



A theoretical approach to the measurement of radon diffusion and adsorption coefficients in radonproof membranes

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Abstract

Theoretical expressions for the determination of radon diffusion and adsorption coefficients in radonproof membranes using the standard method for testing the radon insulation effectiveness of a membrane tightly placed between two compartments are derived from the steady-state solution of the differential equation for radon diffusion. These expressions are applied to an experimental set-up designed specifically to carry out such a determination under controlled environmental conditions.

Minimum detectable radon diffusion and adsorption coefficients of the method are shown to be typically well below those measured in most of the materials commonly used as radon barriers.

Besides the radon diffusion and adsorption coefficients a new parameter called radon flux transmission factor and a more suitable definition of the radon permeability coefficient are also proposed to characterize the applicability of any membrane for radon insulation purposes.

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1. Introduction

The application of radonproof membranes under or on top of floor slabs as passive barriers to mitigate radon entry in dwellings from the soil, in almost all the cases the main source of indoor radon, is one of the most widely used radon protective construction techniques [1–3].

The standard method for testing the insulation effectiveness of a membrane as a radon barrier usually consists in enclosing the membrane to be tested between two chambers of the same or different volumes. In one of them, the so-called primary chamber, a high radon concentration is generated from a source with a given radon production rate. By measuring the radon flux into the other chamber, the secondary chamber, and the gas concentrations in both compartments either in steady-state [4,5] or transient conditions [6], the radon insulation effectiveness of the tested membrane is commonly characterized by means of the

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proportionality constant between the radon flux density out of the membrane in the secondary chamber and the ratio between the difference in radon concentration in the air on both sides of the membrane and the membrane thickness. This proportionality constant, in the bibliography usually referred to as permeability coefficient [7] or permeation coefficient [8], is actually an effective parameter that takes into account the combined effects of both the radon adsorption and diffusion processes in the membrane and then it can not be used to estimate the radon adsorption and diffusion coefficients in the membrane material unless the differential equation for radon diffusion be solved with appropriate boundary conditions.

Furthermore, the above definition of the permeability coefficient for radon is not entirely satisfactory and should be avoided because (1) it implicitly involves that such a coefficient is a parameter characteristic of each membrane which is not exactly true for radon since, as we will show further on, it is also a function of the surface area of the tested membrane sample as well as of the volume of the secondary chamber and (2) it is based on the wrong assumption that the radon concentration gradient within the membrane is constant, a condition that is hardly fulfilled in radonproof membranes due to the short diffusion length for the gas in the membrane material, usually of the same order of magnitude or even lower, 10^{-3} – 10^{-4} m, than the specimen thickness used for laboratory testing [9].

The aim of this paper is to describe a theoretical approach to the measurement of the radon diffusion and adsorption coefficients in radonproof membranes by applying the standard testing method mentioned above to an experimental set-up designed specifically to carry out such a measurement under controlled environmental conditions. Besides the radon diffusion and adsorption coefficients, a new parameter called radon flux transmission factor (RFT) and a more appropriate definition of the radon permeability coefficient than commonly used in the bibliography are also proposed to characterize the “radon tightness” of any membrane.

2. Theory

According to the standard method used for testing the applicability of a membrane as a radon barrier, let us suppose, as shown in Fig. 1, a membrane tightly placed between two compartments in which radon losses by leakage are assumed to be negligible and each connected in a closed circuit to a radon monitor RM provided with temperature and relative humidity sensors for a continuous recording of the radon concentration, temperature and relative humidity in the air of both compartments. The internal pump of each radon monitor drives continuously an air stream at a constant flow rate, the same for both chambers. After passing through the radon source RS with a constant production rate, the air stream induces an increasingly high radon concentration in the primary chamber. Once adsorbed on the membrane, radon diffuses through the membrane material to the secondary chamber where it desorbs causing the increase of the gas concentration in this chamber with time.

As derived by applying the method of trial function approximation [10] to the time-dependent one-dimensional differential equation for radon diffusion, the time constant τ of the overall transmission process of the gas through a membrane of thickness d , in the literature usually referred to as “reshaping time”, can be approximately estimated from the formula

