po, as shown in Figure 6:



Figure 6. Fitting 214Po Energy spectrum

Then deduct 214Po from the original energy spectrum, as shown in Figure 7:



Figure 7. Deduct 214Po Energy spectrum

Continue to fitting 218Po energy spectrum with the fitting model, as shown in Figure 8:



Figure 8. Fitting 218Po Energy spectrum

Finally, 218Po is deducted from the energy spectrum to extract the energy spectrum of the nuclide to be tested, as shown in Figure 9:



Figure 9. Deduct 218Po,214Po Energy spectrum

Calculate the count value in the nuclide energy spectrum to be measured, and use Formula (14) to calculate the activity concentration of artificial radioactive aerosol.

The energy spectrum fitting deduction method can better retain the energy peak of artificial radionuclides. It can not only obtain the activity concentration of radionuclides but also analyze the composition of artificial radionuclides. However, this method is applicable to the situation where the concentration of radon daughters is relatively low and the tailing effect of the energy spectrum is relatively small. For the situation where the radon background is particularly high, the proportional deduction method is considered to deduct the interference of radon daughters.

#### 4.2 Proportional deduction method

The proportional deduction method is also known as the fixed proportional coefficient deduction method. This method considers that the spectrum shape of radon daughters is consistent under different measurement conditions, so the interference caused by radon daughters can be counted and deducted according to the ratio of the peak position count of radon daughters to the count generated in the artificial radiation performance area, thus realizing the measurement of artificial radionuclides. There are many methods to divide energy zones, including two energy zone method and multi energy zone method. The two energy region method divides the energy region into two energy regions: artificial nuclide energy region and radon daughter energy region. This method does not consider  $\alpha$  The interference of particles in the artificial nuclide energy zone is different, and the analysis error is large. Based on the analysis accuracy, the energy spectrum is divided into three energy regions by multi energy region division method. The energy division is shown in Figure 10.



Figure 10. Schematic diagram of energy division

The energy region is mainly divided according to the broadening degree of P particles in the energy spectrum [14]. The range of three energy regions and counting sources are shown in Table 1.

Tuote II The funge of three energy regions and counting sources.				
Area	Energy Range	Peak Count Source	Trailing Count	
	(MeV)	Peak Position (MeV)	Source	
1	4.08~5.58	241Am(5.48),239Pu(5.16) 235U(4.40),238U(4.20)	218Po, 214Po	
2	5.80~6.14	218Po(6.00)	214Po	
3	7.49~7.83	214Po(1.69)		

Table 1. The range of three energy regions and counting sources.

It can be seen from Table 1 that the tailing count in energy zone 1 mainly comes from the tailing count of 218Po and 214Po, and the tailing count in energy zone 2 mainly comes from the tailing count of high energy zone 214Po. Record 218Po and 214Po, and the proportional coefficients of tailing counting in energy zone 1 are k21 and k31; The proportional coefficient of 214Po's tailing count in energy zone 2 is k32, so the net count in artificial radionuclide energy zone 1 can be calculated by the following formula:

$$N_0 = N_1 - k_{31} N_3 - k_{21} \left( N_2 - k_{32} N_3 \right)$$
(8)

N0 is the net count of artificial radionuclides, N1 is the total count of artificial radionuclides, N2 is the total count of 218Po in energy zone 2, and N3 is the total count of 214Po in energy zone 3. The scale factor can be calculated by the following formula:

$$k_{ij} = \frac{N_{ij}}{N_i} \tag{9}$$

kij is the proportional coefficient of the tailing count generated by the i energy zone in the j energy zone, Ni is the total count of the i energy zone, and Nij is the count value of tailing generated by the i energy zone in the j energy zone. Nij can be obtained by integrating the specific energy zone using the fitting model in the function fitting method. The calculated N0 can be brought into Formula (13) to calculate the concentration value of the nuclide activity to be

# 5. Verification and analysis

To verify the feasibility of the algorithm, The data of U and Ra mixed samples in 7 radon environments with different concentrations  $\alpha$  Energy spectrum analysis. The total count of nuclides to be measured calculated by spectrum solution is compared with the total count of energy spectrum measured after cooling by decay method, to verify the feasibility of the two spectrum solution algorithms in a high radon environment. Table 2 shows the results of the fitting method.

classific ation	Radon concentratio n (Bq/m3)	Coolin g time and counting	Juxtaposition Count value	Relativ e deviation	Proportio nal deduction method	Relati ve deviation
U Mixed sample	742	6 d 4710	4572	2.9%	4322	5.4%
	1860	2 d 4620	4651	0.67%	4759	3%
	2160	16 h 6230	5946	4.6%	6078	2%
Ra Mixed sample	442	6 h 2291	2222	3.3%	2093	8.3%
	1200	14 h 2190	2266	0.35%	2142	2.2%
	2600	20 h 2190			2190	3.5%
	3400	8 h 1758			1810	2.9%

Table 2. Summary of spectrum analysis results.

The U and Ra mixed spectra under 5 different concentrations of radon have been measured by using the fitting method, and the relative deviation is within 5%, showing a high measurement accuracy; The proportional coefficient method was used to analyze 7 mixed samples with different radon concentrations. The relative error was large in the analysis of 742Bq/m3 and 442Bq/m3 mixed samples with relatively low radon concentrations, and the other analysis results were all within 4%.

The fitting method and the proportional deduction method show a large deviation when analyzing the U mixed sample with a radon concentration of 724Bq/m3 and the Ra sample with a radon concentration of 442Bq/m3. This deviation is more obvious in the proportional coefficient deduction method (more than 5%), because when the radon concentration is relatively low, there is less data in the energy spectrum for the fitting method, and the fitting accuracy of the function is reduced; For the proportional deduction method, the insufficient amount of data brings about a large statistical error, which causes a certain deviation when determining the proportional coefficient, thus leading to a large deviation between the spectrum solution result and the true value.

When the radon concentration is 2160Bq/m3, the relative deviation generated by the fitting method for energy spectrum analysis is greater than that generated by the proportional coefficient deduction method. This is because when the radon daughter concentration increases, the

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interference in the energy region to be measured is more serious, and the fitting accuracy of the function decreases. When the radon concentration is relatively low, the spectral solution effect of the function fitting method is better than that of the proportional coefficient deduction method. When the radon concentration increases, the relative deviation of the energy spectrum analysis results of the proportional coefficient deduction method gradually decreases, and the analysis accuracy is higher in the strong radon environment. Based on the above considerations, in order to reduce the analysis error, only the proportional coefficient deduction method is used to analyze the energy spectrum when measuring the radon concentration of 2600Bq/m3 and 3400Bq/m2 Ra mixed samples, and the relative deviation is within 4%.

# 6. Conclusion

From the spectrum data, the fitting method has higher analysis accuracy when the radon concentration is relatively low. In the case of relatively high radon concentration, the analysis accuracy of the ratio coefficient deduction method is relatively high. Therefore, the corresponding radon concentration threshold should be set when analyzing the alpha spectrum in a high radon environment, and different energy spectrum analysis methods should be selected according to the relative size of the radon concentration on site. Due to the limited experimental conditions, there is no in-depth analysis of threshold selection. By analyzing the characteristics of  $\alpha$  energy spectrum of radon daughters, the types of interfering nuclides in the energy spectrum are determined. The fitting method and the proportional coefficient method are used to analyze the energy spectrum of U and Ra mixed samples at different radon concentrations, and good measurement results are obtained, which proves the applicability of the two methods in a high radon environment.

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