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Determination of Radon Emanation and Back Diffusion Characteristics of Building Materials in Small Chamber Tests

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A methodology has been established to estimate the radon emanation rates and back diffusion rates of different building materials in small chamber tests. The two parameters, emanation rate and back diffusion rate, can be obtained from the growth curve of the chamber radon activity. Small impervious containers with a volume of 14 900 c.c. were used in the measurement and solid-state radon detectors were used to monitor the chamber radon levels continuously. In the study, the building materials were left inside the chamber for more than 400 hours in order to reach equilibrium radon concentrations. The emanation rates and the back diffusion rates of the samples were found to be fairly constant under the experimental conditions. The emanation rate was determined by the initial slope of the growth curve and the back diffusion rate was determined by the equilibrium value of the activity curve. Leakage of the chamber was estimated by checking the radon free decay rate inside the chamber before each experiment and a correction was included in the calculation. Two kinds of granite and one bare concrete were selected as test samples. The first granite sample had a mean emanation rate of 13.44 $Bq \cdot m^{-2} \cdot hr^{-1}$ *and a back diffusion rate of* $6.89 \times 10^{-4}\,hr^{-1}$. *The second granite sample had an emanation rate of* 0.58 $Bq \cdot m^{-2} \cdot hr^{-1}$ *with a back diffusion rate of* $5.40 \times 10^{-3} \cdot hr^{-1}$. *The bare concrete had an emanation rate of* 8.69 $Bq \cdot m^{-2} \cdot hr^{-1}$ *and a back diffusion rate of* $6.18 \times 10^{-3}\,hr^{-1}$. *The methodology has been found to provide an accurate method of estimating radon emanation and back diffusion characteristics of commonly used building materials.* © 1997 *Elsevier Science Ltd.*

NOMENCLATURE

C	radon concentration inside chamber at time t [$Bq \cdot m^{-3}$]
C_o	background mean radon concentration in laboratory [$Bq \cdot m^{-3}$]
C_i	initial radon concentration inside chamber [$Bq \cdot m^{-3}$]
$C(\infty)$	equilibrium radon concentration inside chamber [$Bq \cdot m^{-3}$]
λ	radon-222 decay constant ($7.553585 \times 10^{-3}\,hr^{-1}$) [$hr^{-1}$]
E_o	initial radon emanation rate of building material (at $C = 0$) [$Bq \cdot m^{-2} \cdot hr^{-1}$]
E	radon emanation rate of building material at concentration C [$Bq \cdot m^{-2} \cdot hr^{-1}$]
D	radon back diffusion rate of building material [hr^{-1}]
α	specific radon back diffusion coefficient of building material [$m \cdot hr^{-1}$]
A	surface area of the sample material [m^2]
V	effective volume of the test chamber [m^3]
q	chamber leakage rate [$m^3 \cdot hr^{-1}$]
t	time unit [hr]
M_e	initial slope of the radon growth curve inside chamber [$Bq \cdot m^{-3} \cdot hr^{-1}$]
C_b	initial concentration of radon inside chamber in leakage measurement [$Bq \cdot m^{-3}$]
C_f	ideal radon concentration inside chamber without building material inside (no leakage) [$Bq \cdot m^{-3}$]
C_L	non ideal radon concentration inside chamber without building material inside (with leakage) [$Bq \cdot m^{-3}$]
M_f	initial slope of the ideal radon concentration decay curve inside chamber, without building material inside (no leakage) [$Bq \cdot m^{-3} \cdot hr^{-1}$]
M_L	initial slope of the non ideal radon concentration decay curve inside chamber, without building material inside (with leakage) [$Bq \cdot m^{-3} \cdot hr^{-1}$]
ACH	(q/V) air exchange rate of the radon chamber [hr^{-1}]

1. INTRODUCTION

Granite and concrete are commonly used in buildings for various purposes such as finishing, decoration, etc. These natural materials contain ^{40}K, ^{235}U, ^{238}U, ^{226}Ra and ^{232}Th [1, 2]. All these tracer elements are radioactive. Radon and thoron are the decay products of the uranium and thorium series respectively. Thoron is one of the isotopes of radon. The existence of uranium and thorium in the building material means the chance of radon gas diffusing into the indoor environment, and there is a potential health hazard to occupants. Therefore, it is necessary to understand their radioactive properties, such as gamma radiation and α radiation. Gamma radiation is due to the existence of ^{40}K, ^{235}U and ^{226}Ra in the building material. In the indoor environment, radon comes from concrete walls, ceiling, floor, and building materials containing uranium and thorium series elements, or it is drawn into the building from the sur-

