

## CHAPTER 15

# Criteria for Closed Chamber Measurements of Radon Emanation Rate

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### ABSTRACT

Determination of the radon emanation rate from solid materials has been the objective of many studies. The experimental method most often used for determining the radon emanation rate is the so-called "closed chamber" method, in which the growth of radon concentration in a closed chamber is used as the basis for the calculation. Usually, only certain regions of the concentration growth curve are used for calculation purposes, specifically, the initial region of the curve, or the equilibrium value to which it converges. In spite of the experimental simplicity of this method, the mathematical procedures used to calculate the free emanation rate (defined as the emanation rate to an infinite space of air) raise many questions, the answers to which are often contradictory from one implementation of the method to another. The objective of this chapter is to provide quantitative values of the inherent bias in determining the emanation rate for various experimental conditions in the closed chamber method. Quantitative comparison is made among the mathematical models most often applied. An assessment is provided of the usefulness of each model for its assumed geometry of measurement.

### INTRODUCTION

Knowledge of the radon emanation properties of a material is important for many practical purposes as well as being of fundamental scientific interest. The experimental methods commonly used to investigate radon emanation may be divided into two groups. To the first group belong methods based on the accumulation of radon emanated from the sample into an emanation chamber. This is the so-called static method. The principle of the second group is dynamic and is based

#6738

on an air or gas flow over the surface of the sample to collect radon emanating therefrom.

The parameters that characterize emanation are emanation coefficient (sometimes called — with less precision — emanating power) and emanation rate. The radon emanation coefficient is defined as the ratio of radon atoms born in the material and released into the free pore space to all of the radon atoms born in the material. The emanation coefficient depends on the radium concentration and distribution in the material, the material porosity, and on what fills the inner pore volume (water or air). The emanation coefficient is not affected by external factors such as temperature, pressure, or radon concentration outside the sample.

The emanation rate characterizes the flux of radon from the material to the outer air volume. The emanation rate can be expressed as the total emanation rate of a sample [ $\text{Bq s}^{-1}$ ], the mass emanation rate of a material [ $\text{Bq kg}^{-1}\text{s}^{-1}$ ] and the surface emanation rate [ $\text{Bq m}^{-2}\text{s}^{-1}$ ]. In the case of so-called free emanation, in which the volume of the air surrounding the sample is infinite relative to the volume of the sample, correlation between the emanation coefficient and the emanation rate depends upon the diffusion length and the dimensions of the sample. However, unlike the emanation coefficient, the emanation rate is affected by external agents such as temperature and pressure changes. If the volume of the air surrounding the sample is not infinite and the radon concentration outside the sample cannot be assumed to be zero, the radon emanation process taking place cannot be referred to as free emanation. It is described as bound emanation. The bound emanation rate is lower than the free emanation rate. From the point of view of applications, it is important to know the free emanation rate.

The experimental method most often used for determination of the emanation rate is the closed chamber method, which, in spite of its simplicity, has a substantial disadvantage, namely, that the measured emanation rate is often not the free emanation rate. The closed chamber method is based on measurement of radon concentration as a function of time in a chamber enclosing the sample. This measurement method is not new. In different forms, it has been applied for the past few decades. Krisiuk et al. present mathematical expressions for the time- and position-dependent concentration of radon in the material and provide a time-dependent solution to this problem.<sup>1</sup> As shown by Krisiuk et al. and Samuelsson and Pettersson, it is possible on the basis of this theory to calculate the time-dependent emanation rate and the radon concentration growth in the sample chamber.<sup>1,2</sup> The derived expressions are so complicated, however, that in practice it is not possible to determine the change in the emanation rate from the experimental data on radon concentration in the chamber. Furthermore, properties of the material, necessary for such calculations (porosity and diffusion length), are often not known. In many cases, the change from the free emanation rate to the bound steady-state emanation rate (after closing the chamber) is so fast that it cannot be detected by measurement. These problems raise questions about the validity of the closed chamber method for determination of the free emanation rate.

