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Accurate measurement of the radon exhalation rate of building materials using the closed chamber method

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Abstract

The construction department of the Chinese government is now discussing the possibility of reducing indoor radon exposure through controlling the radon exhalation rate of building materials. However, which quantity is suitable for control and how to specify and measure this quantity are still not clear. A comparison of a theoretical model and measurement process of radon exhalation between the surface of soil and building materials was carried out. As a result, the so-called intrinsic exhalation rate is thought to be a suitable quantity for control. Through theoretical analysis and experimental comparisons of different pre-treatment methods, it was indicated that the closed chamber method could be used to measure the intrinsic exhalation rate of building materials. Measurement results show that the average intrinsic radon exhalation rate of building materials commonly used in Beijing is 4.891 mBq m^{-2} s^{-1}, with a range of 0.323–21.250 mBq m^{-2} s^{-1}, and the average diffusion length is 16.448 cm, with a range of 2.371–41.960 cm.

(Some figures may appear in colour only in the online journal)

1. Introduction

Radon (^{222}Rn) is a naturally existing radioactive gas with a half-life of 3.83 d, which can emanate from soil as well as building materials to indoor air. Radon decays into a series of short-life progeny (^{218}Po, ^{214}Pb and ^{214}Bi), which could be inhaled into the human respiratory tract and impose a significant health hazard on occupants. In tall buildings in large cities in China, the major contributor of indoor radon concentration is building materials. With new kinds of building material such as porous concrete and coal ash brick used in the newly built dwellings, the indoor radon concentration seems to be growing [1]. Recently, the construction department of the Chinese government has been discussing the possibility of reducing indoor

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radon exposure by controlling the radon exhalation rate from building materials. However, which quantity is suitable for control and how to measure this quantity accurately is still not clear.

The radon exhalation rate is specified as the liberation quantity of radon from the surface area of building materials per unit time (Bq m^{-2} s^{-1}). Using this value and the area of the indoor surface, we could easily calculate the radon emanated per unit time (Bq s^{-1}), which could be used to estimate the radon concentration in the indoor environment.

Various methods have been developed to measure the radon exhalation rate of building materials, such as the accumulation method first proposed by the Lawrence Berkeley Laboratory, the charcoal method and the SSNTD (solid state nuclear track detector) method [2–5]. The closed chamber method (CCM) proposed by Chao *et al* is essentially one kind of accumulation method which can take into account the leakage of the accumulation chamber and the back diffusion rate, so it is widely used in the measurement of the radon exhalation rate of building materials [6]. Due to a possible misunderstanding of the radon exhalation model, the CCM sometimes might be misused during measurements, especially in pre-treatments of building materials [7–9]. Chao discussed the difference between the one-dimensional model and the three-dimensional model by comparing experiments (sealing on four sides versus no sealing on surfaces), but no more theoretical analysis was given and no details on how to make different models consistent were shown [10].

In order to find a reasonable quantity for controlling the exhalation rate from building materials, this paper starts with a comparison of the radon exhalation progress/model between soil and building materials. Using theoretical analysis and experimental comparison of the one-dimensional model and three-dimensional model, especially dealing with different pre-treatments of CCMs, an accurate method for measuring this quantity was suggested. Discussions of the measuring methods and the factors influencing radon exhalation rate from building materials are also given. The radon exhalation rates of commonly used building materials in Beijing are also measured and reported.

2. Theoretical analysis

2.1. Intrinsic exhalation rate

Radon is generated from naturally existing radium and uranium by decay processes in soil or building materials. It then emanates to pore gases or fluids and migrates a significant distance from the site of generation to the surface of materials through diffusion or seepage; finally, a fraction enters the atmosphere before undergoing radioactive decay in an exhalation process. The magnitude of the exhalation progress is usually called the (surface) exhalation rate, which is defined as the number of radon atoms leaving unit surface area of the material per unit time (Bq m^{-2} s^{-1}), or the mass exhalation rate, which is defined as the number of radon atoms leaving unit mass of the material per unit time (Bq kg^{-1} s^{-1}). The former is more frequently used for evaluating the indoor radon concentration [11].