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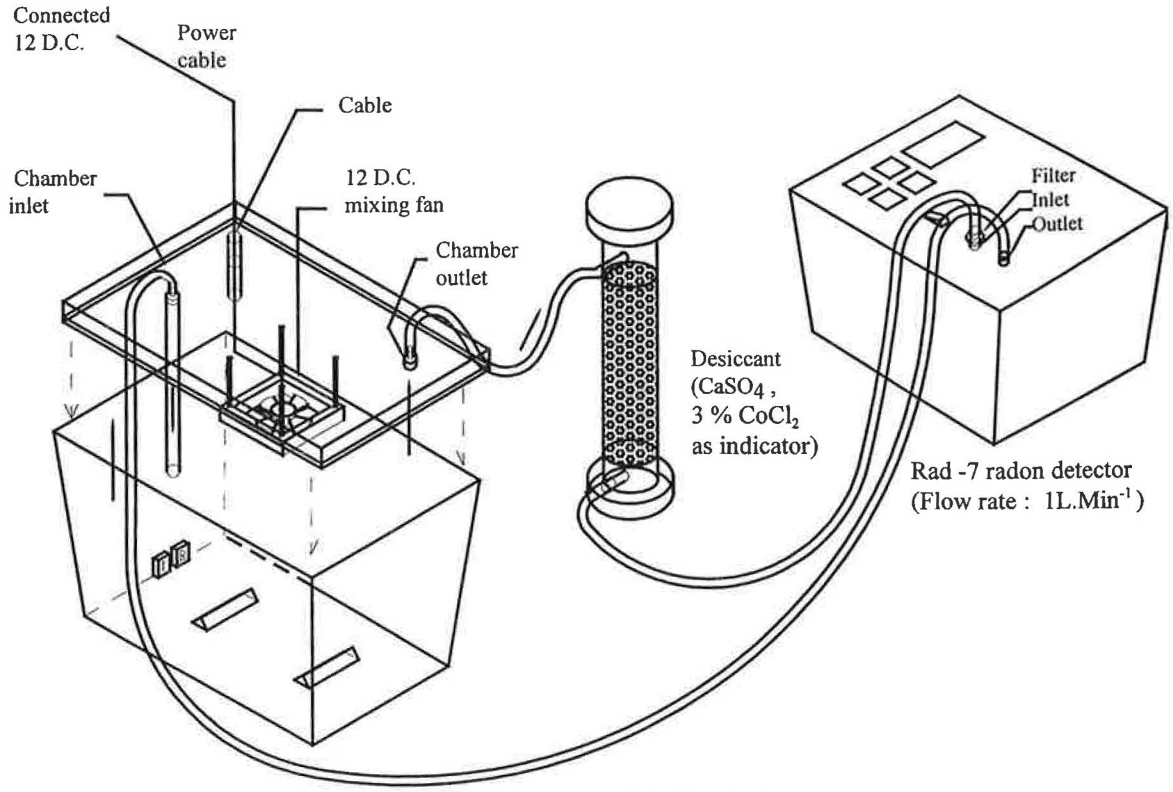

Fig. 1. Experimental set up of the chamber test.

rounding soil. Radon gas and its daughters can be inhaled into the respiratory system. As they decay in our lungs, they emit α-particles that can damage our lung tissue. Performing daily activities in a high radon level environment increases the risk of lung cancer. The Hong Kong Environmental Protection Department has set the guideline of indoor radon level at 200 $\mathrm{Bq \cdot m^{-3}}$.

Similar to many Asian cities, Hong Kong has a lot of high rise buildings using high radon emanation materials as interior finishing [3]. Study of radon emanation rates indicates that the radon risk due to the influence of building material is higher in Hong Kong than in countries like the US and European countries, where radon entry from soil turns out to be more dominant [4, 5].

Several radon emanation tests have been carried out by researchers [6–8] using chamber test techniques. In this study, a similar arrangement was used and a new calculation methodology was proposed taking into account both the back diffusion rate and the chamber leakage rate. Typical granites and concrete were selected for demonstration. These materials are popularly used in Hong Kong for construction and interior finishing.

2. EXPERIMENTAL SET UP AND THEORETICAL APPROACH

2.1. *Experimental set up*

Small vessels made of PVC were designed and used as radon chambers in our experiments. Figure 1 shows a schematic of the chamber. Each chamber is equipped with temperature and humidity sensors, and a small 12 V D.C. circulation fan (7-cm diameter and four-blade) for mixing purposes. The inlet and outlet are diagonally located on top of the chamber lid. The inlet tube is extended down to the bottom of the chamber to provide better circulation and mixing. The power cable conjunction of the mixing fan is sealed by silicone to prevent leakage. A gasket is wrapped around the rim of the chamber and is inserted between the lid and the main body of the chamber to reduce leakage. All the tubes connected to the chamber are sealed by vaseline at the intersection. The experiments were performed under dry conditions (R.H. was controlled at under 10%) and room temperature from 18 to 26°C. Measurement conditions are summarized in Table 1. Before starting the measurements, the chambers were purged by nitrogen gas with a flow rate of 4 l/min for more than 20 minutes. The radon level inside each chamber tended to zero before the test.

In this study, a solid state radon detector, Niton RAD-7, was used. The detector pulls samples of air through a fine inlet filter into a chamber for analysis. The filtered air decays inside the chamber, producing detectable alpha emitting progeny, particularly the polonium isotopes. The solid state detector converts α radiation directly to an electrical signal using an alpha spectrometry technique which is able to distinguish radon from thoron and signal from noise [9].