Usual practice in measurements involving the closed chamber method is to assume that the volume of chamber is large relative to the pore volume of the sample to assure that free emanation takes place in the chamber. Another approach is to assume that at least at the beginning (after closing the chamber), when the radon concentration in the chamber is still very low, radon emanation may be treated as free emanation.<sup>3,4</sup> On the basis of this assumption, measurement of the initial radon growth rate in the chamber allows calculation of the free emanation rate.

The first of the previous two assumptions may not always be fulfilled, and the second may not always be justified. As shown by Samuelsson and Pettersson, the transition from free to bound emanation may occur within an hour or so after closing the chamber.<sup>2</sup> This means that the initial growth rate is not caused by free emanation.

The emanation rate determined on the basis of closed chamber measurements is the emanation rate occurring under constant pressure. In reality, all materials are affected by atmospheric pressure variations, which cause variations in the emanation rate. For this reason, instantaneous emanation rates may be different from emanation rates determined in the laboratory. Over the long term, however, the effects of atmospheric pressure variations usually cancel each other, and the emanation rate determined under stable conditions may be assumed to be the average value.

Despite its shortcomings, the closed chamber method is still a valuable tool for measurement of emanation rate. However, in view of the problems described above, it is necessary to establish and employ certain practical criteria for application of the method.

The objective of this chapter is to compare the available mathematical approaches to calculation of the free emanation rate from closed chamber experimental data and, on the basis of the comparison, to suggest the optimal method of correlating experimental data with theoretical calculations. Special attention is given to the method that takes into account the effect of reduction in driving force for radon emanation from the sample. A final outcome of the chapter is to present practical criteria for measurement of the emanation rate.

## DIFFUSION AND EMANATION THEORY

The typical case of a porous slab sealed hermetically in an emanation chamber is taken as the example. For mathematical simplicity, the one-dimensional case of radon diffusion and emanation is discussed. If the slab is, for example, of cubic shape, it is assumed that emanation takes place only through one pair of opposite walls of the cube, the other walls being covered with sealant to prevent emanation. A second possible approach is to consider the slab to be infinite in two of its dimensions so that emanation in only one dimension need be analyzed. Mathe-

matically, these cases are identical. Restricting the model to a one-dimensional case does not decrease its usefulness. Real constructional elements (e.g., walls) have two dimensions that are much larger than the third, and much larger than the radon diffusion length in the material, so that effectively the radon flow from the slab to the outside air may be considered to be through a pair of opposite planes.

The one-dimensional differential equation governing the processes of radon production, diffusion, and decay in the slab is:<sup>1</sup>

$$\frac{\partial C(x,t)}{\partial t} = f - \lambda C(x,t) + D_{\text{eff}} \frac{\partial^2 C(x,t)}{\partial x^2} \quad (1)$$

where:

- C(x,t) = concentration of radon in the pore space of the sample
- f = production of radon in the pore space
- λ = decay constant of radon
- D<sub>eff</sub> = effective diffusion coefficient of radon in the material

The radon emanation rate E can be expressed as:

$$E(t) = -\delta D_{\text{eff}} \left. \frac{\partial C(x,t)}{\partial x} \right|_{x=d} \quad (2)$$

where

d = half-thickness of the sample (the cartesian coordinate system has its origin in the middle of the sample)

and

δ = porosity of the sample material.

As a result of radon emanation from the sample, the radon concentration growth in the chamber is described by the equation:

$$\frac{dN(t)}{dt} = -\lambda N(t) + \frac{q(t)}{V} \quad (3)$$

where

- q(t) = E(t)·S is the total emanation rate of a sample (S is the surface area of the sample)
- V = the volume of the chamber accessible for emanation (this is the volume of the chamber minus the volume of the sample)
- N(t) = radon concentration in the chamber at time t after closing the chamber.

