Study on the influence of the gas superficial velocity on the radon adsorption of activated carbon

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Abstract. In order to improve the radon adsorption efficiency of activated carbon and provide a reference for reducing the cost of radon purification, the radon adsorption experiment of activated carbon under different gas superficial velocities under the active adsorption mode was studied to measure the radon adsorption quantity of activated carbon. The radon adsorption quantity of apricot shell activated carbon and coal-based activated carbon did not change significantly with the increase of the gas superficial velocity. When the gas superficial velocity increased from 0.9cm/s to 2.0cm/s in turn, the radon adsorption quantity of coconut shell activated carbon did not change significantly. The radon adsorption quantity of coconut shell activated carbon increased first and then decreased when the gas superficial velocity increased from 2.0cm/s to 3.1cm/s. When the gas superficial velocity was about 2.7cm/s, the radon adsorption quantity of coconut shell activated carbon reached its peak value. 2.7cm/s can be selected as the optimum gas superficial velocity for the radon adsorption of coconut shell activated carbon.

1. Introduction

As a radioactive gas, radon can be absorbed into the human body and forms the internal radiation. Radon exists widely in the closed environment, such as underground mine tunnels, nuclear fuel factories or related laboratories. Therefore, it is necessary to take corresponding measures to reduce and eliminate radon with relatively high radon concentration or potential radon threat. The activated carbon adsorption method is one of the most widely use methods for radon reduction and radon removal [1-4]. At present, the research on the radon adsorption by activated carbon mainly focuses on: (1) the influence of the structure of activated carbon on the radon adsorption [5-7]; (2) the influence of external conditions on the radon adsorption by activated carbon [8]. While in the research on the external conditions, it mainly focused on either the adsorption effect of inert gases, such as krypton, xenon, etc. [9-10], or the influence of temperature, humidity, pressure and other factors on the adsorption performance [11-12]. There are relatively few references on the factors affecting the gas superficial velocity in the process of radon adsorption of activated carbon. In our work, based on
previous studies, the influence of different gas superficial velocities on the radon adsorption of activated carbon is further discussed by referring to relevant researches and designing the experiment.

2. Experimental platforms

2.1. Adsorption platform
The experimental adsorption platform consists of the activated carbon adsorption bed, electronic flow meter, diaphragm pump and multi-functional radon self-controlled chamber. The radon extracted from the radon chamber by the diaphragm pump forms an internal circulation through the activated carbon adsorption bed and the flow meter, as shown in Figure 1 and the electronic flow meter has been verified by National Institute of Measurement and Testing Technology. The gas flow rate of the diaphragm pump is controlled by the working voltage to provide different gas superficial velocity for the experiment. The volume of the multi-functional radon self-controlled chamber is 3.8m³, and the radon concentration regulation range is 0-5000Bq/m³. The volume of the radon chamber is much larger than the volume of the radon gas extracted from the radon chamber (The maximum extraction gas volume is 300L). Therefore, the influence of the experiment about radon adsorption of activated carbon on the radon concentration in the radon chamber can be neglected.

The activated carbon materials used in the experiment were three kinds of granular activated carbon materials; these include coconut shell activated carbon, coal-based activated carbon and apricot shell activated carbon. The particle sizes of activated carbons were between 1mm and 2mm, and their iodine values were between 850 and 1050. Before experiment, the deionized water was used to remove the ash on the surface of activated carbon and the gas adsorbed from the air. Then they were put into the drying oven and dried for 4 hours at 120°C. After drying, they were placed in the activated carbon adsorption bed as shown in Figure 2.

![Figure 1. Schematic diagram of the adsorption experimental platform](image1)

![Figure 2. Schematic diagram of the structure of the activated carbon adsorption bed](image2)

2.2. Measurement platform
After the adsorption experiment was completed, the activated carbon was taken 200g from the gas admission end of the activated carbon adsorption bed, and then it was filled in polyethylene holder (φ7.5cm×7.5cm). The activated carbon samples were measured by HPGe γ-ray spectrometer, and the γ-ray spectrometer with high energy resolution was produced by ORTEC. A lead shield with the thickness of 4mm was used in the measuring system to minimize the environmental background. The γ-ray spectrum of both the environmental background and the activated carbon included radon and its daughters were measured separately. The radon in the radon chamber reached the radioactive equilibrium before the adsorption experiment. According to the radioactive equilibrium theory, 214Bi which is the decayed daughter of radon had higher net counts of 0.609 MeV peak in γ-ray spectrum than other decayed daughters, therefore it was selected to analyze the radon adsorption quantity of activated carbon samples [13]. And the radiation background of coconut shell activated carbon, apricot
shell activated carbon and coal-based activated carbon were 349h⁻¹, 239h⁻¹ and 228h⁻¹ about net counts of 0.609 MeV peak.

3. Determination of adsorption time about activated carbon
The radon concentration in radon chamber was set to 1400 Bq/m³, and the temperature and relative humidity were stable at 18°C and 50% (the following experiments were all under the conditions of temperature and relative humidity), and the gas superficial velocity was set to 1cm/s. After the coconut shell activated carbon adsorption bed adsorbed for 45, 60, 75 and 90 min separately, and then the activated carbon samples were measured by γ-ray spectrometer. The changes of 0.609 MeV peak counts with measurement time are shown in Figure 3.