The radon exhalation progress from building materials is quite similar to that from soil. The basic theory of radon exhalation is nearly the same whether the radon comes from the soil or from a slab of building material. There remain some differences: for example, the soil's humidity is usually much higher than that of building materials, so the effect of seepage is much larger. Another difference is that the thickness of soil is naturally unlimited while that of building materials in a wall is limited, although it can be considered to extend indefinitely [12]. This difference results in the distributions of radon concentration in soil and building materials being quite different, as figure 1 shows.

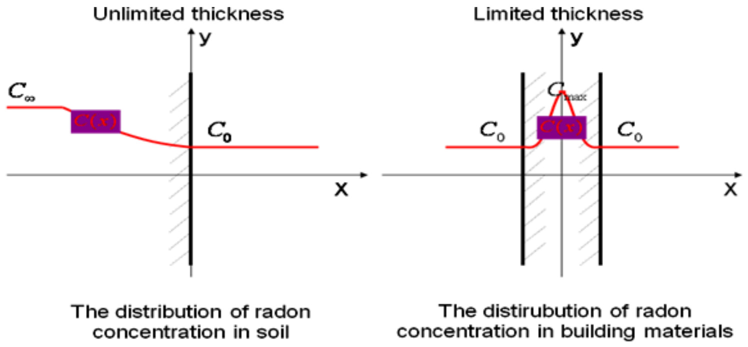

Figure 1. Diagram of radon concentration distributions in soil and building materials.

When radon diffuses out of soil or building materials, the surface exhalation rate can be calculated with

$$E = D_{\mathrm{air}} \frac{\partial C(x)}{\partial x}\bigg|_{x=0,\pm d/2} \tag{1}$$

where D_{air} is the diffusion coefficient of radon in air (m^2 s^{-1}), $C(x)$ is the radon concentration distribution in soil or in building materials (Bq m^{-3}), and $x = 0$ means the exhalation rate from soil, while $x = \pm d/2$ means the exhalation rate from the building materials in the wall. C_0 is the radon concentration in the air (Bq m^{-3}).

When $C_0 = 0$, which represents the intrinsic exhalation power (without being affected by outside radon concentration or soil thickness), then the exhalation rate of soil is usually called the ‘intrinsic exhalation rate’ of soil. In contrast, we could define the ‘intrinsic exhalation rate’ of building materials assuming the thickness of building materials in the wall is unlimited while extending indefinitely, and also the radon concentrations outside two surfaces of building materials are zero.

This quantity could represent the intrinsic exhalation power of building materials under certain conditions without being affected by the thickness of the wall or the outside environment. This quantity is influenced by the radium content, solid grain properties and pore space properties, as well as the temperature and water content of the building materials and so on. So it is quite suitable for controlling the radon exhalation rate of building materials. However, this quantity can not be measured directly, so how to measure it is a problem. The following will deal with this problem.

3. Experimental setup and theoretical approach

The most common situation is only a limited building material sample at hand, sometimes even in an anomalous shape, so it needs some pre-treatment so as to accord with the radon exhalation model in figure 1. The most commonly used method is to cut the sample into cuboids and seal with some radon tight materials [13]. If we seal four surfaces, the radon exhalation could be described by the limited thickness indefinite extent model as shown in the right diagram of figure 1. If we seal five surfaces, radon exhalation could be described by the unlimited thickness indefinite extent model as shown in the left diagram of figure 1. Without sealing the surfaces, radon could emanate from all of the surfaces and a three-dimensional model would need to be set up to calculate the radon exhalation rate.

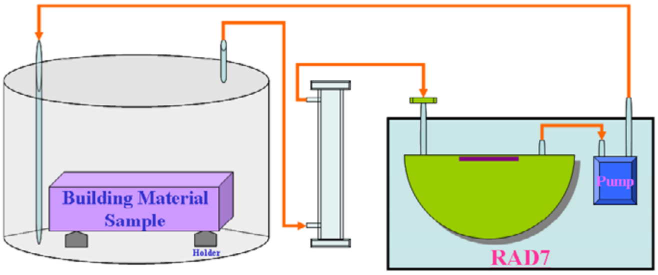

Figure 2. A diagram of our experimental setup.

The closed chamber method is commonly used for the measurement of the radon exhalation rate of building materials, because it can take into account the leakage of the chamber and the back diffusion of the building sample.