The emanation chamber is assumed to be hermetically sealed. In practice, it is always necessary to check whether the chamber is hermetically sealed. If it is not possible to prevent a small leakage and if the leakage rate is constant, the solution may be modified by replacing the value λ by the sum λ + λ<sub>1</sub> in Equation 3 and in the boundary conditions of Equation 1, where λ<sub>1</sub> is the escape constant of radon

from the chamber.<sup>4</sup> Pressure in the chamber should be kept constant throughout the measurement.

If the sample emanates for a period of time much greater than the radon half life, steady-state is achieved (∂C/∂t = 0) and the solution is:

$$C(x) = -\frac{f/\lambda}{\cosh \beta} \left\{ \frac{1}{1 + \tanh \beta/\alpha\beta} \right\} \cosh \frac{x}{L} + \frac{f}{\lambda} \quad (4)$$

where

- L = diffusion length of radon in the material (L = √D<sub>eff</sub>/λ)
- β = d/L (d is the half-thickness of the sample)
- α = V/V<sub>s</sub> (V<sub>s</sub> is volume of the sample)

Substituting Equation 4 into Equation 2 and solving leads to the steady-state emanation rate:

$$E(\infty) = \frac{\delta L f (\tanh \beta)}{1 + \tanh \beta/\alpha\beta} \quad (5)$$

If α → ∞, which corresponds to the situation that the volume of the air surrounding the slab is infinite relative to the pore volume of the slab, Equation 5 takes a simpler form, and the free emanation rate is given by:

$$E = \delta f L \tanh \beta \quad (6)$$

For simplicity, the free emanation rate is denoted by E, as it is independent of time.

The emanation rate cannot be measured directly and is calculated from the growth of the radon concentration in the chamber. After closing the chamber, the emanation rate changes from the free emanation rate given by Equation 6 (providing that, before closing the chamber, steady-state free emanation conditions were present) to the bound emanation rate given by Equation 5, based on actual values of V and V<sub>s</sub>. The values of E(t) — between E and E(∞) — can be found by substituting the full time-dependent solution of Equation 1 into Equation 2, and solving. The time-dependent solution of Equation 1 is complicated and has been given by Krisiuk et al. and Samuelsson and Pettersson.<sup>1,2</sup> Equation 3 should be solved with the explicit form of E(t). The final form of N(t) should allow calculation of one of the following parameters: L, δ, η (emanation coefficient), if all required parameters are known. Alternatively, the complete set of parameters could be fitted into the experimental data. In practice, however, neither procedure is usually possible, because all required parameters are usually not known. The complicated form of the expression N(t) also makes fitting very difficult.

The dependences of E(t) and N(t) were presented graphically by Samuelsson and Pettersson, Samuelsson, and Samuelsson and Erlandsson for various values of L, δ, and η.<sup>2,5,6</sup>

However, from a practical point of view, knowledge of the change in the emanation rate with time is not necessary, because the change in the emanation rate characterizes the entire system, not just the sample. The sample is characterized by its free emanation rate.

Difficulties in correlating experimental data of radon concentration growth in the chamber with the free emanation rate are usually overcome by two approaches. The first approach is to assume that the value of  $\alpha$  is large enough that the emanation in the chamber may be approximated with negligible error by the free emanation rate. In this case, solution of Equation 3 takes the simple form:

$$N(t) = \frac{q}{\lambda V} (1 - \exp(-\lambda t)) \quad (7)$$

From Equation 7, on the basis of experimental data, it is easy to find the constant value of  $q$ , the total free emanation rate. However, in practice, the value of  $\alpha$  is not always large enough to allow free emanation to be approximated in the chamber. A conflicting requirement is that for material of a relatively low emanation rate, the volume into which emanation takes place should be small in order to achieve good counting statistics.

