

Study on the Decay Law of γ Energy Spectrum of Radon and Its Daughters in Radon Chamber

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Abstract: Radon is one kind of radioactive gas which mainly distributed in soil and rocks, because of its strong upward migration ability, it is easy to diffuse into the air and cause radiation damage as the respiratory system enters the human body, which may cause lung cancer. Accurate measurement of concentration has always been a research area that has attracted much attention. There are various methods for measuring radon. Commonly used at home and abroad are thermoluminescence radon method, solid nuclear track radon method, activated carbon method, electret method, and scintillation chamber method. It is qualified for measurement under normal conditions, but in special applications such as the deduction of radon background in aerial gamma measurement, the above method is no longer applicable. At present, the detectors used in aviation gamma instruments at home and abroad are mainly large crystal sodium iodide detectors. The high-resolution array detector aviation gamma spectrometer developed by Ge Liangquan of Chengdu University of Technology also uses this kind of detection. In order to explore the effect of the detector on the direct measurement of the gamma energy spectrum of radon and its daughters. An experimental platform was built based on the HD-6 multifunctional automatic control radon chamber, the large crystal NaI(Tl) detector was used for measurement, and the HPGe detector was used for comparative measurement. Finally, the measurement results of the two were measured with the RAD7 radon meter. The obtained radon concentration was compared, and the reason for the abnormal data was studied. Through experiments, the large crystal NaI(Tl) detector can be applied to the direct measurement of the γ energy spectrum of radon and its daughters, and its practical effect is better than that of the electric cooling HPGe detector. The experiment found that the temperature is The count rate of the large crystal NaI(Tl) detector cannot be ignored, and temperature correction is required.

Keywords: Large Volume NaI(Tl) Detector, γ Energy Spectrum Method for Measuring Radon, Daughters of Radon

1. Introduction

Radon is one kind of natural radioactive gas that exists widely in nature. It is mainly produced by the decay of radionuclides ^{238}U and ^{226}Ra in rocks and soil. The atmosphere contains only a trace amount of radon, the content is less than 0.1%, but due to human activities or The increase in local radon concentration caused by natural changes may cause harm to human or animal health. Therefore, the measurement of radon is an important research area. Aeronautical gamma spectroscopy measurement refers

to the installation of an aeronautical gamma spectrometer system on an aircraft to measure the gamma spectra at the ground-air interface, so as to obtain the content of uranium, thorium, potassium and other nuclides in the surface medium to determine the area of mineral resources distributed. The counts caused by the gamma rays emitted by radon and its decay daughters in the atmosphere need to be deducted from the gamma energy spectrum measured by the aerial gamma detector, while conventional radon detection methods such as thermoluminescence radon measurement method, The solid nuclear track radon method is not suitable for such use occasions. Therefore, it is necessary to explore the feasibility

and effect of the NaI(Tl) detector, which is often used in aerial gamma detectors, for direct measurement of radon and its progeny. [1-5].

Liu ZhiHe studied the gamma spectrum measurement method of radon in the Ge(Li) detector activated carbon method, and calculated the gamma spectrum count and the response coefficient of radon concentration; Wu Rui et al. studied the technology of in-situ HPGe gamma spectrometer to measure the radon balance factor in the tunnel air. It shows that the HPGe γ spectrometer can directly measure the characteristic gamma rays of ^{214}Pb (or ^{214}Bi) in the tunnel; research has proved that the double probe γ -recombination method using NaI(Tl) crystal can measure the concentration of ^{222}Rn in water with high measurement accuracy. Compared with the traditional method, the dual-probe γ -weighting method can improve the signal-to-noise ratio of the measurement; Yi proposed a correction method based on fast Fourier transform to subtract the scattered rays of atmospheric radon. This method has a better effect on radon deduction, saves application costs, and solves the problem that traditional spectral lines are better than atmospheric radon correction. In summary, it is feasible to use nuclear radiation detectors to directly measure the gamma energy spectrum of radon to estimate the radon concentration. Use NaI(Tl) detector or HPGe detector to obtain corresponding measurement results. Correcting the radon concentration from the measurement method and data can improve the measurement accuracy. However, there are relatively few studies on directly using large-volume NaI (Tl) to measure the γ radioactivity of radon and its progeny in the air, so this experiment has certain reference significance for application research in related fields. [6-10].