2.2. *Theoretical and experimental approach*

Mass balance analysis shows that the radon chamber activity can be expressed by the following equation:

$$\frac{\mathrm{d}C}{\mathrm{d}t} = -\lambda C - DC + \frac{E_0 A}{V} + \frac{q(C_0 - C)}{V}. \quad (1)$$

The first term on the right hand side represents the decay term and the last term represents the exchange of cham-

Table 1. Chamber temperature and relative humidity

Building material	Mean temperature (°C)	Conc. fluctuation due to temperature variation (%) (Appendix A)	Mean R.H. (%)
Granite-1 (chamber A)	21.6	0.92–1.60	1
Granite-1 (repeat measurement, chamber B)	21.0	0.93–1.75	1
Granite-2 (chamber B)	20.2	0.71–0.91	5
Concrete slab (chamber A)	22.5	0.76–1.12	20

ber and ambient laboratory radon due to chamber leakage.

The second and third terms imply that the radon emanation rate is not a constant. It has been found that a certain amount of radon in the air will get back to the building material [8, 10]. A parameter called back diffusion is defined here, showing the rate of radon getting back to the building material as radon level builds up in the air. E_o represents the radon emanation rate when $C = 0$. The radon emanation rate follows a linear relationship with the radon concentration inside the chamber and can be expressed by the following equation [8]:

$$E = E_o - \alpha C, \tag{2}$$

where $\alpha = \dfrac{DV}{A}$ (see Appendix B).

The coefficient, α, is defined as the material specific radon back diffusion coefficient. Radon back diffusion is induced by the existence of the concentration gradient across the material boundary. In equation (1), the influence of back diffusion is characterized by the back diffusion rate, D. Use of D in equation (1) is equivalent to use of mass transfer coefficient in Fick's law of mass diffusion.

Solving equation (1) and using initial conditions $t = 0$, $C = C_i$

$$C = \left(C_i - \frac{E_oA + qC_o}{V\left(\lambda + D + \frac{q}{V}\right)}\right) e^{-[\lambda + D + (q/V)]t} + \frac{E_oA + qC_o}{V\left(\lambda + D + \frac{q}{V}\right)}. \tag{3}$$

As the chamber is purged by nitrogen gas before the experiment, C_i tends to zero. Equation (3) can be simplified as

$$C = \left(\frac{E_oA + qC_o}{V\left(\lambda + D + \frac{q}{V}\right)}\right)\left(1 - e^{-[\lambda + D + (q/V)]t}\right). \tag{4}$$

Before the emanation rate of the building material, E_o, can be found, the system leakage rate, q, and radon back diffusion of the building material, D, have to be determined.

2.3. *Determination of the chamber leakage rate, q*

If the chamber is empty of any testing samples and the radon in the air inside the chamber undergoes natural decay with the initial concentration, C_i , the dynamic equation can be written as follows:

$$\frac{dC_L}{dt} = -\lambda C_L + \frac{qC_o}{V} - \frac{qC_L}{V}. \tag{5}$$

As $t \to 0$, $C_L(0) \to C_b$, where C_b is the initial radon concentration inside the chamber.

$$C_L = \left(C_b - \frac{qC_o}{V\left(\lambda + \frac{q}{V}\right)}\right) e^{-[\lambda + (q/V)]t} + \frac{qC_o}{V\left(\lambda + \frac{q}{V}\right)} \tag{6}$$

$$\frac{dC_L}{dt_{(t\to 0)}} = M_L = -\left(C_b - \frac{qC_o}{V\left(\lambda + \frac{q}{V}\right)}\right)\left(\lambda + \frac{q}{V}\right)$$

$$M_L = -\frac{qC_b}{V} - \lambda C_b + \frac{qC_o}{V}, \tag{7}$$

where M_L is the initial slope of the decay curve with chamber leakage.

In the ideal case where the chamber has zero leakage rate

$$C_f = C_b e^{-\lambda t} \tag{8}$$

$$\frac{dC_f}{dt_{(t\to 0)}} = M_f = -\lambda C_b, \tag{9}$$

where C_f is the concentration of the chamber at time t without building material inside and M_f is the initial slope of the decay curve without leakage. To compare the initial slopes in equation (7) and equation (9)

$$M_f - M_L = (C_b - C_o)\frac{q}{V}$$

$$\frac{q}{V} = \frac{M_f - M_L}{(C_b - C_o)}$$

$$q = V\left[\frac{M_f - M_L}{(C_b - C_o)}\right]. \tag{10}$$

From equation (10), it can be seen that the chamber leakage rate, q, can be determined by measuring the two initial slopes of radon decay.

2.4. *Determination of the radon emanation rate*

In order to keep the initial radon activity inside the chamber as low as possible, the chamber is purged by pure nitrogen gas before the measurement. The initial radon concentration of the chamber will be close to zero. Thus the chamber concentration growth curve can be represented by equation (4). Taking the differential to the equation and letting time equal zero, equation (11) and equation (12) are obtained.