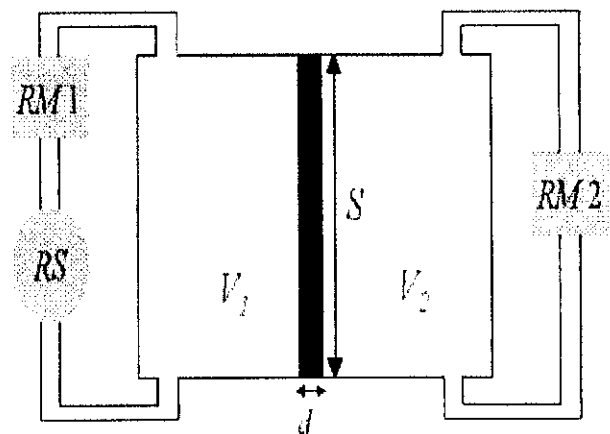


Fig. 1. Schematic view of the experimental set-up designed for testing the insulation effectiveness of radonproof membranes.

$$\tau = \frac{1}{\left(\lambda + \frac{\pi^2 D}{d^2}\right)}, \quad (1)$$

where D is the diffusion coefficient for radon in the membrane material and λ its decay constant. This time constant is a measure of the time taken for the system to change from its initial to its final state. Specifically, it is the time for a fraction 0.63 (strictly $1 - e^{-1}$) of the whole change to be complete. Accordingly, after an elapsed time $t = 5\tau$ steady-state conditions can be assumed for practical purposes since this is the time required for radon concentrations in both chambers and inside the membrane to reach about 99% of their final steady-state values. As estimated from formula (1), for radonproof membranes 1 mm thick with radon diffusion coefficients typically between 10^{-12} and $10^{-13} \text{ m}^2 \text{ s}^{-1}$ this time will range from approximately 5 to 19 days.

To find the steady-state radon concentrations and radon fluxes into and out of the membrane in the primary and secondary chambers, we have to solve, assuming no radon production within the membrane, the stationary one-dimensional differential equation for radon diffusion

$$D \frac{d^2 C}{dz^2}(z) - \lambda C(z) = 0, \quad (2)$$

where $C(z)$ is the steady-state radon concentration at any depth z in the membrane.

The general solution of Eq. (2) must be made to fit the boundary conditions expressing the balance between radon entries and losses in both the primary and secondary chambers which for the arrangement shown in Fig. 1 are

$$f + DS \frac{dC}{dz}(0) - \frac{\lambda V_1}{\beta} C(0) = 0, \quad (3)$$

$$-DS \frac{dC}{dz}(d) - \frac{\lambda V_2}{\beta} C(d) = 0, \quad (4)$$

with

d thickness of the membrane (m)
 f radon production rate of the source RS (Bq s^{-1})

D diffusion coefficient for radon in the membrane material ($\text{m}^2 \text{ s}^{-1}$)
 β adsorption coefficient for radon in the membrane material
 V_1, V_2 volumes of the primary and secondary chambers, respectively (including those of the internal measurement chambers of radon monitors RM 1 and RM 2) (m^3)
 S surface area of the membrane (m^2)
 $C_1 = \frac{1}{\beta} C(0)$ steady-state radon concentration in the primary chamber (Bq m^{-3})
 $C_2 = \frac{1}{\beta} C(d)$ steady-state radon concentration in the secondary chamber (Bq m^{-3})
 $\Phi_{\text{in}} = -DS \frac{dC}{dz}(0)$ steady-state radon flux into the membrane (Bq s^{-1})
 $\Phi_{\text{out}} = -DS \frac{dC}{dz}(d)$ steady-state radon flux out of the membrane (Bq s^{-1})

The steady-state radon concentration profile within the membrane is then given by

$$C(z) = \frac{\beta f}{\lambda} \left(\frac{\beta SL \cosh\left(\frac{d-z}{L}\right) + V_2 \sinh\left(\frac{d-z}{L}\right)}{\beta SL(V_1 + V_2) \cosh\left(\frac{d}{L}\right) + (V_1 V_2 + \beta^2 S^2 L^2) \sinh\left(\frac{d}{L}\right)} \right), \quad (5)$$

where $L = \left(\frac{D}{\lambda}\right)^{1/2}$ is the radon diffusion length in the membrane material.

Consequently, the steady-state radon concentrations and radon fluxes into and out of the membrane in the primary and secondary chambers are given by

$$C_1 = \frac{f}{\lambda} \times \left(\frac{\beta SL \cosh\left(\frac{d}{L}\right) + V_2 \sinh\left(\frac{d}{L}\right)}{\beta SL(V_1 + V_2) \cosh\left(\frac{d}{L}\right) + (V_1 V_2 + \beta^2 S^2 L^2) \sinh\left(\frac{d}{L}\right)} \right), \quad (6)$$

$$C_2 = \frac{f}{\lambda} \times \left(\frac{\beta SL}{\beta SL(V_1 + V_2) \cosh\left(\frac{d}{L}\right) + (V_1 V_2 + \beta^2 S^2 L^2) \sinh\left(\frac{d}{L}\right)} \right), \quad (7)$$