2. Theoretical Background

Radon (^{222}Rn) in the uranium series and its progeny ^{214}Bi , ^{214}Pb and ^{210}Pb are the main gamma radiators, of which ^{214}Bi and ^{214}Pb account for 98% of the total uranium radiation exposure, while ^{218}Po and ^{214}Po are short of radon. The main alpha radiators in the life-span progeny, there are three long-lived radon progeny in the uranium series: ^{210}Bi (half-life of 5 days, is the main beta radiator), ^{210}Pb (half-life of 22.3 years, is a weaker beta radiation Body) and ^{210}Po (with a half-life of 138.4 days).

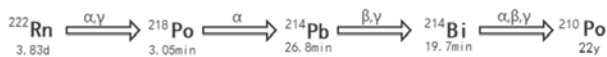


Figure 1. Radon daughters simplify the decay chain.

According to the decay law of radionuclides, after 10 half-lives of the longest half-life nuclides in the daughters, the radionuclide and its daughters can be considered to reach radioactive equilibrium. From the decay chain in Figure 1, it can be seen that radon and It takes about 268 minutes to establish the radioactivity balance of its daughters. According to formula (1), the radioactivity of radon can be calculated by measuring the radioactivity of its daughters.

$$\lambda_a \times N_a = \lambda_b \times N_b = \lambda_c \times N_c \quad (1)$$

In this formula:

$\lambda_a, \lambda_b, \lambda_c$ are the decay constants of the nuclides a, b, and c in the decay chain. N_a, N_b, N_c are the counts of nuclides a, b, and c by the detector.

3. Experimental Design

The experiment uses the HD-6 multifunctional automatic control radon chamber developed by East China University of Technology, with a minimum working concentration of 370Bq/m^3 , and a chamber volume of $220 \times 110 \times 110$ (cm); the RAD7 model produced by DurrIDGE, the United States, is used to measure radon. The lower limit of detection can reach 0.1pCi/L ; the portable electric-cooled high-purity germanium gamma spectrometer produced by ORTEC in the United States with a resolution of $1.72\text{keV}@0.662\text{MeV}$; the NaI(Tl) detector produced by Saint-Gobain in France, The crystal volume is $10 \times 10 \times 40$ (cm), and the resolution FWHM is $8.0\%@0.662\text{MeV}$.

Before starting the experiment, place the NaI(Tl) detector and HPGe detector at the designated position. Use lead bricks to shield the detector to reduce the influence of the natural radioactive background. Only the measurement surface is left, and the measurement surface faces the radon chamber. RAD7 It is equipped with a suction pump, which can carry out cyclic drying measurement by connecting the air inlet and outlet holes with the radon chamber. Start the radon chamber. After 24 hours of radon replenishment, continue to wait for 268 minutes to ensure that the radon and its progeny reach the radioactive equilibrium, then start the detector, and continuously measure the half-life of 2 to 3 radon.

4. Data Processing

Due to the interaction between γ -rays and matter, mainly including photoelectric effect, Compton scattering and electron pair effect, the output energy spectrum is more complicated, and all-power peaks, Compton plateaus, single escape peaks and double escape peaks are formed in the energy spectrum. Wait. In the actual γ spectrum, due to the influence of scattered photons and backscatter peaks, annihilation radiation peaks, characteristic X-rays, bremsstrahlung radiation, cumulative effects, peak effects, edge effects and other factors, the measured energy spectrum becomes more Complex, so we must first deduct the background of the experimental data [11].

Use Matlab data analysis software to extract data from the instrument output file. It is planned to use the SNIP algorithm to directly perform background subtraction. It is divided into four steps. First, it is necessary to transform each address count of the energy spectrum, and use the LLS operator (formula (2)), compress the counting range through natural logarithm operation, calculate the high-count spectrum, and the square root operation can extract the weak peak information.

$$V(i) = \ln[\ln(\sqrt{y(i) + 1} + 1) + 1] \quad (2)$$

In this formula:

i represents the road address; $y(i)$ represents the count of the i road address; $V(i)$ represents the transformed value after LLS operation.