Here, to try to find an appropriate way to measure the intrinsic exhalation rate of building materials, a careful analysis with different theoretical approaches of different pre-treatments combined with the closed chamber method was carried out.

A diagram of our experimental setup is shown in figure 2.

The closed chamber is a stainless steel barrel with an effective volume of 16.150 l. The inlet and outlet are located on the top of the chamber with the inlet tube extended down to the bottom of the chamber. A quite small 24 V DC circulation fan is put at the bottom to keep radon mixing fully in the chamber. The lid of the chamber is wrapped with a gasket and then pinned by eight bolts. The leakage of the chamber as measured by the pure radon decay method is 169.51 $\mathrm{cm}^3\ \mathrm{h}^{-1}$, which can be reduced by nearly half after sealing with adhesive tape.

The building materials are cut into square shapes and sealed (four surfaces versus five surfaces versus no surfaces) with aluminum foil to let radon leave only from the uncovered surface. Then the building materials are left in an indoor room for at least one month to establish a balance between the building materials and the environment. After putting the building material samples on the holders of the chambers, the chambers are immediately purged with nitrogen gas with a flow rate of nearly 10 $\mathrm{l\ min}^{-1}$ for half an hour, then the radon levels inside the chambers are close to zero. The increasing radon concentrations in the chambers are measured by a RAD7 continuous radon monitor (Durridge), which was calibrated using the Standard Chinese Radon Chamber.

For the purpose of comparing different radon exhalation models, each sample ($a \times b \times c$, $b > c > a$) is sealed with three modes: one totally unsealed for the three-dimensional radon exhalation model; one sealing four surfaces leaving one pair of symmetrical surfaces for the one-dimensional limited thickness radon exhalation model; one sealing five surfaces leaving only one surface for the one-dimensional unlimited thickness radon exhalation model. Their diagrams are shown in figure 3.

The radon exhalation rate could be calculated using equation (1) when the radon distribution in the building materials is known. Under the steady state exhalation situation, the radon concentration distribution in a building material sample can be calculated from

$$-\lambda C(x,y,z) + \frac{C_{\mathrm{Ra}} S_{\mathrm{e}}}{\eta} + \frac{D}{\eta}\left(\frac{\partial^2}{\partial x^2} + \frac{\partial^2}{\partial y^2} + \frac{\partial^2}{\partial z^2}\right) C(x,y,z) = 0. \tag{2}$$

For one dimension, this equation can be simplified as

$$\frac{D}{\eta}\frac{\partial^2 C(x)}{\partial x^2} - \lambda C(x) + \frac{C_{\mathrm{Ra}} S_{\mathrm{e}}}{\eta} = 0 \tag{3}$$

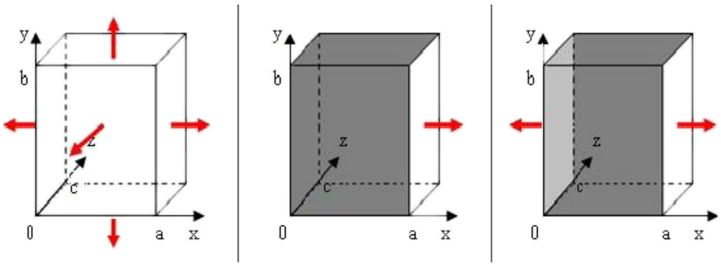

Figure 3. The diagrams of different sealing modes.

where $C(x, y, z)$ and $C(x)$ are respectively the three-dimensional and one-dimensional radon concentration distributions in building material (Bq m^{-3}) and D is the diffusion coefficient of radon in building materials ($\mathrm{m}^2\ \mathrm{s}^{-1}$). η is the porosity of the sample, C_{Ra} is the radium content (Bq m^{-3}) and S_{e} is the emanation coefficient.