![Figure 3. Measurement results of the activated carbon at different adsorption times](image)

According to the growth rate of 0.609 MeV peak counts of the activated carbon samples at different adsorption times, the adsorption depth of activated carbon at different adsorption times can be judged. When the adsorption time is between 60 min and 90 min, the changes of 0.609 MeV peak counts at the same measurement time are very weak, which can almost be ignored. Therefore, when the adsorption time is 60min, the adsorption depth reaches 100%. When the gas superficial velocity is about 1cm/s or greater than 1cm/s, the adsorption depth of the carbon bed can reach 100% within the adsorption time of 60min.

4. Analysis of the activity of radon adsorbed by activated carbon
The detection efficiency of the energy spectrometer is obtained by measuring the standard material, when the activated carbon samples to be detected and the standard material are in the radioactive equilibrium, the 0.609MeV peak counts can be obtained by the γ-ray energy spectrum of activated carbon, and the activity of radon (i.e., the radon adsorption quantity) adsorbed by activated carbon can be calculated by the formula (1):

\[ A_s = A_\text{Ra} \times \left( \frac{N_s}{N_\text{Ra}} \right) \]  

In the formula: \( A_s \) is the activity of radon, \( A_\text{Ra} \) is the activity of the standard material \(^{226}\text{Ra},312\text{Bq}\), \( N_\text{Ra} \) is the net counts of 0.609MeV peak of the standard material, \( N_s \) is the net counts of 0.609MeV peak of activated carbon after it adsors the radon.

In order to obtain the net counts of 0.609MeV peak, the background of gamma spectrum is subtracted by the algorithm of Sensitive nonlinear iterative peak (SNIP). Before applying SNIP algorithm, the LLS (twice log operators plus square-root operator) operator is used to transform each energy spectrum count with the formula (2):

\[ V(i) = \log[10\left(\sqrt{y(i)} + 1\right) + 1] \]

Where i represents the channel, \( y(i) \) are the energy counts in the channel i, \( V(i) \) is the saving vector after \( y(i) \) is transformed by LLS operator, then \( V1(i) \), \( V2(i) \) are calculated up to \( Vm(i) \) step by step,
and the parameter of \( m \) is generally the FWHM. The new value in the channel \( i \) in the \( p \)-th iteration is obtained by the formula (3):

\[
V_p(i) = \min \left\{ \frac{1}{2} \cdot \left[ V_p(i - p) + V_p(i + p) \right], V_p(i) \right\}
\]

(3)

Where \( p \) grows iteratively from 1 to a given \( m \) value with step size 1. When \( V_m(i) \) is calculated, the net value of the spectrum can be obtained by performing the inverse calculation of LLS operator.

After background subtraction, the spectrum needs to be smoothed and denoised before calculating the activity of radon. And the spectrum is denoised by the denoising method of Least squares support vector machine (LS-SVM). In the method, the spectrum was segmented according to the principle of three-window method, and the method can be described by the formula (4):

\[
Y(i) = \begin{cases} 
  y_1(i); & 1 \leq i < 34 - t \\
  y_2(i); & 34 + t < i < 114 - t \\
  y_3(i); & 114 + t < i < 256 \\
  k_j \cdot y_1(i) + \overline{k_j} \cdot y_2(i); & 34 - t \leq i \leq 34 + t \\
  k_j \cdot y_2(i) + \overline{k_j} \cdot y_3(i); & 114 - t \leq i \leq 114 + t
\end{cases}
\]

(4)

Where \( i \) is the spectrum channel, \( y_1, y_2, \) and \( y_3 \) are the results of spectrum three-section regression fitting, the channel distribution of the three-energy segment is \((1 \sim 34 + t), (34 + t \sim 114 + t), \) and \((114 + t \sim 256)\), respectively. The overlapping channel address of the spectrum is \(2t + 1\). \( k_j \) and \(-k_j\) are the smoothing weighting factors and their mathematical expressions are described in formula (5):

\[
k_j = \frac{1}{2} + \frac{1}{2} \sin\left( \frac{j - 1}{2t} \pi + \frac{\pi}{2} \right) j = 1, 2, \ldots, 2t + 1
\]

(5)

\[
\overline{k_j} = \frac{1}{2} + \frac{1}{2} \sin\left( \frac{j - 1}{2t} \pi + \frac{\pi}{2} \right) j = 1, 2, \ldots, 2t + 1
\]

Where \( j \) states the position number of the spectrum in the overlapped data segment. The net counts of 0.609 Mev can be obtained by the spectrum that was eliminated noise, and then the activity of radon can be calculated by the net counts.

