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NUCLEAR SCIENCE AND TECHNIQUES

Nuclear Science and Techniques, Vol.17, No.2 (2006) 92–96

Calculation and measurement of migration coefficient of radon under laboratory conditions

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Abstract Under laboratory conditions (i.e. a sealed system in which the temperature may vary according to air temperature), the migration of radon in upward, downward and horizontal directions has been investigated. After a period of accumulation, the spatial distribution profile of radon was drawn on the basis of the experimental data. The profile showed whorl-shape with bigger ends. The longer the accumulation time, the bigger the whorl end, and the higher the radon concentration is. By fitting the experimental data by least-square statistical method, we find that the distributions of radon follow negative exponential functions in the upward, downward and horizontal directions. However, exponents for the three directions are not exactly identical. The upward migration is more effective than the downward one and both upward and downward migrations are more effective than the horizontal one.

Key words Radon, Migration coefficient, Diffusion coefficient

CLC number P631.6

1 Introduction

The migration of radon has been widely studied in the fields of earth and environmental sciences since the discovery of radon in 1898. In the early 1939, Flügge and Zimens[1] thought that the migration mechanism of radon was diffusion, following Fick's diffusion law. Until now, this is still the best-known mechanism of radon migration. With the extensive applications and further investigation of radon measurement, researchers found that diffusion alone cannot explain the long-distance migration of radon in the environment.[2-4] The physical conditions of rock, soil and atmosphere such as temperature and pH value may affect the migration of radon. To study the migration of radon in the building, we designed experimental equipment (a sealed system in which the temperature may vary according to air temperature). The results show that the diffusion law is not enough to explain the migration of radon under laboratory conditions or the migration in nature[5,6]. In this paper, we studied the migration of radon under laboratory conditions. The migration coefficients of radon in upward, downward and horizontal directions are calculated, respectively, on the basis of the experimental data.

2 Basic theory of describing the migration of radon

The diffusive movement of radon is described by Fick's law:

$$N = N_0 \mathrm{e}^{-\sqrt{\lambda_{\mathrm{Rn}} / D_{\mathrm{m}}} \cdot x} \qquad (1)$$

where N is the radon concentration at the distance x,

Supported by the Bureau of Science and Technology of Fujian Province, China (No. 2003J010) and National Natural Science Foundation of China (No.10575022)

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Received date: 2005-10-27

N_0 is the radon concentration at the source center, λ_{Rn} is decay constant of radon and D_m ($cm^2 \cdot s^{-1}$) is the molecular diffusion coefficient of radon in the air.

The molecular diffusion coefficient of radon in the air is $D_m = 0.1 cm^2 s^{-1}$ [3,7], and the decay constant of radon is $\lambda_{Rn} = 2.1 \times 10^{-6} s^{-1}$. Thus, Eq. (1) becomes

$$N = N_0 e^{-0.00458x} \quad (2)$$

Using Fick's law we obtain

$$N = N_0 e^{-\sqrt{\lambda_{Rn}/p} \cdot x} \quad (3)$$

where the definitions of all parameters are the same as Eq. (1), except for *p*. Here *p* is defined as the migration coefficient related with the migration capability of radon and its progeny. Taking the logarithm on both sides of Eq. (3)

$$\ln N = \ln N_0 - \sqrt{\lambda_{Rn}/p} \cdot x \quad (4)$$

and let $y = \ln N, a = \ln N_0, b = -\sqrt{\lambda_{Rn}/p}$, a reorganized form of Eq. (4) is created:

$$y = a + bx \quad (5)$$

where *a* and *b* are unknown, but they can be calculated by using the least-squares method, if a group of experimental data y and x are available. Then *p* can be computed.

3 Experimental devices and methods

For the purpose of keeping radon migration in the three directions under the same conditions (e.g. room temperature, normal pressure and static state), a device was designed as shown in Fig. 1. The ^{226}Ra source was placed in the center of the pipe. Each time, before we put the source in, the pipe was cleaned. The active sorbents were placed at the points of interest in the pipe. Then the top, bottom and horizontal entries of the PVC pipe were sealed.

A total of 30 mL of standard liquid ^{226}Ra source, with 1.32×10^{-8} g of ^{226}Ra, was used. Sorbent is a kind of active carbon. A bag of sorbent contained 12 g of active carbon.