The second approach is to assume that, at least in the beginning, soon after closing the chamber, when the radon concentration in the chamber is still low, the emanation may be treated as free emanation. In this case, from the slope of the initial growth rate of the radon concentration in the chamber, the free emanation may be calculated from

$$r = \frac{N(t)}{t} = \frac{q}{V} \quad (8)$$

This approach, however, introduces several controversies. As shown by Samuelsson and Pettersson, the very period after closing the chamber is characterized by the fastest change in the emanation rate.<sup>2</sup> For example, for  $V = V_p$  and for  $L = 100$  cm, the emanation rate reaches a bound steady-state value within 4 h after closing the chamber. The ratio of bound to free emanation rates for these conditions is 0.65. It is obvious that, in this case, the slope of the initial growth rate would not provide the value of the free emanation rate. Sometimes, however, estimation of the free emanation rate from the initial growth rate may be possible. This may be true when, because of the geometry of the measurement, emanation is at all times close to free emanation and, for small values of  $t$  ( $t < 24$  h), the exponent in Equation 7 may be approximated by the two first terms of the series:

$$\exp(-\lambda t) = 1 + \frac{-\lambda t}{1!} + \frac{(-\lambda t)^2}{2!} + \frac{(-\lambda t)^3}{3!} + \dots \approx 1 - \lambda t, \quad (9)$$

Then Equation 7 takes the form:

$$N(t) = \frac{q}{\lambda V} (1 - 1 + \lambda t) = \frac{qt}{V} \quad (10)$$

where  $q$  is the total free emanation rate.

In applying this method to determine the free emanation rate, important assumptions are that the emanation rate has a constant value during the course of the measurement, and that the exponential growth of radon activity in the chamber is approximately linear. Assuming a constant value of the emanation rate at a time when the emanation rate changes the most rapidly is synonymous with assuming that free emanation takes place in the chamber all of the time.

In summary of the previous discussion, the two commonly used methods for determining the free emanation rate may be applied only when free emanation takes place in the chamber all of the time. The emanation rate may be determined (1) from Equation 7 (for example, by fitting experimental data to the exponential curve given by this equation), or (2) from Equation 10. The advantage of method (1) is that the time of the measurement is not restricted to the period after closing the chamber, which is important in the case of samples of low emanation rate. For such samples, the radon concentration in the chamber within the first few hours after closing the chamber may not be high enough to allow reasonable counting statistics. Use of method (2) allows the time of measurement to be shortened significantly. Method (2) can be used for samples having a relatively high emanation rate.

Another approach to the problem of free emanation rate measurements was presented by Wojcik and Morawska.<sup>7</sup> The model developed does not focus on the processes taking part inside the sample, but rather on their influence on growth of the radon concentration in the chamber. The principal assumption of this model is that if the volume of the chamber is not considerably greater than the volume of the sample, the radon present in the chamber reduces the driving force for emanation from the sample. The smaller the volume of the emanation chamber relative to the sample, the greater is the effect of the driving force reduction. The radon concentration vs. time relationship in the chamber is given by the following equation in which the effect of the reduction in driving force is taken into account:

$$\frac{dN(t)}{dt} = -\lambda N(t) + \frac{q}{V} \left( 1 - \frac{N(t)}{N_p} \right) \quad (11)$$

In this equation,  $N_p$  is the maximum radon concentration in the pore space of the sample material, whose value depends on the radium concentration in the material, the porosity of the material, and the radon emanation coefficient. The maximum radon concentration in the pore space of the sample depends only on the condition of the sample at the time of the measurement (the moisture content, which affects

the emanation coefficient) and not on the way in which the measurement is performed. The maximum concentration is reached when the difference between the volume of the chamber and the volume of the sample is much smaller than the volume of pore space within the sample. The reduced driving force effect described here may be compared to the term back diffusion discussed with different meanings, by Jonassen and Samuelsson and Pettersson.<sup>4,6</sup>

The second term on the right-hand side of Equation 11 may be compared to the second term on the right-hand side of Equation 3 to give:

$$E(t) = E \left( 1 - \frac{N(t)}{N_p} \right) \quad (12)$$

which may be considered to be an approximation of the time-dependent emanation rate.