5. Results and discussion

When Guo Liangtian et al. [14] explored the influence of the gas superficial velocity on the adsorption coefficient of krypton of activated carbon, the gas superficial velocity was controlled in the range of 0.1cm/s-5cm/s when this gas flowed through the activated carbon adsorption bed. Radon and krypton belong to the same family elements, and their physical properties are similar. When studying the influence of the gas superficial velocity on the radon adsorption of activated carbon, based on the experimental conditions of predecessors and the existing experimental instruments, the experimentally selected gas superficial velocity is controlled in the range of 1cm/s-3cm/s, the radon concentration in radon chamber is set to 1000Bq/m³, the adsorption time of activated carbon is 60min, and the measurement time is 60min. The influence of the gas superficial velocity on the radon adsorption of activated carbon is analyzed mainly by the radon adsorption quantity of the activated carbon bed. The radon adsorption experiments were carried out on the coconut shell, apricot shell and coal-based activated carbon under the adsorption conditions of different gas superficial velocities respectively, and the radon adsorption quantity was calculated. The results are shown in Table 1.
Table 1. Radon adsorption measurement results of coconut shell activated carbon at different gas superficial velocities

| Gas superficial velocity /cm·s⁻¹ | The radioactivity of radon /Bq |
|---------------------------------|-------------------------------|
| Coconut shell | Apricot shell | Coal-based | Coconut shell | Apricot shell | Coal-based |
| 0.98 | 1.10 | 0.91 | 57.01 | 66.01 | 16.12 |
| 1.10 | 1.26 | 1.01 | 77.11 | 65.19 | 22.43 |
| 1.27 | 1.35 | 1.28 | 76.52 | 61.22 | 19.51 |
| 1.55 | 1.46 | 1.64 | 45.80 | 51.41 | 17.41 |
| 1.94 | 1.63 | 1.69 | 75.71 | 42.18 | 17.06 |
| 2.10 | 1.93 | 1.91 | 86.80 | 61.34 | 19.63 |
| 2.35 | 2.12 | 2.15 | 151.76 | 60.99 | 20.33 |
| 2.69 | 2.45 | 2.24 | 166.02 | 80.03 | 18.11 |
| 2.83 | 2.64 | 2.49 | 160.88 | 70.57 | 23.37 |
| 2.90 | 3.10 | 2.72 | 118.93 | 71.38 | 17.76 |
| 2.98 | —— | 2.74 | 145.34 | —— | 18.69 |
| 3.01 | —— | 2.79 | 121.27 | —— | 19.16 |
| —— | —— | 3.08 | —— | —— | 18.34 |

The radon adsorption quantity of three kinds of activated carbon under different gas superficial velocities is shown in Figure 4. When the gas superficial velocity is in the range of 0.9cm/s-2.0cm/s, the changes of the radon adsorption quantity of coconut shell activated carbon and apricot shell activated carbon are roughly the same and at the same level. When the gas superficial velocity is in the range of 2.0cm/s-3.1cm/s, the radon adsorption quantity of coconut shell activated carbon is significantly higher than other two kinds of activated carbon. The radon adsorption quantity of coconut shell activated carbon reaches its peak when the gas superficial velocity is 2.7cm/s. The radon adsorption quantity of coal-based activated carbon has no obvious change under different gas superficial velocity, and the radon adsorption quantity is much smaller than the others.

When the gas superficial velocity is more than 2.7cm/s, the radon adsorption quantity of coconut shell activated carbon shows a downward trend. The reason for this change may be that after the gas superficial velocity increases to a certain value, the resistance of external diffusion material begins to decrease, and the adsorbed molecules are separated from the active center of the surface of the adsorbent, causing a part of the radon atoms adsorbed by activated carbon to be desorbed [15], and resulting in a decrease in the radon adsorption quantity.
6. Conclusion

Through radon adsorption experiments of the activated carbon bed in radon chamber and the measurement of activated carbon samples by the HPGe $\gamma$-ray spectrometer with high resolution, the conclusions of the analysis are as follows: (1) When the gas superficial velocity is 0.9 cm/s-3.1 cm/s, the radon adsorption capacity of three kinds of activated carbon is: coconut shell activated carbon $>$ apricot shell activated carbon $>$ coal-based activated carbon, the radon adsorption capacity of coal-based activated carbon is low, so it is not recommended to use. (2) Among the three kinds of activated carbon, the radon adsorption capacity of coconut shell activated carbon is greatly affected by the gas superficial velocity, and the radon adsorption capacity of coal-based activated carbon is basically not affected by the gas superficial velocity. (3) When the gas superficial velocity is in the range of 0.9 cm/s-3.1 cm/s, the optimal gas superficial velocity that the coconut shell activated carbon absorbs radon is 2.7 cm/s. The above conclusions have certain reference significance for the selection of activated carbon and the setting of the gas superficial velocity in the practical application of the radon adsorption of activated carbon.

When the gas superficial velocity is between 1.5 cm/s and 1.8 cm/s, the radon adsorption quantity of the three kinds of activated carbons presents the V-shaped fluctuation in varying degrees, and the measurement results of repeated experiments are the same as those of the previous ones at this gas superficial velocity, the abnormal results caused by improper operation in the experiment are excluded. There is no clear explanation for the fluctuation of the radon adsorption quantity of activated carbon at the gas superficial velocity, which needs to be further explored.

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