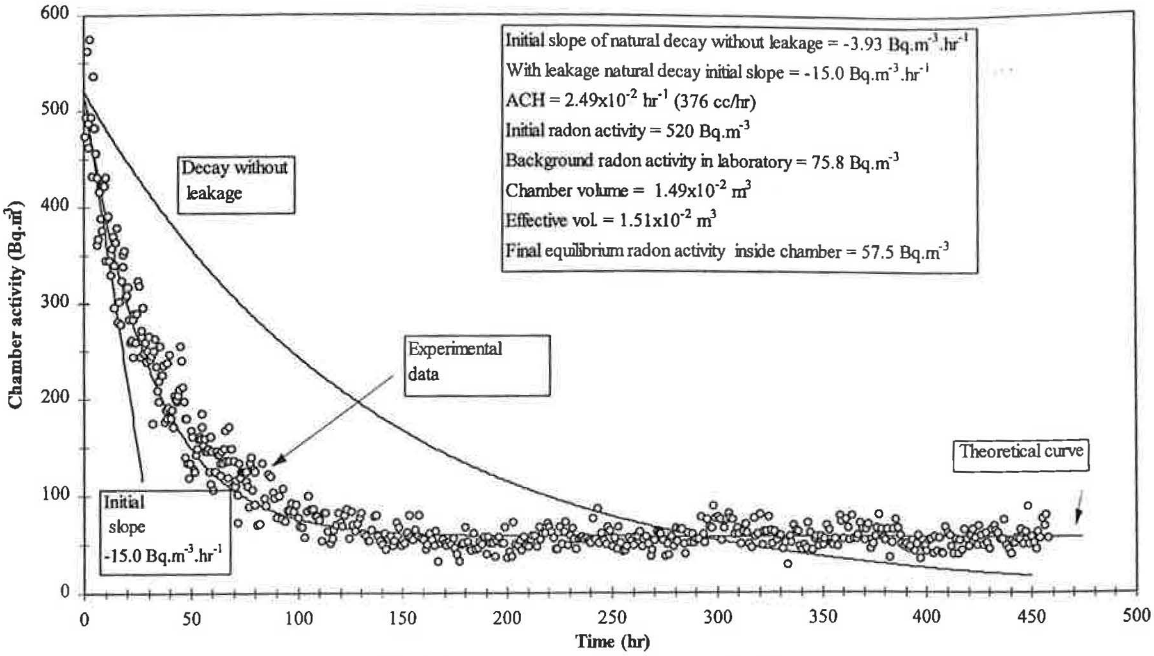

Fig. 2. Leakage measurement of chamber A.

$$\left.\frac{\mathrm{d}C}{\mathrm{d}t}\right|_{(t\to 0)} = M_e = \frac{E_o A + qC_o}{V} \tag{11}$$

$$E_o = \left(M_e - \frac{q}{V}C_o\right)\frac{V}{A}. \tag{12}$$

Since the measurement starts with zero radon activity inside the chamber, the initial slope of the curve, M_e, is independent of the back diffusion. The emanation rate of the sample can be directly determined after knowing the chamber leakage rate, q, and the ambient radon concentration inside the laboratory, C_o. The first portion of the growth curve was nearly a straight line and through the origin. The initial slope can be obtained by visual inspection or by considering the data of the first few hours modified by linear regression technique. The emanation rate can then be calculated by equation (12).

2.5. *Determination of the back diffusion rates of building materials*

The radon chamber activity is expressed by equation (3). After leaving the sample inside the chamber for a long period of time, the chamber radon concentration will approach an equilibrium value as shown below.

$$C(\infty) \cong \frac{\dfrac{E_o A}{V} + \dfrac{q}{V}C_o}{\left(\lambda + D + \dfrac{q}{V}\right)} \tag{13}$$

then

$$D = \frac{\left(\dfrac{E_o A}{V} + \dfrac{q}{V}C_o\right)}{C(\infty)} - \left(\lambda + \frac{q}{V}\right)$$

$$D = \frac{M_e}{C(\infty)} - \left(\lambda + \frac{q}{V}\right). \tag{14}$$

The back diffusion rate of the material can then be obtained by equation (14).

3. RESULTS AND DISCUSSION

3.1. *Leakage and laboratory radon background level*

All the measurements were performed under fairly dry conditions. Two chambers were built and were shown to have different leakage rates. Chamber A had a higher leakage rate than chamber B. Chamber A was built earlier and significant improvement was achieved in sealing chamber B in order to reduce the leakage rate. The leakage curves of the two chambers are shown in Figs 2 and 3 respectively.

In chamber A, an initial radon level of $520\,\mathrm{Bq\cdot m^{-3}}$ was used. This was obtained by leaving a high radon emitting material inside the chamber for a long time before taking it out. The radon level was allowed to decrease due to natural decay and exchange of outside and inside air. The ideal decay curve with no chamber leakage was also plotted. From Fig. 2, it was shown that equilibrium radon concentration was $57.5\,\mathrm{Bq\cdot m^{-3}}$ and was attained after about 150 hours. Using the initial slopes of the two decay curves and the theory shown in Section 2.3, the leakage rate of chamber A was found to be 375.5 cc/hr. After calculating the chamber leakage rate q, a theoretical decay curve was plotted using equation (6). It has been shown that the theoretical curve fits very well with the experimental data.

Similarly, the leakage rate of chamber B has been found to be 25.7 cc/hr from Fig. 3 using a different initial radon level ($3200\,\mathrm{Bq\cdot m^{-3}}$).