The solution to Equation 11 is:

$$N(t) = \frac{q}{V(\lambda + q/VN_p)} \left\{ 1 - \exp \left[ -t \left( \lambda + \frac{q}{VN_p} \right) \right] \right\} \quad (13)$$

Thus it is possible to determine the total free emanation rate,  $q$ , of a sample, and the maximum radon concentration in the pore space of the sample,  $N_p$ , by making a time series of measurements of radon concentration growth in the chamber and applying Equation 13. The advantages of this method are that it is not necessary to assume that emanation in the chamber is free (and it does not have to be free) and that knowledge of the internal parameters of the sample ( $\delta$  and  $L$ ) is not necessary in order to determine the free emanation rate from the experimental data.

The validity of the results obtained in this way may be verified via comparison with theoretical calculations performed on the basis of Equation 5. As a basis for these comparisons, an experimental set of data may be used.<sup>8</sup> A sample of lightweight concrete of volume  $3.375 \times 10^{-3} \text{ m}^3$  was enclosed hermetically in a chamber of volume  $3.95 \times 10^{-2} \text{ m}^3$ . The porosity of the material was 0.0733. The values obtained for the total free emanation rate and the maximum concentration of radon in the pore space of the material by fitting the time series of radon concentration in the chamber to Equation 13 are  $q = 4.11 \times 10^{-5} \text{ Bq s}^{-1}$  (19.6 atom  $\text{s}^{-1}$ ), and  $N_p = 69.7 \times 10^3 \text{ Bq m}^{-3}$ . The half-thickness of the sample was much smaller than the diffusion length, so that the ratio  $\tanh \beta/\beta$  may be assumed to be unity.

According to Equation 12, the ratio of the bound steady-state emanation rate to the free emanation rate may be expressed as:

$$\frac{E(\infty)}{E} = 1 - \frac{N(\infty)}{N_p} \quad (14)$$

Table 1. Ratio of the Bound Steady-State to the Free Emanation Rate Calculated from Experimental Data and Theoretical Model as a Function of the Volume of the Chamber

$V[\text{M}^3]$	$\frac{V}{V_s}$	$\alpha = \frac{V^a}{\delta V_s}$	$\frac{E(\infty)}{E} = 1 - \frac{N(\infty)}{N_p}$	$\frac{E(\infty)}{E} = \frac{1}{1 + \gamma\alpha}$
0.0361	10.67	145.9175	0.9928	0.9932
0.0200	5.93	80.8407	0.9861	0.9878
0.0100	2.96	41.4204	0.9725	0.9758
0.0050	1.48	20.2102	0.9465	0.9528
0.0010	0.30	4.0420	0.7795	0.8017

<sup>a</sup> In this case,  $dV_s = 0.2474 \times 10^{-3} \text{ m}^3$

The values of this ratio were calculated for different values of the volume of the chamber for the values  $q$  and  $N_p$  given above ( $q(\infty)/q = E(\infty)/E$ ). The results are shown in the fourth column of Table 1. The same ratio, calculated from the theoretical Equations 5 and 6, is given by:

$$\frac{E(\infty)}{E} = \frac{1}{1 + \tanh \beta/\beta\alpha} \quad (15)$$

Values of this ratio were calculated for different values of  $a$ , assuming that  $\tanh b/b$  is unity. The results are shown in the fifth column of Table 1.

From Table 1, it can be seen that values of the ratio of the bound steady-state emanation rate to the free emanation rate calculated from the experimental data and from the theoretical model are in excellent agreement. Small differences may be due to errors in porosity determination or to the fact that the ratio  $\tanh b/b$  might not be exactly equal to unity.

As a summary of this discussion, it may be concluded that the free emanation rate can be determined by the closed chamber method using Equation 13 independent of the geometry of the measurement, that is, independent of whether free or bound emanation takes place in the chamber. This method is the simplest for determining the free emanation rate from experimental data without using complicated theoretical expressions. The result is the same as would be obtained by exact theory.

The values of time-dependent emanation rate, calculated accordingly to Equation 13 for small values of  $t$ , differ slightly from those calculated from Equation 2, but approach the same final value.<sup>5</sup> The change from free to bound emanation is faster according to the theoretical model described by Equation 2. However, the theoretical calculation is based on simplifying assumptions, for example, the assumption of instantaneous air mixing in the outer volume, which would cause the emanation process to be more rapid. As mentioned previously, experimental emanation rate vs. time data are not available, because the emanation rate is not measured directly.