When all surfaces are unsealed, we consider the boundary conditions:

$$C(x, y, z)|_{x=0,a;\,y=0,b;\,z=0;c} = C_0 \qquad \left.\frac{\partial C(x, y, z)}{\partial x}\right|_{x=0,a} = 0$$
$$\left.\frac{\partial C(x, y, z)}{\partial y}\right|_{y=0,b} = 0 \qquad \left.\frac{\partial C(x, y, z)}{\partial z}\right|_{z=0,b} = 0 \tag{4}$$

where C_0 is the radon concentration outside the building materials (Bq m^{-3}). In this situation, equation (2) hardly gives an analytical solution of the radon concentration distribution; it could only be solved by a numerical method, then the radon exhalation rate of this situation is too complicated to calculate. The relationship between the increasing radon concentration and the intrinsic exhalation rate is not easily established. Only when $a = b = c$ might we establish a relationship between them. This is nearly impossible for a real situation. So the three-dimensional model is not suitable for measurement of the radon exhalation rate of building materials.

For the one-dimensional model, there remain two modes. One has unlimited thickness just as the soil does; the other has limited thickness as do the building materials in the wall. Their boundary conditions are

$$C(x)|_{x=a} = C_0 \qquad \left.\frac{\partial C(x)}{\partial x}\right|_{x=a} = 0 \qquad \text{versus}$$
$$C(x)|_{x=0,a} = C_0 \qquad \left.\frac{\partial C(x)}{\partial x}\right|_{x=0,a} = 0. \tag{5}$$

From equations (3) and (4), the radon concentration in these two modes can be expressed in a unified form of equal as

$$C(x) = A\mathrm{e}^{-\sqrt{\frac{\lambda\eta}{D}}x} + B\mathrm{e}^{\sqrt{\frac{\lambda\eta}{D}}x} + \frac{C_{\mathrm{Ra}}S_{\mathrm{e}}}{\lambda\eta}. \tag{6}$$

However, these constants A and B have different values. For the unlimited thickness model and the limited thickness model, the A and B are separately expressed as

$$A = B = \frac{1}{2}\left(C_0 - \frac{C_{\mathrm{Ra}}S_{\mathrm{e}}}{\lambda\eta}\right)\cosh^{-1}\left(\sqrt{\frac{\lambda\eta}{D}}a\right)$$

for the unlimited thickness model (one surface exposed) (7)

$$A = \left(C_0 - \frac{C_{\mathrm{Ra}} S_{\mathrm{e}}}{\lambda\eta}\right) \Big/ (1 + \mathrm{e}^{\sqrt{\frac{\lambda\eta}{D}}a})$$

$$B = \left(C_0 - \frac{C_{\mathrm{Ra}} S_{\mathrm{e}}}{\lambda\eta}\right) \Big/ (1 - \mathrm{e}^{\sqrt{\frac{\lambda\eta}{D}}a})$$

for the limited thickness model (two surfaces exposed). (8)

Then we can easily calculate the radon exhalation rate of each surface of the sample using equation (1).

In a closed chamber, the equation describing the radon concentration inside the chamber can be expressed as follows [10]:

$$\frac{\mathrm{d}C_1(t)}{\mathrm{d}t} = -\lambda C_1(t) + \frac{(E_0 - \alpha C_1(t))S}{V} + \frac{q(C_1(t) - C_2)}{V} \tag{9}$$

where $C_1(t)$ is the radon concentration in the chamber at time t (Bq m^{-3}), C_2 is the radon concentration outside the chamber (Bq m^{-3}), which can be considered a constant during measurement, λ is the radon decay constant (s^{-1}), α is the back-diffusion rate (s^{-1}), q is the leakage rate of the chamber (m^3 s^{-1}), S is the area of the radon exhalation surface (m^2) and V is the effective volume of the chamber (m^3). E_0 is the previously defined intrinsic exhalation rate of the building material (Bq m^{-2} s^{-1}).

When the building material sample is in the chamber, $C_1(t) = C_0$ and also $C_1(0) = 0$. The net exhalation rate can be expressed in terms of the radon concentration gradient at the material surface:

$$E_0 - \alpha C_1(t) = -D \frac{\mathrm{d}C(x)}{\mathrm{d}x}\bigg|_{x=0;\, x=0,a}. \tag{10}$$

From the initial slope of the increase of radon concentration in the chamber, we can obtain the measured value of surface radon exhalation rate E_{meas} (Bq m^{-2} s^{-1}), which is defined by the slope divided by the total area of the radon exhalation surface.