In Fig. 2, the experimental data show larger scattering than those in Fig. 3. This was due to the higher leakage rate of chamber A. The data scattering was also related to the diurnal variation of ambient radon level and temperature. A simple analysis shown in Appendix A indi-

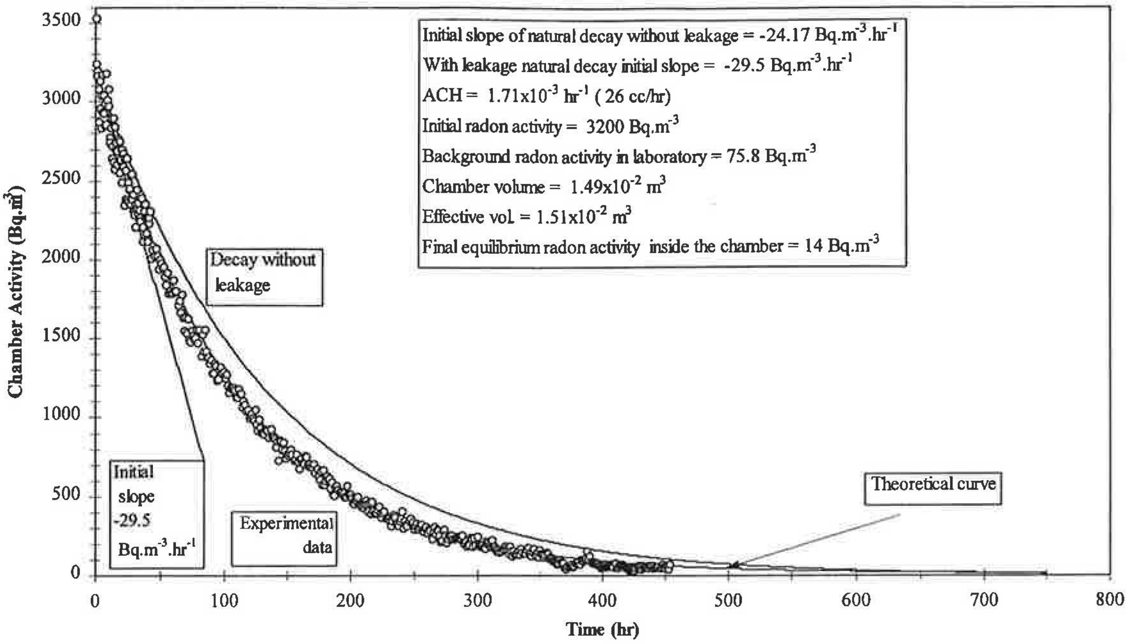

Fig. 3. Leakage measurement of chamber B.

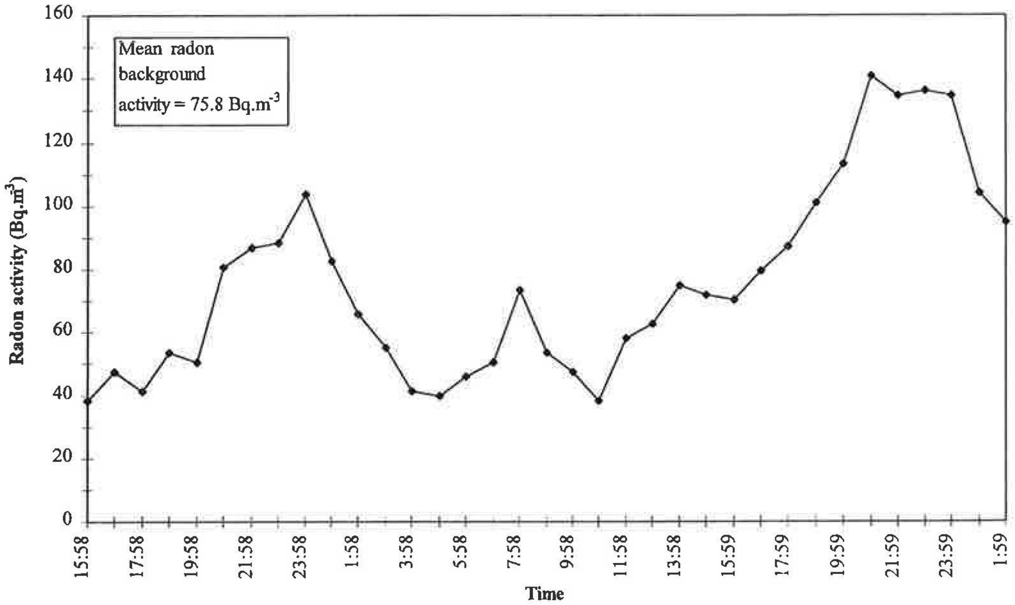

Fig. 4. Background radon activity in laboratory.

cates that temperature variation influences the radon level inside the chamber. It was estimated that in our experiments less than 2% radon level variation was encountered. A more accurate leakage rate can be determined if the temperature variation can be kept as low as possible.

Background radon level inside the laboratory is also a parameter in making an accurate estimation of the leakage rate in addition to the radon emanation and back diffusion characteristics. Since no chamber can guarantee zero leakage, background radon gas can get into the chamber by way of leakage or diffusion through the small gaps along the chamber intersections. In our tests, the background radon level has been carefully monitored and a typical trend is shown in Fig. 4. In calculating the leakage rates shown in Figs 2 and 3, the mean background radon activity of 75.8 Bq $\cdot$ m^{-3} was used. For Figs 2 and 3, it was shown that experimental data reached an equilibrium value which was not zero because ambient radon gas diffused into the chamber.