$$\Phi_{\text{in}} = \frac{\beta f SL [V_2 \cosh\left(\frac{d}{L}\right) + \beta SL \sinh\left(\frac{d}{L}\right)]}{\beta SL(V_1 + V_2) \cosh\left(\frac{d}{L}\right) + (V_1 V_2 + \beta^2 S^2 L^2) \sinh\left(\frac{d}{L}\right)}, \quad (8)$$

$$\Phi_{\text{out}} = \frac{\beta f S L V_2}{\beta S L (V_1 + V_2) \cosh\left(\frac{d}{L}\right) + (V_1 V_2 + \beta^2 S^2 L^2) \sinh\left(\frac{d}{L}\right)} \quad (9)$$

Combining Eqs. (6) and (7) it is finally found that the diffusion and adsorption coefficients for radon in the tested membrane material can be evaluated from the following expressions:

$$\cosh\left(\frac{d}{L}\right) = \frac{C_1}{C_2} \left[1 - \frac{1 - \left(\frac{C_2}{C_1}\right)^2}{\frac{V_1}{V_2} \left(\frac{f}{\lambda V_1 C_1} - 1\right) + 1} \right], \quad (10)$$

$$\beta = \frac{V_2 \sinh\left(\frac{d}{L}\right)}{S L \left(\frac{C_1}{C_2} - \cosh\left(\frac{d}{L}\right)\right)}. \quad (11)$$

3. Results

Figs. 2–4 show the dependence of the steady-state radon concentrations C_1 and C_2 in the air

on both sides of the tested membrane as well as of the ratio $\frac{C_2}{C_1}$ on the diffusion coefficient for radon D in the membrane material for two typical membrane thickness, 0.5 and 1 mm, and different radon adsorption coefficients β assuming $f = 0.1 \text{ Bq s}^{-1}$, $V_1 = V_2 = 8 \times 10^{-3} \text{ m}^3$ and $S = 4 \times 10^{-2} \text{ m}^2$.

As it is seen in these figures, for a given membrane thickness the radon concentration C_1 in the primary chamber decreases and that C_2 in the secondary chamber increases as D increases. Hence, the ratio $\frac{C_2}{C_1}$, independent of f and V_1 , increases with D and with β . When the radon diffusion length L becomes greater than the membrane thickness $\frac{C_2}{C_1}$ approaches unity. More exactly, for a given membrane of thickness d and surface area S the ratio $\frac{C_2}{C_1}$ reaches a value 0.95 when approximately $D = D_0 = 10 \lambda d^2 \left(1 + \frac{2V_2}{\beta S d}\right)$. It must be emphasized that for D values higher than D_0 the standard method for testing the insulation effectiveness of a material as a radon barrier should not be applied in any case because no reliable determination of the radon diffusion and adsorption