Combining equations (6)–(10), we finally obtain the relationship of the measured value of surface radon exhalation rate and the intrinsic exhalation rate of the building material. The relationship of the two models is expressed as

$$E_{\mathrm{meas}}1 = E_0 \tanh(a/L) \qquad \text{for the unlimited thickness model (one surface exposed)} \tag{11}$$

$$E_{\mathrm{meas}}2 = E_0 \tanh(a/2L) \qquad \text{for the limited thickness model (two surfaces exposed)} \tag{12}$$

where $L = \sqrt{D/\lambda\eta}$ is the radon diffusion length in the material. The most interesting result is that $E_0 = \sqrt{\lambda\eta D} C_{\mathrm{Ra}} S_{\mathrm{e}}/\lambda\eta$, which is only influenced by the porosity, diffusion coefficient, radium content and emanation rate of the sample, and has nothing to do with the environmental parameters or measurement related parameters, and even has nothing to do with the thickness or shape of the building material sample! It therefore characterises an intrinsic property of the material and is suitable for reflecting the intrinsic difference between different building materials and for controlling the quality of different building materials.

Comparing equations (11) and (12) and assuming $R = E_{\mathrm{meas}}1/E_{\mathrm{meas}}2$, the radon diffusion length is obtained:

$$L = \frac{a}{\arccos h(\frac{1}{R-1})}. \tag{13}$$

Substituting the value of L into either equation (11) or (12), we finally obtain the intrinsic radon exhalation rate of building materials.

Table 1. The experimental results of different sealing methods.

		Measured value of the surface radon exhalation rate (mBq m^2 s^{-1})					
Building materials	Size (cm)	No sealing	Sealing on four surfaces	Sealing on five surfaces	R value	Intrinsic exhalation rate (mBq m^2 s^{-1})	Diffusion length (cm)
Porous concrete	$24^{\mathrm{a}} \times 15 \times 15$	0.074 ± 0.016	0.201 ± 0.100	0.342 ± 0.105	1.697	0.479 ± 0.238	16.748 ± 4.613
	$24 \times 15^{\mathrm{a}} \times 15$	0.069 ± 0.021	0.170 ± 0.091	0.312 ± 0.120	1.832	0.560 ± 0.216	23.835 ± 20.778
	$24 \times 15 \times 15^{\mathrm{a}}$	0.079 ± 0.018	0.186 ± 0.098	0.332 ± 0.134	1.785	0.543 ± 0.286	21.012 ± 12.719
Lightweight brick	$10^{\mathrm{a}} \times 10 \times 10$	0.437 ± 0.147	1.395 ± 0.110	2.535 ± 0.330	1.818	4.405 ± 0.574	15.251 ± 2.782
	$10 \times 10^{\mathrm{a}} \times 10$	0.429 ± 0.155	1.279 ± 0.413	2.189 ± 0.711	1.709	4.067 ± 1.009	11.400 ± 2.658

[a] Using this value as the x-axis direction.

4. Experimental measurements

4.1. Comparison of different sealing methods

In order to compare the three sealing methods and verify the intrinsic radon exhalation rate measurement method, we carried out a series of comparison experiments. Two building materials were chosen for these experiments—porous concrete and lightweight brick, which are commonly used in the buildings of Beijing, China. Each building material sample was cut into cuboids and sealed by the above methods—first without sealing, second sealing on four surfaces and third sealing on five surfaces. The porous concrete has a size of 24 cm × 15 cm × 15 cm and the lightweight brick has a size of 10 cm × 10 cm × 10 cm. In order to exclude the influence of sealing different surfaces, the porous concrete was sealed three times, each time with a different surface, while the lightweight brick was sealed twice, each time with a different surface. The experimental results are shown in table 1.

The measured value of the surface radon exhalation rate shows that the different sealing methods give quite different results. The no sealing situation usually has a rather smaller value than for sealing on four surfaces and on five surfaces, mainly because in this situation the radon exhalation surface S is much larger than the others. The results for the same building materials with no sealing are nearly the same, which is reasonable. The R values are between 1.697 and 1.832, just because the thickness a is usually smaller than the diffusion length L, which causes the R values to be distributed from 1.648 (when $a = L$) to 2.0.