3.2. *Radon emanation rates and back diffusion rates of different building materials*

In our measurements, two different types of granite slabs and one type of bare concrete slab were studied. They were popularly used for finishing and construction purposes in buildings. Radon build-up curves inside the chambers were plotted and are shown in Figs 5–8.

In each figure, it can be seen that the radon level inside the chamber built up and reached equilibrium value after

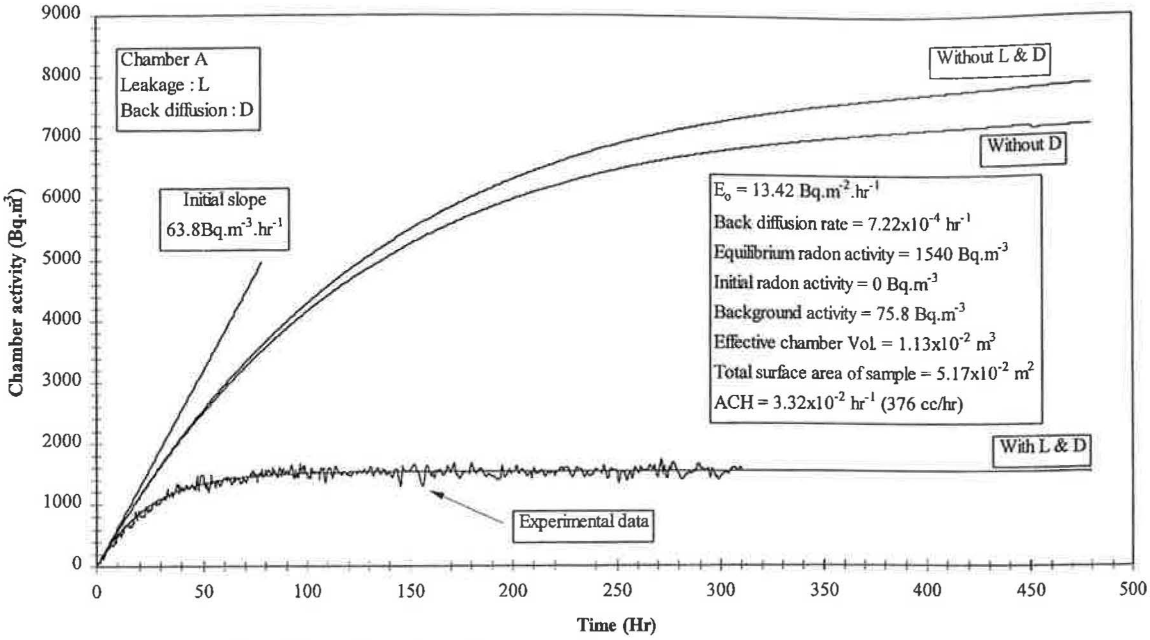

Fig. 5. Emanation rate and back diffusion rate of granite-1 in dry condition.

a long period of time (order of magnitude was 100 hours). The initial slope of the curve and the final equilibrium radon level were used in calculating the radon emanation rate and the back diffusion rate using the theory shown in Sections 2.4 and 2.5. Three curves were plotted in each figure, showing cases where no chamber leakage and back diffusion were considered, where no back diffusion was considered and where both chamber leakage and back diffusion were included. It can be clearly seen that the theoretical curve where both chamber leakage and back diffusion were considered fits very well with the experimental data. These figures indicate clearly the validity of the methodology used in this study in estimating the radon emanation rate and back diffusion rate of building materials. They also support the argument that an accurate correction of chamber leakage rate is very important in determining the correct radon emanation characteristics. Radon emanation and back diffusion rates of the samples tested are shown in Table 2.