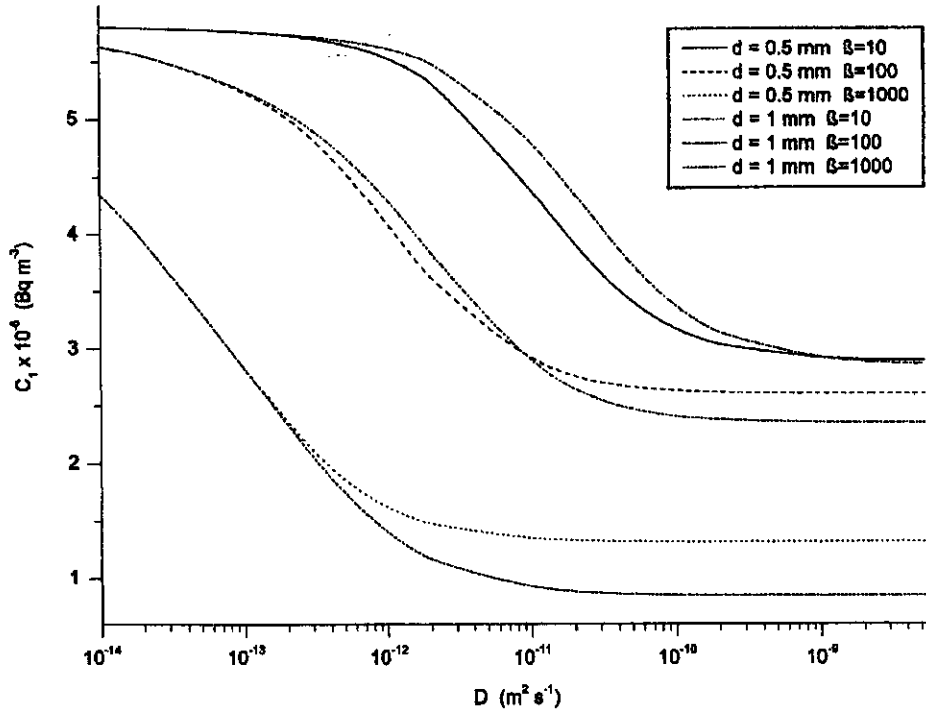


Fig. 2. Steady-state radon concentration in the primary chamber as a function of the diffusion coefficient for radon in the membrane material (parameter values: $f = 0.1 \text{ Bq s}^{-1}$, $V_1 = V_2 = 8 \times 10^{-3} \text{ m}^3$, $S = 4 \times 10^{-2} \text{ m}^2$).

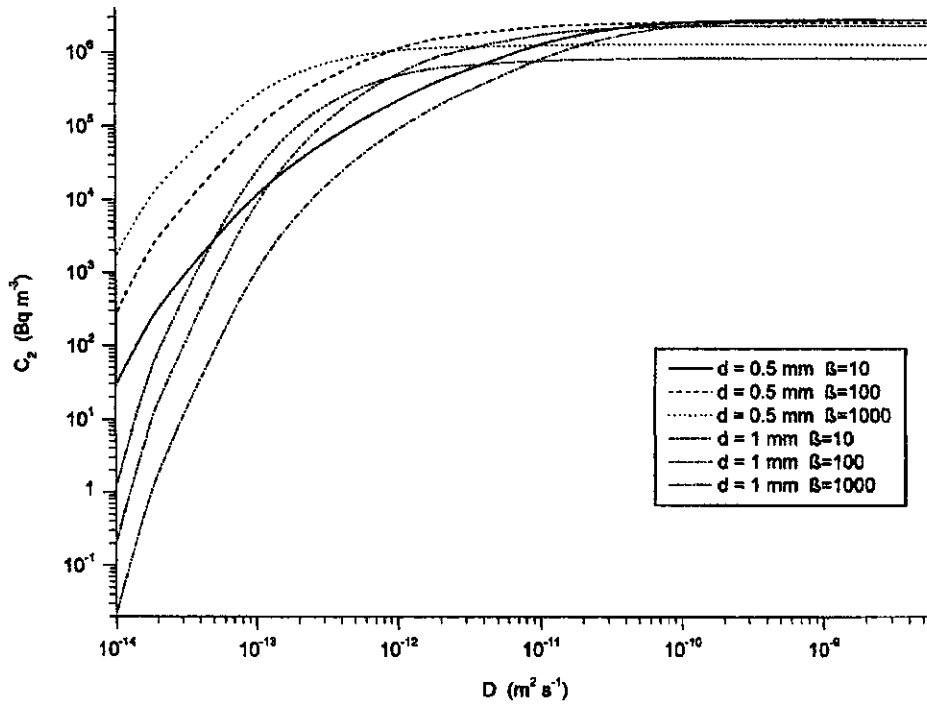


Fig. 3. Steady-state radon concentration in the secondary chamber as a function of the diffusion coefficient for radon in the membrane material (parameter values: $f = 0.1 \text{ Bq s}^{-1}$, $V_1 = V_2 = 8 \times 10^{-3} \text{ m}^3$, $S = 4 \times 10^{-2} \text{ m}^2$).