## EXPERIMENTAL DETERMINATION OF RADON EMANATION PROPERTIES

Experimental methods for applying the closed chamber method to determine radon emanation properties will now be considered. The radon emanation properties are the total emanation rate of a sample, the specific mass emanation rate, the specific surface emanation rate, and the emanation coefficient.

### Total Free Emanation Rate of a Sample

In planning to measure the total free emanation rate of a sample, it is necessary to take into account the properties of the sample as well as the following requirements for the experiments:

- A rough estimate of the order of magnitude of the emanation rate, on the basis of the radium content of the sample. This estimate allows selection of an appropriate sample mass for reasonable counting statistics (for example, several grams for uranium ore, but several kilograms for bricks).
- Estimation of the radon diffusion length and the porosity of the material. Usually, the porosity of materials such as sand, silt, or clay is between 0.4 and 0.6, and the diffusion length in the range 100 to 200 cm for dry materials, and of the order of 1 to 3 cm for materials saturated with water. Both the porosity and the diffusion lengths of brick and concrete are smaller: the porosity of concrete is around 0.1 (bricks may be higher) and the dry diffusion length is usually not greater than 50 cm.<sup>9</sup> Knowledge of porosity and diffusion length is helpful in finding the optimal relationship between the dimensions of the sample and the volume of the emanation chamber.
- The duration of the experiment. The duration of the experiment may be decreased significantly by determining the total free emanation rate of a sample from the initial slope of the radon concentration vs. time curve; however, in this case, it is necessary to arrange conditions such that radon emanation may be considered "free" throughout the measurements. Results for the ratio  $E(\infty)/E$  (determined from Equation 15) are shown in Table 2a—d for different values of diffusion length and porosity as a function of the volume of the material  $V_s$  and the volume of the emanation chamber,  $V$ . Tables 2a and 2b give data for material similar to typical dry and wet soil, respectively (porosity and diffusion length large, and porosity high and diffusion length small, respectively). Results for material similar to typical dry and wet concrete are given in Tables 2c and 2d.

From Table 2, it is clear that for materials similar to dry soil, it is difficult to create a condition of free emanation in the chamber ( $E(\infty)/E \approx 1.0$ ), except for very small samples in a very large chamber. The situation changes slightly for wet soils, but still the volume of the chamber must be 10 to 20 times larger than the volume of the sample in order to ensure free emanation for thick samples.

Table 2. The Ratio  $E(\infty)/E$  Calculated for Different Values of Diffusion Length and Porosity as a Function of the Volume of the Material and the Volume of the Chamber

Table 2a							
Diff. Length = 100.0 cm Porosity = 0.500							
d[cm]	H[cm] <sup>a</sup>						
	1	5	10	20	50	80	100
1	0.7	0.9	1.0	1.0	1.0	1.0	1.0
5	0.3	0.7	0.8	0.9	1.0	1.0	1.0
10	0.2	0.5	0.7	0.8	0.9	0.9	1.0
20	0.1	0.3	0.5	0.7	0.8	0.9	0.9
50	0.9	0.2	0.3	0.5	0.7	0.8	0.8
80	0.0	0.1	0.2	0.4	0.6	0.7	0.8
100	0.0	0.1	0.2	0.3	0.6	0.7	0.7

  

Table 2b							
Diff. Length = 2.0 cm Porosity = 0.500							
d[cm]	H[cm] <sup>a</sup>						
	1	5	10	20	50	80	100
1	0.7	0.9	1.0	1.0	1.0	1.0	1.0
5	0.5	0.8	0.9	1.0	1.0	1.0	1.0
10	0.5	0.8	0.9	1.0	1.0	1.0	1.0
20	0.5	0.8	0.9	1.0	1.0	1.0	1.0
50	0.5	0.8	0.9	1.0	1.0	1.0	1.0
80	0.5	0.8	0.9	1.0	1.0	1.0	1.0
100	0.5	0.8	0.9	1.0	1.0	1.0	1.0