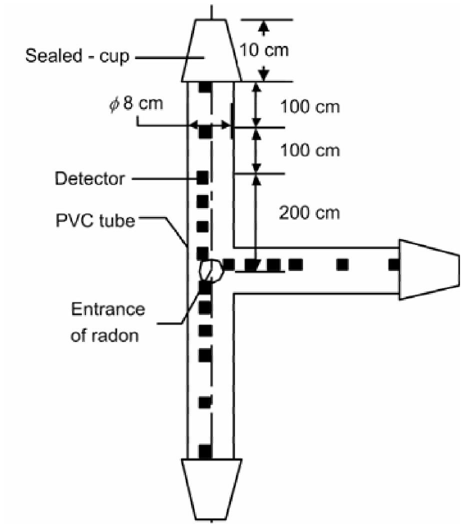


Fig.1 The sketch of device for measuring radon and its progenies' migrations upwards, downwards and horizontally.

Gamma measurement method was applied to measure the radioactivity of radon and its progenies which are absorbed by the sorbent. High-resolution Ge(Li) semi-conductor detector (FWHM = 2 keV at 1333 keV), and 4096 channel ADC pulse height analyzer (made in Shanghai Electronic Instrument Factory) were used. The statistical error is 0.5% when the measurement time is 3000 s. To decrease the background counts, the sorbent was shielded by lead during measurements.

Active carbon sorbents were placed in the sealed PVC pipe for 7 and 19 days, respectively. Then they were taken out and each bag of sorbent was put into a separate glass container so that counts of each bag for 3000 s interval can be obtained.

The gamma counts of the active carbon sorbents are proportional to the concentration of radon. Hence, the gamma counts represent the relative intensity of radon. The counts corresponding to energy peaks at 352 and 609 keV were contributed by ^{214}Pb and ^{214}Bi, respectively. The total counts, counts at peak 352 and 609 keV, followed the same distribution. Therefore, we mainly discuss total counts.

4 Results and discussion

Total counts of each bag of sorbent without background are listed in Table 1.

Table 1 The experimental data on migration of radon and its progenies in PVC pipe

Sorbent position	7 days accumulation			19 days accumulation		
	N_{total}	$N_{352\ keV}$	$N_{609\ keV}$	N_{total}	$N_{352\ keV}$	$N_{609\ keV}$
Up 400 cm	2905±15	164±1	182±1	7795±40	1010±6	711±4
Up 300 cm	3379±17	234±2	175±1	9440±48	1294±7	892±5
Up 200 cm	5543±28	704±4	386±2	11230±57	1430±8	937±5
Up 150 cm	6304±32	773±4	471±3	11884±60	1404±8	869±5
Up 100 cm	8436±43	1038±6	727±4	16594±83	2037±11	1332±7
Up 50cm	10840±55	1120±6	708±4	13497±68	2054±11	911±5
Down 50 cm	6013±30	569±3	724±4	12267±62	1897±10	1242±7
Down 100 cm	5807±29	568±3	379±2	10086±51	1417±8	801±4
Down 150 cm	3364±17	370±2	325±2	10357±52	1002±6	702±4
Down 200 cm	1928±10	180±1	156±1	7685±39	1024±6	597±3
Down 300 cm	2111±11	234±2	161±1	8368±42	1252±7	764±4
Down 400 cm	808±4	70±1	78±1	2652±14	463±3	260±2
Horizon 50 cm	6305±32	556±3	478±3	8099±41	1030±6	753±4
Horizon 100 cm	2582±13	303±2	88±1	4533±23	454±3	304±2
Horizon 150 cm	180±1	27±1	57±1	2201±11	213±2	106±1
Horizon 200 cm	378±2	41±1	0±1	2147±11	160±1	109±1
Horizon 300 cm	133±1	0±1	20±1	1414±8	55±1	61±1
Horizon 400 cm	34±1	13±1	0±1	667±4	84±1	2±1

Note: N_{total} is the total counts without background. $N_{352\ keV}$ is the 352 keV peak counts, without background counts. $N_{609\ keV}$ is the 609 keV peak counts without background.

According to the vertical and horizontal migration data (see Table 1) and migration rule of radon, the spatial migration distribution of a point-shaped radon source can be inferred. The 2D distribution of radon migration in the 400 cm PVC pipe is shown in Fig. 2. The point (0,0) is the location of the radium source. We can see that the distribution is whorl-shaped. The longer the accumulation time, the bigger the end of the whorl is, and the higher the radon concentration is.