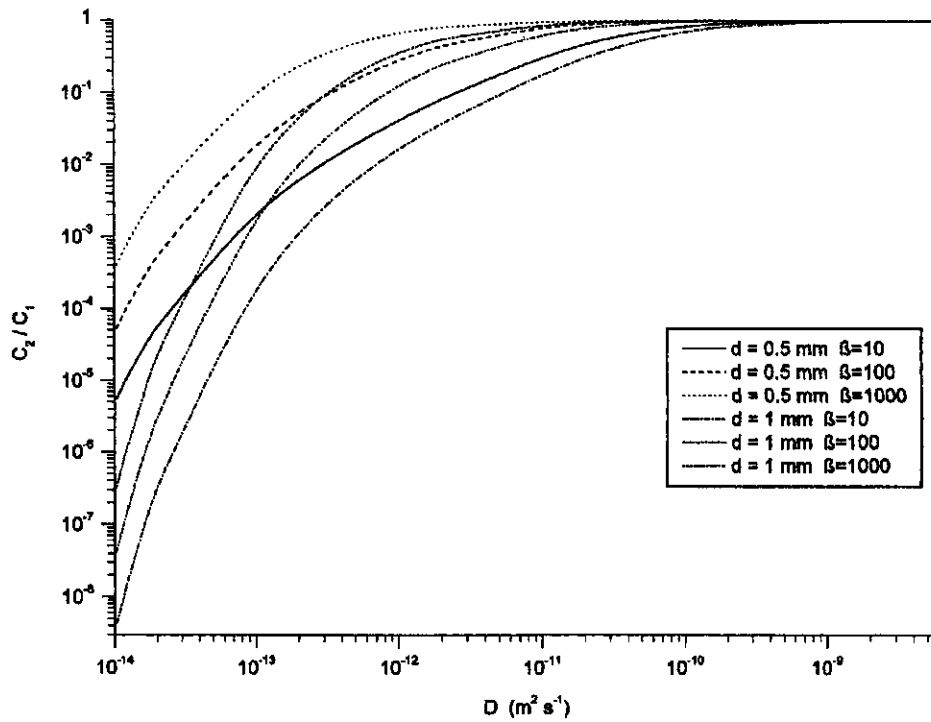


Fig. 4. Ratio between steady-state radon concentrations in the secondary and primary chambers as a function of the diffusion coefficient for radon in the membrane material (parameter values: $V_2 = 8 \times 10^{-3} \text{ m}^3$, $S = 4 \times 10^{-2} \text{ m}^2$).

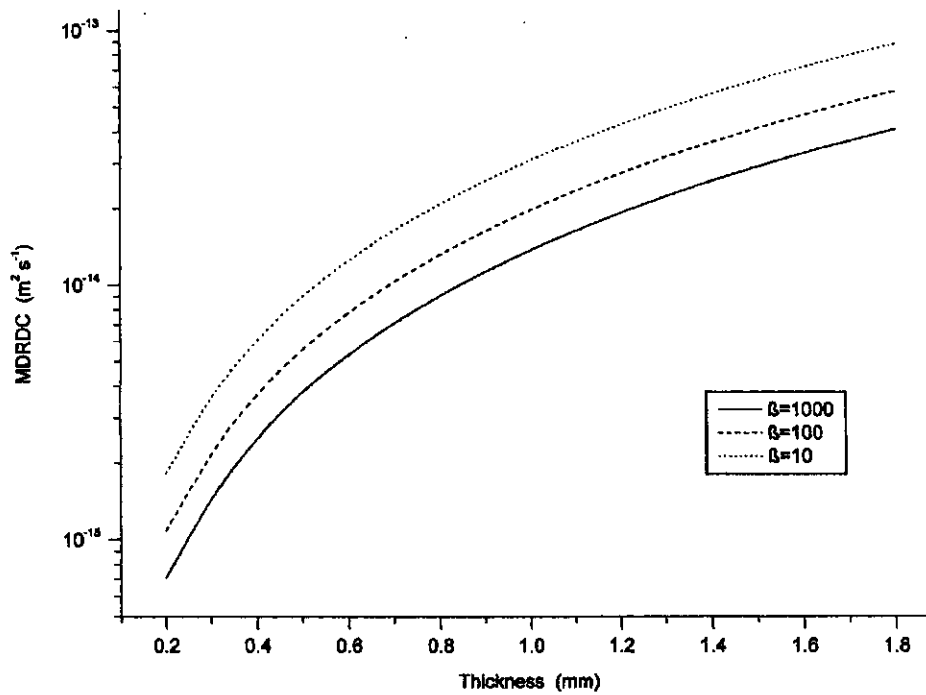


Fig. 5. Dependence of the minimum detectable radon diffusion coefficient MDRDC on the thickness of the tested membrane (parameter values: $f = 0.1 \text{ Bq s}^{-1}$, $V_1 = V_2 = 8 \times 10^{-3} \text{ m}^3$, $S = 4 \times 10^{-2} \text{ m}^2$).

coefficients can ever be made from Eqs. (10) and (11).

It can also be appreciated in Figs. 2 and 3 that for a given membrane thickness the steady-state radon concentration C_1 in the primary chamber always decreases as the adsorption coefficient increases, whereas in the case of the radon concentration C_2 in the secondary chamber, as derived from Eq. (7), a β value for each diffusion coefficient exists, $\beta = \frac{1}{S} \left(\frac{2H_1E_2}{D} \right)^{1/2}$, above which C_2 decreases as β increases because the gas adsorption effects on the membrane predominate over those due to diffusion. This increases the total radon activity absorbed within the membrane and reduces the radon flux out of the membrane in the secondary chamber.