Using different exhalation surfaces, the calculated intrinsic exhalation rates of the porous concrete as well as the lightweight brick are nearly the same, although the surface radon exhalation rates are quite different, and also the diffusion lengths are quite similar, which confirms that the intrinsic exhalation rate and the diffusion length are inherent characteristics of the materials, and have nothing to do with the shape, measurement method or outside environmental parameters. Moreover, the coincident results indicate that the closed chamber method is suitable for measuring the intrinsic exhalation rate of building materials after appropriate cutting and sealing of samples.

4.2. Exhalation rates of different building materials

Few radon exhalation rates of building materials used in Beijing have been published before, so we collected some building materials commonly used in Beijing's buildings and measured their intrinsic exhalation rates and diffusion lengths. The survey results are listed in table 2.

Table 2. The exhalation rates of some building materials in Beijing.

Building materials	Numbers of samples	Intrinsic exhalation rate (mBq m^2 s^{-1})	Diffusion length (cm)
Concrete	3	0.803 (0.323–1.218)	4.905 (2.371–8.844)
Concrete with cinder	4	11.302 (6.577–21.250)	13.621 (3.370–21.490)
Porous concrete	3	0.571 (0.479–0.560)	22.424 (16.749–21.012)
Lightweight brick	4	5.927 (4.067–8.627)	25.218 (11.400–41.960)
Black soil brick	1	0.325 ± 0.043	9.380 ± 0.257

Measurement results show that the intrinsic radon exhalation rates of building materials commonly used in Beijing have an average of 4.891 mBq m^{-2} s^{-1}, with a range of 0.323–21.250 mBq m^{-2} s^{-1}, and the diffusion lengths have an average value of 16.448 cm, with a range of 2.371–41.960 cm.

The exhalation rates of concretes mixed with cinder are obviously higher than those of other concretes, which is because the radium is concentrated during coal burning and remains in the cinder. The radon exhalation rates of porous concrete have no significant difference from common concrete, which is because the intrinsic exhalation rate is not only influenced by the porosity but also by the radium content. The exhalation rate of lightweight brick is higher than that of ordinary concrete, which is mainly affected by the porosity. The black soil brick has the lowest intrinsic radon exhalation rate. The diffusion lengths of porous concrete and lightweight brick are obviously higher than that of ordinary concrete, which is also the effect of porosity.

Ignoring the indoor fitment changes, the indoor radon concentrations might increase with use of lightweight brick and concrete with cinder on a large scale. Poor ventilation with air-conditioning systems more and more popularly used in tall buildings will strengthen the increase.

5. Conclusion

This paper deals with finding an appropriate quantity for controlling the radon exhalation of building materials and pointing out a reasonable method to measure this quantity. Through comparing the radon exhalation models of soil and building materials, we find that the intrinsic exhalation rate is suitable for control. Through theoretical analysis and experimental comparison of different pre-treatment methods, we find that, after appropriate cutting and sealing of samples, the closed chamber method can be used to measure the intrinsic exhalation rate of building materials. Using this method to measure the building materials commonly used in Beijing, we obtained the results that the intrinsic radon exhalation rates of these building materials have an average of 4.891 mBq m^{-2} s^{-1}, with a range of 0.323–21.250 mBq m^{-2} s^{-1}, and the diffusion lengths have an average value of 16.448 cm and a range of 2.371–41.960 cm.

The intrinsic exhalation rate of building materials is one of the inherent characteristics of these materials, and is influenced by the radium content, emanation coefficient and diffusion coefficient of radon in materials. It can therefore be used for controlling the radon exhalation rate of different materials no matter what the shape or environmental parameters. However, this does not mean that the intrinsic exhalation rate is not influenced by the environment, because the environmental temperature and humidity impact on the emanation coefficient and diffusion coefficient of building materials and thus influence the intrinsic exhalation rate. The intrinsic exhalation rates therefore only make sense in certain conditions. However, this will not affect the comparison of different building materials using this quantity, because we can set a standard for the environmental temperature and humidity when measuring the intrinsic exhalation rate.

If we know the intrinsic exhalation rate and diffusion length of each kind of building material, then we can easily calculate the surface radon exhalation rate when these materials are built into a wall, while the surface radon exhalation rate will usually be used to roughly evaluate maximum indoor radon concentrations. More details such as cover effects and ventilation should be made clear if we want to make more precise evaluations.

Acknowledgment

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