After iteratively calculating the value of each change, the background spectrum data is obtained through the inverse LLS transformation, and the exponential operation with e as the base is used for the transformed value twice, and then the

square is calculated, see formula (3).

$$b(i) = [e^{e^{V(i)} - 1} - 1]^2 - 1 \quad (3)$$

The convergence speed of the entire iteration of the traditional SNIP algorithm is slow. Therefore, the Simpson formula is used to use the three points of the interval to divide the integral difference. The Scott coefficients are 1/6, 4/6, respectively. 1/6, which replaces SNIP iterative calculation. Combine formulas (2~4) to program the algorithm in Matlab

$$V_p(i) = \min\{V_{p-1}(i), \frac{1}{6}[V_{p-1}(i+p) + 4 \times V_{p-1}(i) + V_{p-1}(i-p)]\} \quad (4)$$

In this formula:

p represents the p -th iteration, that is, starting from 1 and adding 1 after each iteration operation until it is equal to the given value of m (transformation window width increasing method); $V_p(i)$ represents the LLS transformation value of the p -th iteration [12].

For the selection of the number of iterations m , if a fixed number of iterations is used, the effect of background subtraction in different energy ranges of the γ energy spectrum will be greatly different. Therefore, the adaptive

SNIP algorithm is used to determine the width of different energy peaks. The number of iterations in this range can ensure a good background subtraction effect in the full spectrum range.

The data after subtracting the background is fitted with the nonlinear fitting function GaussAmp in the Origin function drawing software, and the result is shown in figure 2. It can be seen that the energy spectrum after subtracting background fitting can clearly strip out the characteristic peaks of radon and its daughters [13-15].

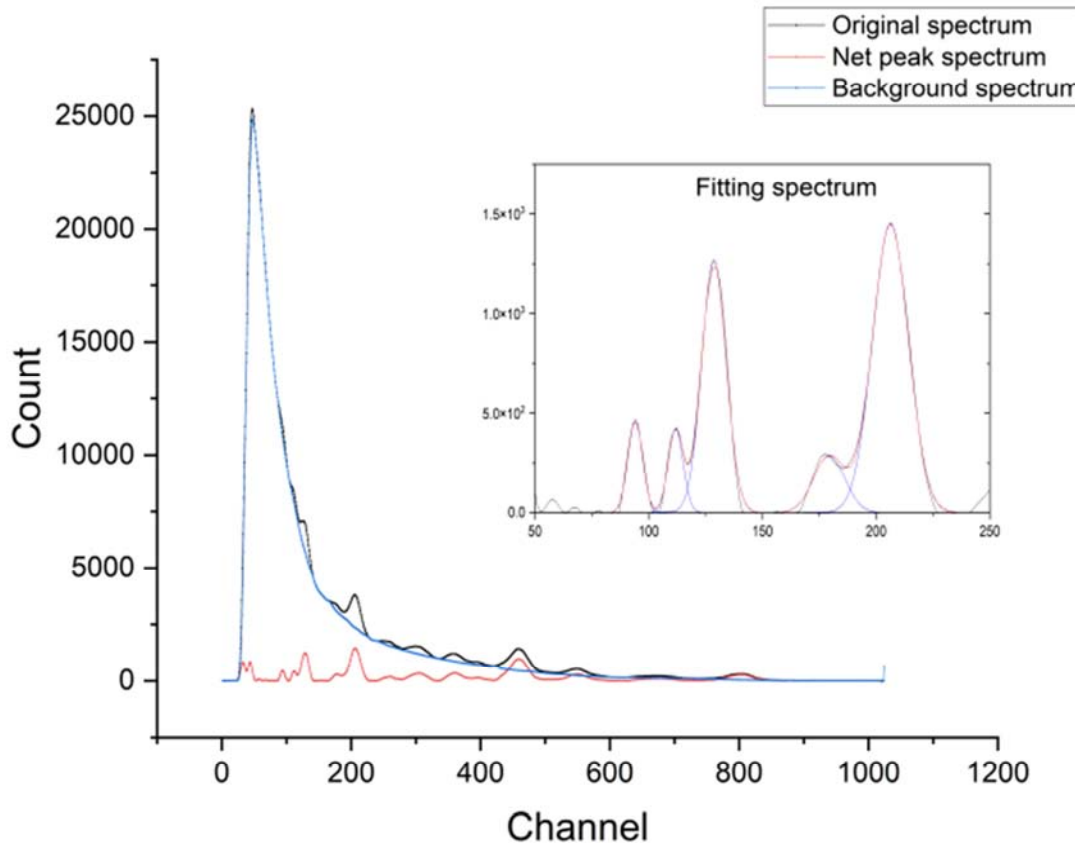


Figure 2. NaI(Tl) measurement data processing.