The minimum detectable radon diffusion (MDRDC) and adsorption (MDRAC) coefficients of the method for membrane testing analysed as a function of the thickness of the tested membrane are shown in Figs. 5 and 6 for different values of β and D , respectively, $f = 0.1 \text{ Bq s}^{-1}$, $V_1 = V_2 = 8 \times 10^{-3} \text{ m}^3$ and $S = 4 \times 10^{-2} \text{ m}^2$, assuming that the minimum detectable radon concentration in the air

of the secondary chamber is 20 Bq m^{-3} for time integration periods of 2 h. With these parameter values the minimum detectable radon adsorption coefficient in a typical membrane 1 mm thick is estimated to range between 0.16 and 2×10^{-3} for D values from 10^{-13} to $10^{-12} \text{ m}^2 \text{ s}^{-1}$ and the minimum detectable radon diffusion coefficient between 3.2×10^{-14} and $1.5 \times 10^{-14} \text{ m}^2 \text{ s}^{-1}$ for β values from 10 to 1000, well below those measured in the most frequently used radon insulation materials [4,9,11].

For a given membrane material the uncertainties in the radon diffusion length and adsorption coefficient determined from Eqs. (10) and (11) are mainly influenced by the volumes of the primary and secondary chambers and by the thickness and surface area of the tested membrane so that they can be significantly improved in practice by decreasing V_1 and V_2 and increasing d and S . As an example, the combined standard uncertainties in the diffusion length and adsorption coefficient for radon in a membrane 1 mm thick are estimated to be from 10.5% to 72% and from 14% to 96%, respectively, for L between 2×10^{-4} and $2 \times 10^{-3} \text{ m}$

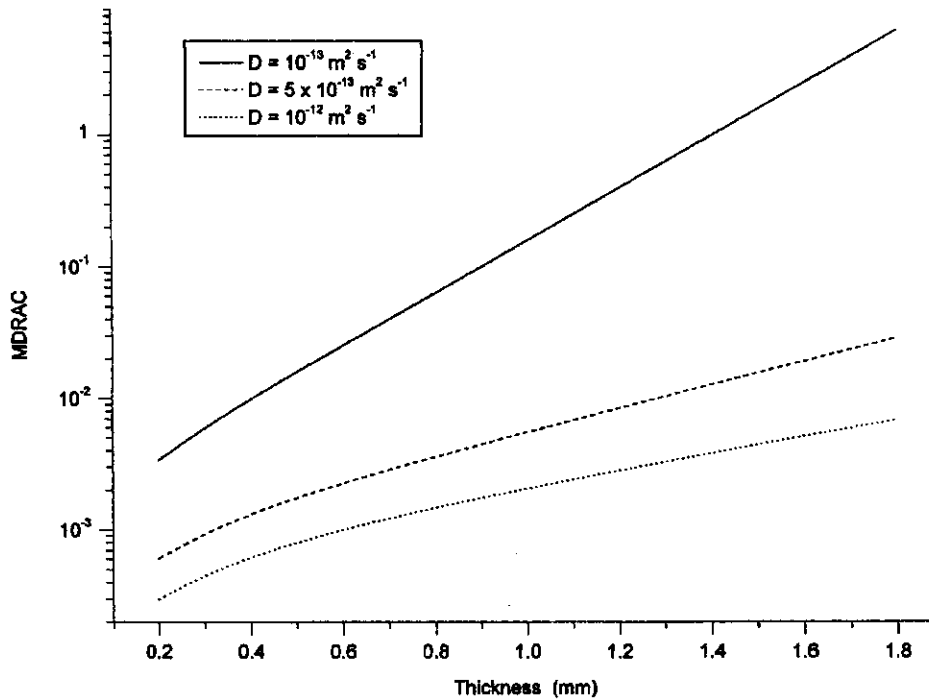


Fig. 6. Dependence of the minimum detectable radon adsorption coefficient MDRAC on the thickness of the tested membrane (parameter values: $f = 0.1 \text{ Bq s}^{-1}$, $V_1 = V_2 = 8 \times 10^{-3} \text{ m}^3$, $S = 4 \times 10^{-2} \text{ m}^2$).

and β between 1000 and 10, using the parameter values mentioned above with typical uncertainties of 4% for f , 2% for V_1 , V_2 , S and C_1 from 9×10^5 to $6 \times 10^6 \text{ Bq m}^{-3}$, 10% for d and between 6% and 2% for C_2 from 7×10^2 to $8 \times 10^5 \text{ Bq m}^{-3}$.