  

Table 2c							
Diff. Length = 20.0 cm Porosity = 0.050							
d[cm]	H[cm] <sup>a</sup>						
	1	5	10	20	50	80	100
1	1.0	1.0	1.0	1.0	1.0	1.0	1.0
5	0.8	1.0	1.0	1.0	1.0	1.0	1.0
10	0.7	0.9	1.0	1.0	1.0	1.0	1.0
20	0.6	0.9	0.9	1.0	1.0	1.0	1.0
50	0.5	0.8	0.9	1.0	1.0	1.0	1.0
80	0.5	0.8	0.9	1.0	1.0	1.0	1.0
100	0.5	0.8	0.9	1.0	1.0	1.0	1.0

Table 2. The Ratio  $E(\infty)/E$  Calculated for Different Values of Diffusion Length and Porosity as a Function of the Volume of the Material and the Volume of the Chamber (continued)

Table 2d							
Diff. Length = 2.0 cm Porosity = 0.050							
d[cm]	H[cm] <sup>a</sup>						
	1	5	10	20	50	80	100
1	1.0	1.0	1.0	1.0	1.0	1.0	1.0
5	0.9	1.0	1.0	1.0	1.0	1.0	1.0
10	0.9	1.0	1.0	1.0	1.0	1.0	1.0
20	0.9	1.0	1.0	1.0	1.0	1.0	1.0
50	0.9	1.0	1.0	1.0	1.0	1.0	1.0
80	0.9	1.0	1.0	1.0	1.0	1.0	1.0
100	0.9	1.0	1.0	1.0	1.0	1.0	1.0

<sup>a</sup> For simplicity, the volume of the chamber is expressed as the height of the air column above the sample, H, and the volume of the sample is expressed as its half-thickness, d.

On the other hand, for materials of properties similar to typical wet concrete, the volume of the chamber need be only slightly higher than the volume of the sample in order to create the conditions for free emanation.

If the conditions for free emanation in the chamber are fulfilled, the total free emanation rate may be calculated from Equations 7 or 10. If these conditions are not fulfilled, or if there are doubts as to whether the emanation is free, Equation 13 should be applied.

### Specific Mass Emanation Rate

The specific mass emanation rate [ $\text{Bq kg}^{-1}\text{s}^{-1}$ ] may be calculated directly from the total free emanation rate when the sample is so thin relative to the diffusion length (small value of  $\beta$ ) that the ratio  $\tanh \beta/\beta$  may be assumed to be unity. Under these conditions, essentially all radon that enters the pore space of the sample will be able to emanate from the sample to the chamber. However, escape of all radon entering the pore space may not occur for all cases of free emanation in the chamber even when  $E(\infty)/E$  is close to unity, because it may be that when  $\tanh \beta/\beta < 1.0$ ,  $1/\alpha \ll 1.0$  (see Equation 15).

The value of  $\tanh \beta/\beta$  may be calculated on the basis of known values of the diffusion length. It is easy to check experimentally whether the assumption that  $\tanh \beta/\beta \ll 1.0$  is justified. In order to do this, a second sample of smaller dimensions should be measured. If the determined values of the mass emanation rate are equal for the original sample and for the smaller sample, the implication is that for both cases  $\tanh \beta/\beta$  equals unity.

### Specific Surface Emanation Rate

The specific surface emanation rate [ $\text{Bq m}^{-2}\text{s}^{-1}$ ] is usually defined as the surface emanation rate of thick samples when further increase in the thickness does not alter the radon flux through the sample surface. In this case, the closed-chamber method can be applied by sealing the chamber to the sample surface, instead of enclosing the sample in the chamber. The surface emanation rate may be calculated from Equation 5 or, for free emanation conditions, from Equation 6. With the increasing  $\beta$  (increasing thickness of the sample)  $\tanh \beta \rightarrow 1.0$ , and the surface emanation rate tends to a constant value given by  $E = \delta fL$ .