The measurements show that granite-1 and granite-2

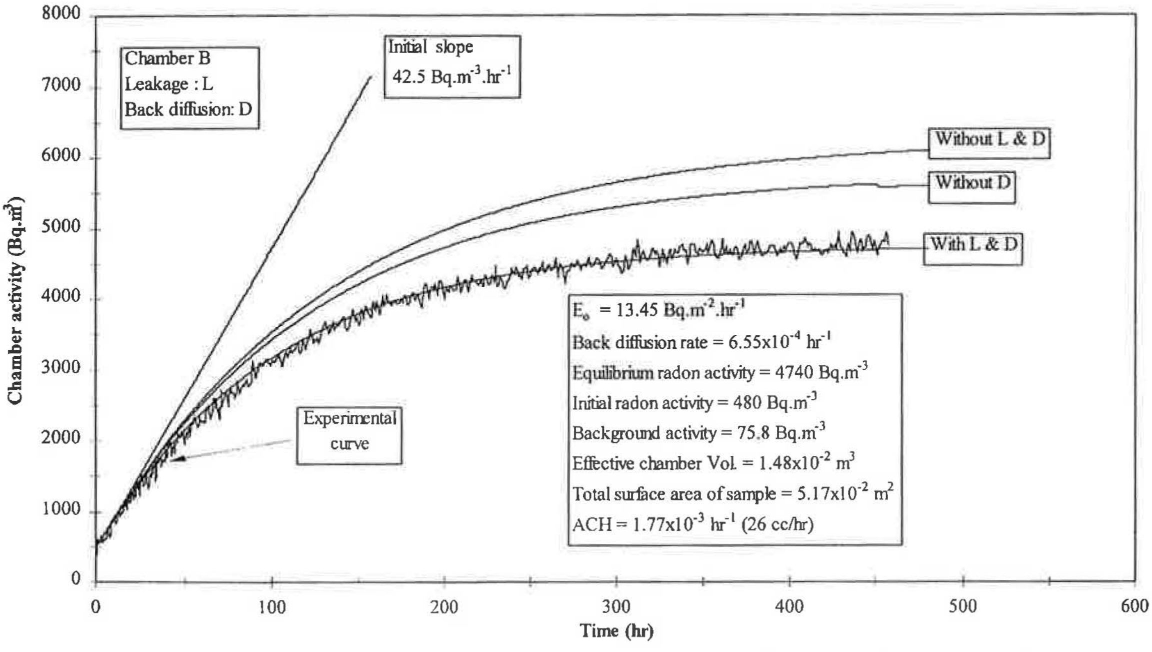

Fig. 6. Emanation rate and back diffusion rate of granite-1 in dry condition (repeat the measurement).

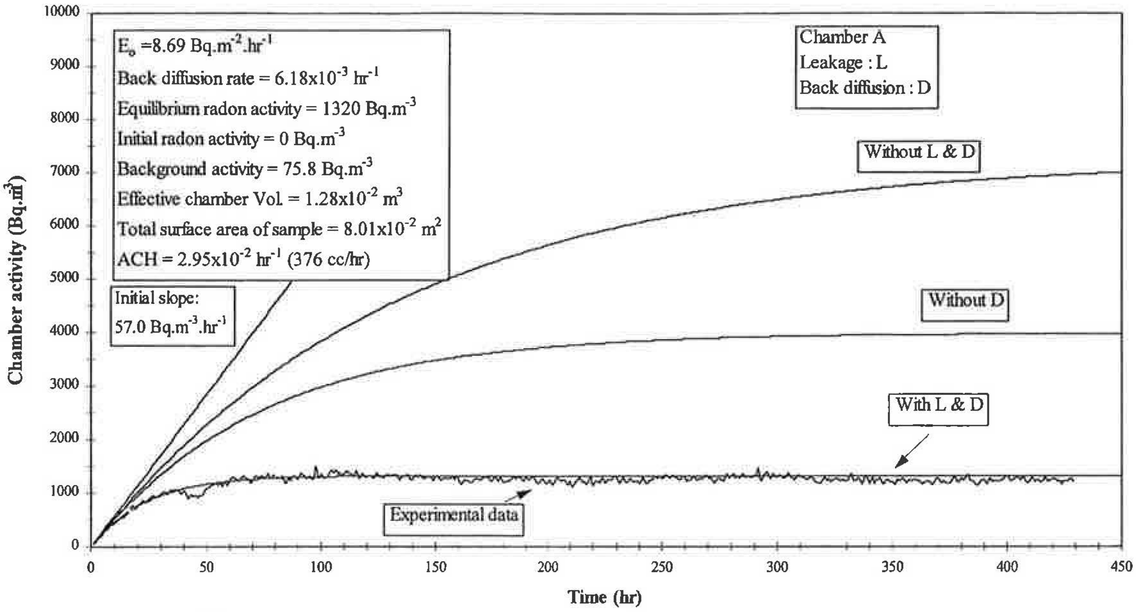

Fig. 7. Emanation rate and back diffusion rate of concrete slab in dry condition.