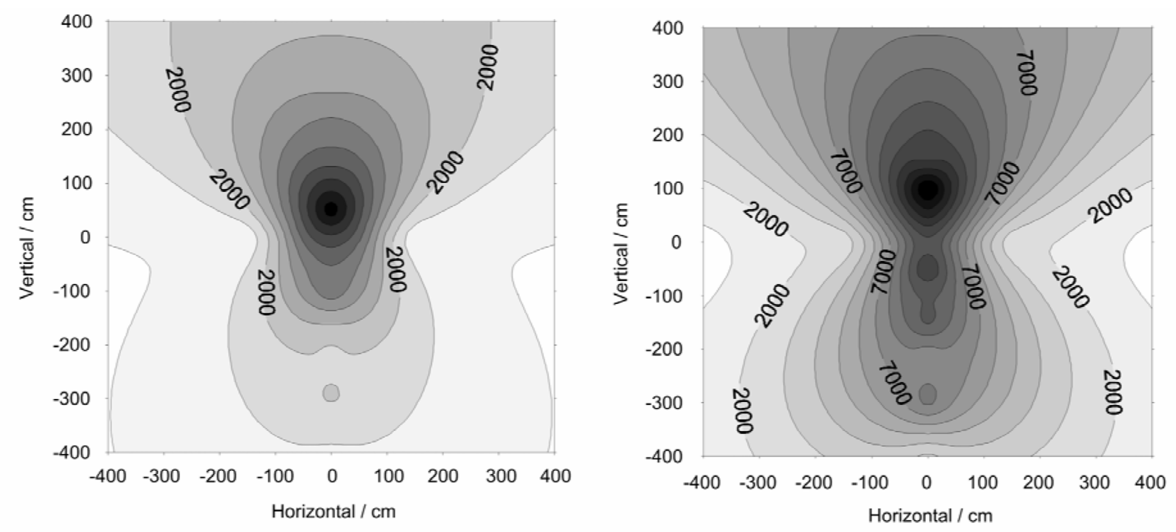
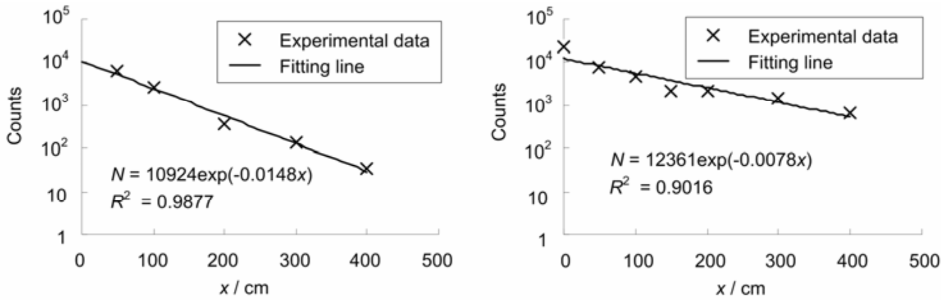


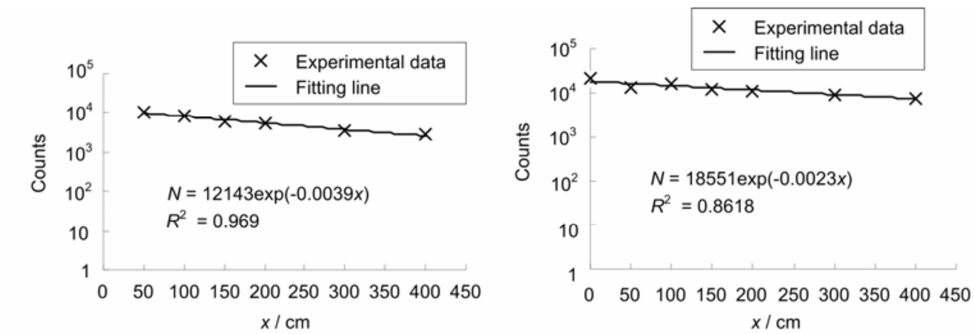
Fig.2 2D distribution of ^{222}Rn concentration after (a) 7-day and (b) 9-day accumulation.