Instead of radon diffusion and adsorption coefficients, two other parameters may be used to characterize the radon insulation effectiveness of a membrane: the radon flux transmission factor RFT and the radon permeability coefficient P . The radon flux transmission factor is defined as the ratio between the radon fluxes out of and into the membrane when the gas diffusing through the membrane desorbs into an infinite volume (radon free desorption). Making $V_2 \rightarrow \infty$ in Eqs. (8) and (9) we obtain that for a membrane of thickness d with a diffusion length for radon L ,

$$\text{RFT} = \frac{1}{\cosh\left(\frac{d}{L}\right)}. \quad (12)$$

As it is seen in Fig. 7 showing the dependence of RFT on the radon diffusion coefficient D for

membrane thickness 0.5 and 1 mm, the radon flux transmission factor increases as d decreases and D increases reaching values higher than 0.99 in membranes of thickness d with radon diffusion coefficients over approximately $D = 50\lambda d^2$.

With respect to the permeability coefficient for radon in a given membrane, it is usually defined in the literature as the proportionality constant between the radon flux density out of the membrane in the secondary chamber and the ratio between the difference in radon concentration in the air on both sides of the membrane and the membrane thickness. From Eqs. (6), (7) and (9) it is easily shown that

$$\frac{\Phi_{\text{out}}}{S\left(\frac{C_1 - C_2}{d}\right)} = \frac{\lambda\beta dL}{\sinh\left(\frac{d}{L}\right) + \frac{\beta SL}{V_2}\left(\cosh\left(\frac{d}{L}\right) - 1\right)}. \quad (13)$$

According to this equation the above definition of the radon permeability coefficient has to be considered rather inappropriate and should be avoided, especially in radonproof membranes. The

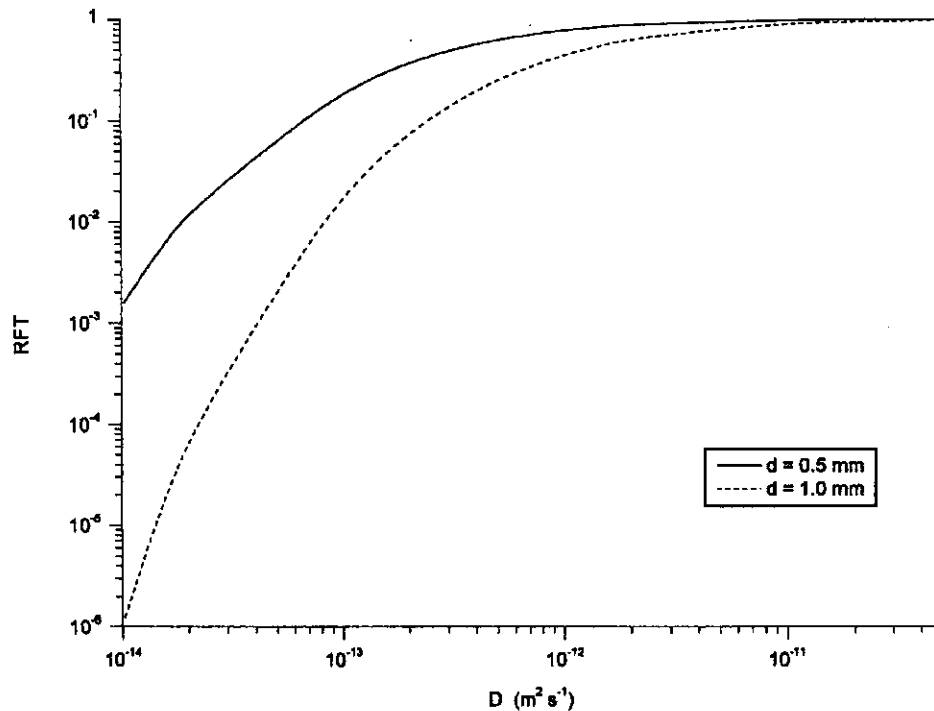


Fig. 7. Relationship between radon flux transmission factor RFT and diffusion coefficient D .

reason is that it implicitly involves that such a permeability coefficient is a parameter characteristic of each membrane which is not the case. As shown in Eq. (13), it also depends on the volume of the secondary chamber and the surface area of the tested membrane sample. Furthermore, it wrongly assumes that the radon concentration gradient within the membrane is constant, a condition which is not fulfilled for radonproof membranes due to the short diffusion length for radon in the membrane material as compared to the sample thickness.