For example, if the half-thickness of the sample is twice the diffusion length ( $d = 2L$ , as recommended by Jonassen 1983), the value of  $\tanh \beta$  is 0.964. In order to satisfy the condition  $\tanh \beta \approx 1.0$ , the thickness of a concrete sample should be of the order to 1 to 2 m (if the diffusion length is in the range 0.25 to 0.5 m), and of a soil sample (which has a higher diffusion length), of the order of several meters. Such large samples usually cannot be enclosed in an emanation chamber unless the dimensions of the chamber are very large. Instead, for practical reasons, it is better to seal a smaller chamber to the surface of the material.

### Emanation Coefficient

The emanation coefficient may be calculated from the value of the total free emanation rate (independent of whether determined from the conditions of free or bound emanation in the chamber), but only when  $\tanh \beta/\beta = 1.0$ . The emanation coefficient is given by the equation:

$$\eta = \frac{q}{C_{\text{Ra}} m} \quad (16)$$

where  $C_{\text{Ra}}$  is the radium concentration of the sample (units of  $\text{Bq kg}^{-1}$ ) and  $m$  is the mass of the sample.

### Diffusion Length and Porosity

The closed-chamber method may allow determination of the porosity and diffusion length of the material, in addition to determination of the radon emanation properties. If emanation rate measurements are performed for two cases for which the values of the ratio  $E(\infty)/E$  are different ( $< 1.0$ ) (calculated from Equation 14), it is possible to calculate the values of  $L$  and  $\delta$  from Equation 15. For example, for  $d = 5$  cm and  $H = 1$  cm (see Table 2a),  $E(\infty)/E = 0.3$ , and for  $d = 5$  cm and  $H = 10$  cm, the value of this ratio is 0.8. From Equation 15, values of  $\alpha$  and  $\beta$  may be determined, and from these,  $L$  and  $\delta$ .

There are, however, restrictions on application of the closed-chamber method for determining diffusion length and porosity because, for some materials, it is difficult to create conditions favoring distinctly different values of the ratio  $E(\infty)/E$  (see, for example, Table 2d).

## CONCLUSIONS

This chapter provides a detailed discussion of application of the closed-chamber method to determination of radon emanation properties. Despite the fact that the closed-chamber method in different forms has been used for decades, and the fact that theoretical models of the processes taking part in the sample and in the chamber have been developed, application of the method is still controversial, because it is very difficult to correlate experimental data with the complicated theoretical expressions in order to calculate the free emanation rate of the sample. Furthermore, such calculations are impossible unless the sample diffusion length and the sample porosity are known.

On the basis of the analysis provided here, the following conclusions may be drawn:

- The two common methods of determining the free emanation rate via the closed-chamber method, namely, (i) fitting experimental data to the exponential form of the radon concentration growth curve in the chamber, and (ii) determining the initial slope of the concentration growth curve, may be used only if the emanation in the chamber, because of the geometry of the measurement, can be treated as free emanation.
- If the requirements for free emanation cannot be fulfilled, or if it is difficult to judge whether emanation in the chamber is free or bound, it is suggested that for free emanation rate calculations, use be made of the expression taking into account the effect of the reduction in driving force caused by the radon present in the chamber. The form of this expression is simple (equation 13) and does not require knowledge of internal properties of the sample such as porosity or diffusion length. It was demonstrated here that values of the free emanation rate determined in this way are in excellent agreement with values calculated from the exact theoretical model.

The last section of this chapter identifies practical criteria for measuring emanation properties. The results presented for the ratio  $E(\infty)/E$ , for various values of diffusion length and porosity, as a function of pore volume of the sample and volume of the emanation chamber, are intended to be of practical use in planning emanation rate measurements, especially if there is a requirement for creating conditions of free emanation in the chamber.

Certain other parameters can be also calculated from the free emanation rate, namely, the specific mass emanation rate, the surface emanation rate, the emanation coefficient, the diffusion length, and the porosity. Conditions are discussed under which these parameters can be determined.

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