Fitting the up, down and horizontally directed distribution data with the least-squares method, the regression equations can be obtained (see Figs. 3-5). Supposing the confidence level is 0.05, all the correlation coefficients between fitted values and experimental data are greater than their critical values. From the



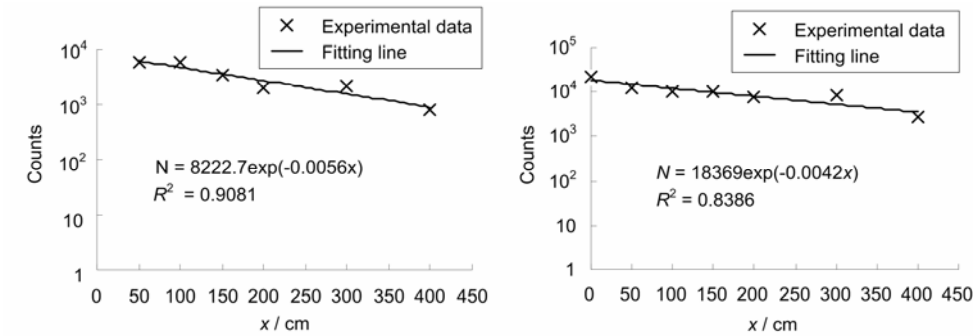
(a) (b)

Fig.3 (a) Least-squares fit plot of ^{222}Rn counts in the horizontal direction after 7 day accumulation; (b) Least-squares fit plot of ^{222}Rn counts in the horizontal direction after 19 day accumulation.



(a) (b)

Fig.4 (a) Least-squares fit plot of ^{222}Rn counts in the upward direction after 7 day accumulation; (b) Least-squares fit plot of ^{222}Rn counts in the upward direction after 19 day accumulation.



(a) (b)

Fig. 5 (a) Least-squares fit plot of ^{222}Rn counts in the downward direction after 7 day accumulation; (b) Least-squares fit plot of ^{222}Rn counts in the downward direction after 19 day accumulation.

statistical point of view, the regression equation is significant.

According to Eq. (4), the slope in the fitting equation is $\sqrt{\lambda_{\mathrm{Rn}} / p}$, hence the migration parameter p can be derived for the three directions, respectively. Their migration coefficients are summarized in Table 2.

In this experiment, although the migration conditions of radon in three directions were the same, the migration coefficients showed much difference. It is shown that the value of the upward migration coefficient is greater than that of the downward one; the upward and the downward are greater than the horizontal ones. In the case of 19 day accumulation, the upward migration coefficient was 0.397 $cm^2 \cdot s^{-1}$, not only greater than the diffusion parameter of radon in the air (0.1 $cm^2 \cdot s^{-1}$), but also greater than the diffusion parameters of radon progenies (0.03—0.085 $cm^2 \cdot s^{-1}$)[8]. But the down migration coefficient (0.119 $cm^2 \cdot s^{-1}$) was slightly greater than diffusion coefficient (0.1 $cm^2 \cdot s^{-1}$). The horizontal migration coefficient (0.0345 $cm^2 \cdot s^{-1}$) is less than the horizontal diffusion coefficient (0.1 $cm^2 \cdot s^{-1}$). In our experiment, the accumulation time was not long enough, so that the radioactive equilibrium

Table 2 Fitting data of radon and its progenies and migration coefficients in different directions

Time / d	Migration direction	$\sqrt{\lambda_{Rn} / p}$	Migration coefficient / $cm^2 \cdot s^{-1}$
7	Upward	0.0039	0.1381
	Downward	0.0056	0.0670
	Horizontal	0.0148	0.0096
19	Upward	0.0023	0.3970
	Downward	0.0042	0.1190
	Horizontal	0.0078	0.0345

was not achieved, which caused the differentia of migration coefficient between 7 and 19 day accumulation. If the accumulation time is long enough (e.g. 38 days or more), the migration coefficient will be a constant.

The migration of radon was under room temperature and normal pressure in the sealed devices. If there exists a little difference in temperature in vertical direction, the upward migration coefficient should be equal to the downward migration coefficient. But the experimental results showed the upward migration coefficient was far greater than the downward migration coefficient. Radon possesses the ability to rise although the specific gravity of radon is higher than the air. The following is one of the explanations. "After α-particles emit from radon and its progenies decelerate, they become He nuclei. The He nuclei can combine with radon and its progeny and form clusters. Because He is very light, it can make the clusters' gravity less than the buoyancy in the medium. As a result, the clusters will rise of their own accord."[5,6]

5 Conclusion

Migration coefficients of radon are more precise than are diffusion coefficients. On the basis of our experiment, the migration of radon is characterized by directional orientation. Hence, one-value diffusion coefficient cannot accurately describe the migration capability in different directions. There are different migration coefficients in different directions. The above-mentioned method can accurately calculate and measure these migration coefficients.

Acknowledgments

The authors appreciate the technical guidance and help of Prof. JIA Wenyi and FANG Fang from Chengdu University of Technology.

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