5. Experimental Results and Analysis

Use Origin function drawing software to visually present the processed energy spectrum data, and the results are as follows:

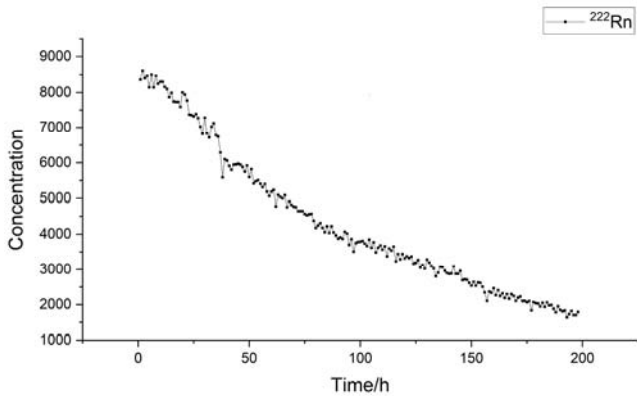


Figure 3. RAD7 measured radon concentration.

The measurement interval of RAD7 is 1h, continuous measurement is 8 days, after fitting, the formula $y=8.7245e^{(-0.008x)}$ is obtained, and the fitting degree is 0.9914, which conforms to the exponential attenuation, but due to the airtightness of the radon chamber itself Part of the radon leakage and the adsorption of radon on the chamber wall lead to the reduction of radon, so this trend is slightly different from the standard exponential decay trend. This curve will be used as a standard curve to verify the correctness of the data measured by the detector.

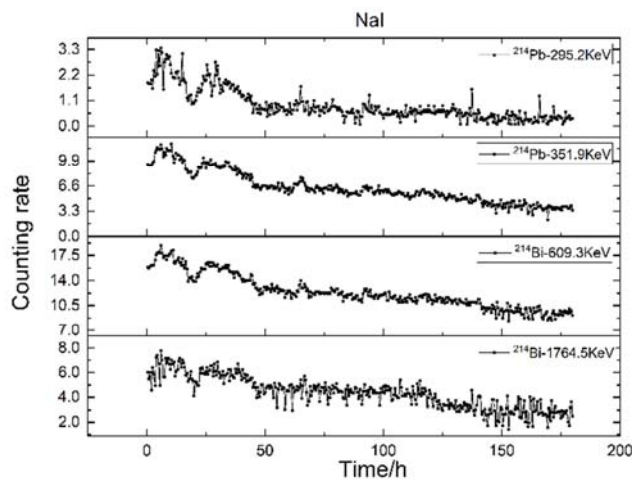


Figure 4. Count rate of characteristic peak of radon gamma decay daughter measured by NaI(Tl) detector.

From the simplified decay diagram of radon daughters in figure 1 and the principle of radionuclide radioactivity balance, it can be seen that ^{214}Pb or ^{214}Bi is the main gamma decay daughter of radon, and the change trend of the gamma ray count rate of ^{214}Pb or ^{214}Bi can reflect the radon count rate change trend. It can be seen from Figure 5 that the data measured by the NaI(Tl) detector and the HPGe detector show an exponential decay trend as a whole, but in comparison, the data measured by the HPGe detector fluctuates greatly, and the large crystal NaI (The data measured by the Tl) detector can better reflect this trend. In the energy spectrum measured by the NaI(Tl) detector, the energy peak of 351.9keV for ^{214}Pb and the energy peak of 609.3keV for ^{214}Bi are more representative.

However, it can be seen in the figure 5 that the image has large fluctuations in certain time periods, and this change is reflected in the energy spectrum measured by the NaI(Tl) detector and the HPGe detector. The fluctuations are found after observation. Location is related to time, and it is speculated that this change is caused by changes in ambient temperature. Therefore, after the experiment, the NaI(Tl) detector was used alone, and the radon chamber was not activated for continuous measurement for 8 days under the same experimental conditions. The experimental data was processed by Origin using the same method to obtain figure 6.