A more suitable definition for the radon permeability coefficient P of a membrane is proposed as the proportionality constant, for $V_2 \rightarrow \infty$, between the radon flux density out of the membrane and the ratio between the concentration difference on both sides of the membrane and the membrane thickness. In that case Eq. (13) becomes

$$P = \frac{\lambda \beta d L}{\sinh\left(\frac{d}{L}\right)}. \quad (14)$$

For $\lambda = 0$ (in practice for approximately $\lambda \leq \frac{0.06D}{d^2}$) Eq. (14) reduces to the well known expression $P = \beta D$ of the permeability coefficient for any non-radioactive gas in a given membrane material, but for a relatively short-lived radioactive gas like radon it also takes into account that the diffusion length for the gas may be of the same order of magnitude or even lower, 10^{-4} – 10^{-3} m, than the thickness of the tested membrane. In that case the permeability coefficient will be a parameter characteristic of the membrane which depends not only on the intrinsic properties of the membrane material through the radon diffusion and adsorption coefficients but also on the membrane thickness. In short, the product βD represents the maximum permeability coefficient for radon in a given membrane with radon adsorption and diffusion coefficients β and D , respectively, and thickness $d \leq 0.245\left(\frac{D}{\beta}\right)^{1/2}$.

In particular, Fig. 8 shows the relationship between the radon permeability and diffusion coefficients for two typical membrane thickness, 0.5 and 1 mm, and different radon adsorption coefficients.

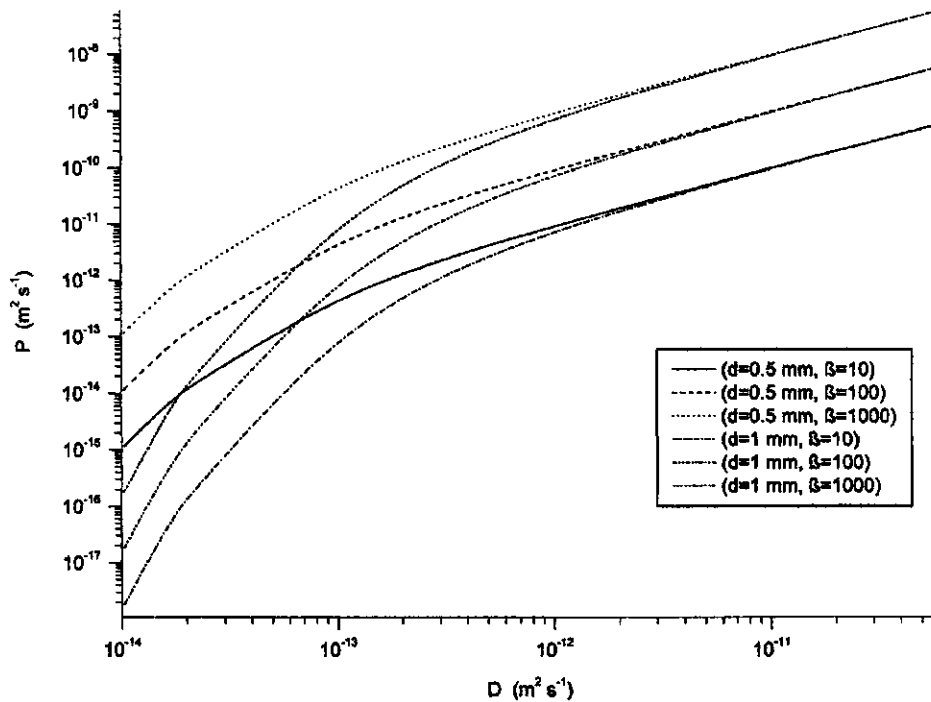


Fig. 8. Relationship between radon permeability P and diffusion D coefficients.

It can be seen that the permeability coefficient for radon increases as the thickness of the membrane decreases and D and β increase. For a given membrane thickness d the increase in the radon permeability coefficient with D becomes almost linear over $D = 1.3\lambda d^2$, reaching about 99% of its maximum value βD in membrane materials with radon diffusion coefficients $D = 16\lambda d^2$, approximately.

4. Conclusion

A theoretical approach to the measurement of radon diffusion and adsorption coefficients in radonproof membranes has been described. It is based on the application of the standard method of tightly enclosing the membrane between two compartments of the same or different volumes. Theoretical expressions for evaluating both coefficients have been derived from the steady-state solution of the differential equation for radon diffusion through the membrane. Minimum detectable radon diffusion and adsorption coeffi-

cients of the method are estimated to be typically well below those measured in the most frequently used radon insulation materials.

Finally, besides the radon diffusion and adsorption coefficients a new parameter called radon flux transmission factor and a more suitable definition of the radon permeability coefficient than usually given in the literature are also proposed as an alternative way of characterizing the insulation effectiveness of any membrane as a radon barrier.

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