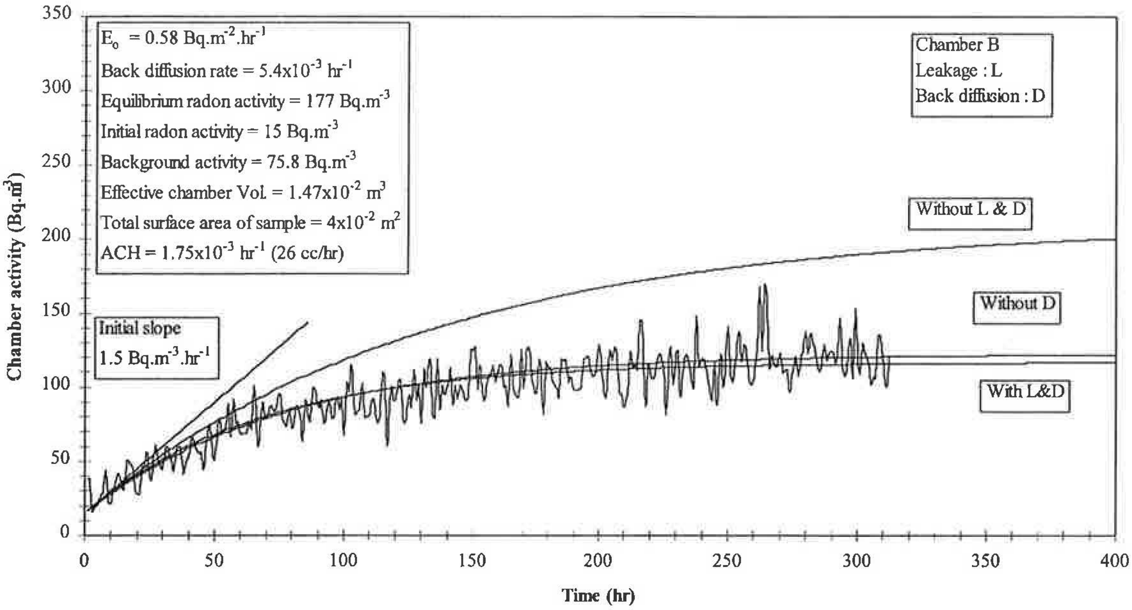

Fig. 8. Emanation rate and back diffusion rate of granite-2 in dry condition.

have very different emanation rates and back diffusion rates. Granite-1 was tested in both chamber A and chamber B and very close radon emanation rates were measured. The back diffusion rates vary a bit but are still within sufficient accuracy.

The bare concrete has an emanation rate less than

Table 2. Building material emanation rate and back diffusion

Building material	Leakage rate, ACH (hr^{-1})	Initial activity ($Bq \cdot m^{-3}$)	Emanation rate, E_o ($Bq \cdot m^{-2} \cdot hr^{-1}$)	Building material back diffusion (hr^{-1})	Specific back diffusion coeff. ($m \cdot hr^{-1}$)
Granite-1 (chamber A)	3.32×10^{-2}	0.0	13.42	7.22×10^{-4}	1.58×10^{-4}
Granite-1 (repeat measurement, chamber B)	1.77×10^{-3}	480	13.45	6.55×10^{-4}	1.88×10^{-4}
Granite-2 (chamber B)	1.75×10^{-3}	15.0	0.58	5.40×10^{-3}	1.99×10^{-3}
Concrete slab (chamber A)	2.95×10^{-2}	0.0	8.69	6.18×10^{-3}	9.83×10^{-4}

that of granite-1, but greater than that of granite-2. It is believed that granite-1 contains a lot more uranium and thorium radioactivity than other test samples.

4. CONCLUSION

Chamber tests have been conducted to estimate the radon emanation and back diffusion rates of building materials. The method provided is accurate and convenient for use in the laboratory.

It has been found that the leakage rate of the chamber can be accurately determined by the natural radon decay method. The chamber with a small leakage rate can be used to measure the radon emanation characteristics of low radioactive materials.

The emanation rate of a building material can be found from the chamber radon concentration. It is believed that radon back adoption and the concentration gradient existing across the material boundary cause back diffusion. The emanation rate can be split into two terms as E_o and αC. Then the emanation rate, E_o, of the building material is the zero order estimation while the back diffusion rate, D, is the first order interaction caused by the build up of radon level in air. The back diffusion of the material reduces radon activity in air. Radon molecules adopt back to a site within the material. Different materials have different porosities. Temperature and humidity also influence the material radon emission and adoption. These mechanisms are very complicated and require further investigation.

A larger emanation database is being developed using the method shown in this study. Further emphasis will be given to the effect of temperature and relative humidity on the radon emanation and back diffusion characteristics.

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APPENDIX A. INFLUENCE OF TEMPERATURE DIURNAL VARIATION ON RADON CONCENTRATION FLUCTUATION INSIDE CHAMBER

By ideal gas law

$$c \propto \frac{1}{T}\text{(keep pressure and volume as the constants)},$$

where C is the radon concentration inside the chamber and T is the extreme air temperature inside the chamber (K).

Then

$$\frac{\Delta C}{C} = \left| \frac{\dfrac{1}{T} - \dfrac{1}{T_m}}{\dfrac{1}{T_m}} \right| \times 100\%$$

or

$$\frac{\Delta C}{C} = \left| \frac{T_m - T}{T} \right| \times 100\%,$$

where T_m is the mean air temperature inside the chamber (temperature variation in the laboratory was from 291.1 to 298.8 K).

APPENDIX B. RELATIONSHIP BETWEEN THE RADON BACK DIFFUSION RATE D AND SPECIFIC RADON BACK DIFFUSION COEFFICIENT α

If the back diffusion effect is not considered, the mass balance equation of radon concentration becomes

$$\frac{dC}{dt} = -\lambda C + \frac{EA}{V} + \frac{q(C_o - C)}{V}. \qquad (A1)$$

If the back diffusion term is included and the emanation rate, E, is modified as $(E_o - \alpha C)$, equation (A1) becomes

$$\frac{dC}{dt} = -\lambda C + \frac{(E_o - \alpha C)A}{V} + \frac{q(C_o - C)}{V}$$

or

$$\frac{dC}{dt} = -\lambda C - \frac{\alpha A}{V}C + \frac{E_o A}{V} - \frac{q(C_o - C)}{V}. \qquad (A2)$$

Comparing equations (A1) and (A2), the following term is obtained:

$$D = \frac{\alpha A}{V}$$

or

$$\alpha = \frac{DV}{A},$$

where α is defined as the material specific radon back diffusion coefficient in this study.