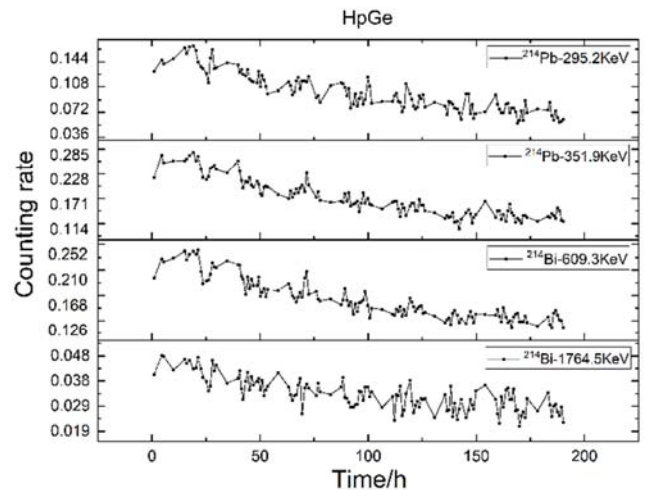


Figure 5. The HPGe detector measures the characteristic peak count rate of radon gamma decay daughters.

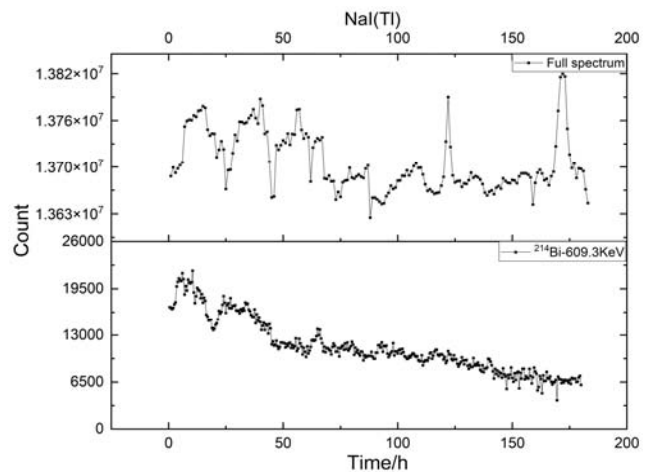


Figure 6. Comparison of NaI(Tl) full spectrum count and ^{214}Bi -609.3keV peak.

The upper half of figure 6 shows the trend of the measured full-spectrum data with temperature. Due to the large temperature change during this measurement period, there are obvious differences in the data before and after the measurement, but the trend of changes in each day is still roughly identical. It can also be seen that temperature is an important factor affecting the count rate of large volume NaI(Tl). The lower part is the ^{214}Bi -609.3keV peak count curve shown in Figure 3. The two experiments were completed in the same month, and the overall environmental

temperature has not changed much. It can be concluded from the comparative observation that the temperature change has a significant effect on the full spectrum count and radon. The impact of the feature peak count of the daughters is roughly the same.

6. Conclusions

The experimental results show that:

- 1) It is feasible to use the NaI(Tl) detector and the electrically cooled HPGe detector to directly detect the γ energy spectrum of radon and its daughters. Under the same conditions, the two detectors have a low energy end of ^{214}Pb at 351.9keV and ^{214}Bi at 609.3keV. The peak measurement results are better, and the stability of the data obtained with the NaI(Tl) detector is better than the data obtained with the HPGe detector.
- 2) Temperature has a great influence on the performance of the large volume NaI(Tl) detector. The response relationship of the counting rate to temperature should be measured in advance during use, so that the temperature correction of the experimental data can be carried out in the later stage, so as to obtain more accurate radon concentration and temperature. Relationship.
- 3) The improved Simpson_SNIP algorithm has a good effect on the background deduction of the γ energy spectrum of radon and its progeny. Combining the nonlinear fitting function GaussTemp to fit the data after subtracting the background, the characteristic peaks of radon and its progeny can be fitted. More obvious separation.

7. Shortcomings and Prospects

Due to the limited experimental conditions, the depth of the research in this experiment is still insufficient, especially for the temperature effect. Only the influence of temperature changes on the count of the large volume NaI(Tl) detector is discussed, but the detector cannot be obtained. The response curve to temperature has led to the failure to perform mathematical corrections, which can be further explored when the experimental equipment is perfected in the future.

Furthermore, because the application object of the experiment is the deduction of the γ background caused by radon and its daughters in aerial γ energy spectrum detection, the radon concentration in the environment of the aerial γ instrument system during operation is different from that on the ground., the relationship between the simulated radon concentration and the count rate in the radon chamber is not completely representative, and the relative relationship between the radon concentration and the count rate can be further fitted.

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