MEASUREMENT OF RADON DIFFUSION LENGTH IN THIN MEMBRANES

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Building regulations in Israel require the insulating of buildings against radon $^{222}$Rn penetration from soil. In radon-prone areas membranes stretched between the soil and the building foundation are used, together with sealing other possible penetration routes. Designing the radon mitigation procedure requires checking that all sealing materials are practically radon tight, having a thickness of at least three times the radon diffusion length. In this work, a very simple technique to evaluate the radon diffusion length in thin membranes, using a radon source of known activity and an activated charcoal canister as radon detector is presented. The theoretical formalism and measurement results for polyethylene membranes of different densities obtained in a recent comparison exercise are presented.

INTRODUCTION

Radon ($^{222}$Rn) is known to diffuse through porous building materials such as concrete, cement, paints, etc. and therefore it may penetrate from soil through slabs and walls and accumulate in dwellings(1, 2). Basements and shelters should use building materials or membrane coverings to block radon penetration. The choice of membrane depends, among other factors such as environmental stability, on its radon diffusion length. A membrane thickness equal to at least three diffusion lengths is considered radon tight(3–5).

Building regulations in Israel require applying radon mitigation techniques in radon-prone areas by insulating the building from soil by radon-tight membranes. The radon-insulating quality of the membranes should be checked by a certified laboratory. The purpose of the present work was to set up a simple radon diffusion length measurement method and to check its performance in an international comparison exercise. The measurements results are presented.

FORMALISM AND PROCEDURES

Radon flow through a medium is described by the diffusion equation:

$$L^2 \frac{\partial^2 C(x,t)}{\partial x^2} - C(x,t) = \frac{1}{\lambda_{Rn}} \frac{\partial C(x,t)}{\partial t}$$

written here for the one-dimensional case and neglecting radon production in the medium. $C(x,t)$ is the radon concentration (in Bq m$^{-3}$) at point $x$ (in m) and time $t$ (in s), $\lambda_{Rn} = 2.1 \times 10^{-6}$ s$^{-1}$ is the radon decay constant, calculated as $\lambda_{Rn} = \ln(2)/T_{Rn}$ with $T_{Rn} = 3.82$ d the $^{222}$Rn half-life and $L$ is the radon diffusion length (in m), related to $D$ the diffusion coefficient (in m$^2$ s$^{-1}$) by the relation:

$$L = \sqrt{\frac{D}{\lambda_{Rn}}}$$

Figure 1 shows schematically the experimental setup for the diffusion length measurement in this study. Radon atoms emitted from a radon source in chamber 1 diffused via the partition material into chamber 2. The partition to be a homogenous diffusive medium with a surface area much larger, compared with its thickness, such as to apply Eq. (1) was considered. The equilibrium state solution is as follows(3):

$$C(x) = \frac{C_1 \sinh(\ell - x/L) + C_2 \sinh(x/L)}{\sinh(\ell/L)}$$

where $C(x)$ is the radon concentration (in Bq m$^{-3}$) inside the partition material at distance $x$ (in m) from entry surface, $C_1$ and $C_2$ are the constant radon concentrations (in Bq m$^{-3}$) in chambers 1 and 2, respectively, $\ell$ is the thickness of the partition (in m) and $L$ is the radon diffusion length (in m) in the partition material. Physically, the material is not homogeneous: radon atoms penetrate it by diffusing between the air molecules contained in its pore space. Technically,
In order to calculate the radon atom flow rate at the partition equals the loss rate by radioactive decay. In the partition, the flow rate from chamber 1 equals by radioactive decay and by flow into the partition. In each of the three volumes, the radon emission rate from source equals the loss rates between the two chambers and the chambers from the two volumes of chambers 1 and 2 (in m$^3$), respectively, $\ell$ the partition thickness (in m), $S_M$ the partition surface area (in m$^2$), $V_M = \ell S_M$ the partition volume (in m$^3$) and $a = \ell/L$ the ratio between the partition thickness $\ell$ and the diffusion length $L$.

The above parameters are divided into three groups: (1) experimental setup parameters $V_1$, $V_2$, $\ell$ and $S_M$ which are fixed and measurable with small uncertainty, (2) physical parameters $A_S$, $C_1$, $C_2$ and $A_M$ which are measured by detectors with higher uncertainty and (3) finally the unknown $L$, to be calculated mathematically.

The diffusion length $L$ may be evaluated in several ways: (1) from Eq. (4) by measuring $C_1$ and $A_S$, (2) from Eq. (5) by measuring $C_2$ and $A_S$, (3) from Eq. (6) by measuring $C_1$ and $C_2$ or by replacing $C_1$ and $C_2$ from Eqs (4) and (5) and measuring $A_S$ or $a$ (by dividing Eq. (4) by (5):

$$\frac{C_1}{C_2} = \frac{V_2}{V_M} \cdot \frac{a \cdot \sinh(a) + \cosh(a)}{a \cdot \sinh(a) + \cosh(a)}$$  \hspace{1cm} (7)

and measuring $C_1$ and $C_2$. In case (3) the partition material activity $A_M$ is usually very low. In order to be measurable with acceptable uncertainty one should use a very strong radon source and a very low detection limit measurement method. For example, by using high purity germanium (HPGe) spectrometry, $A_M$ should be larger than the detection limit of a few tenth of 1 Bq.

In order to measure $C_1$ and $C_2$ one needs to keep the system closed for a long enough time to reach the equilibrium state. One may determine it theoretically by solving the time-dependent Eq. (1) using a range of expected $L$ values or experimentally by measuring the radon concentration in both chambers every few hours using a continuous radon monitor (CRM) in each chamber, which would be the ideal measurement method. A simpler method would be to measure $C_1$ and $C_2$ at equilibrium time by taking small air assays from the chambers or by passive detectors, inserted in the chambers at closure time. One may estimate a lower limit of the equilibrium
time by simple arguments. The time necessary to reach equilibrium diffusion flow in the partition may be evaluated\(^{(1, 2)}\) from Eq. (1) \(A_S t_{\text{diff}} = \frac{|T_{Rn}/\ln(2)|}{[1 + (\pi L/\ell)]}.\) It varies between 0, in the limit of a thin partition (\(\ell << L\)) and maximum 5.5 d in the limit of a thick partition (\(\ell >> L\)). In a typical measurement, when \(L \approx \ell\), it would be 0.5 d. The equilibrium time of the radon concentration in the chambers is practically \(4 T_{Rn} = 15.4\) d, during which the radon concentration reaches 94 \% of the equilibrium value. In the present measurements, the chambers are closed for a minimum of 21 d in order to ensure reaching the equilibrium state.

A precise radon diffusion measurement method should be designed theoretically, before constructing the experimental setup, by using Eqs. (4)–(6) and assuming a range of expected \(L\) values. This procedure allows the predicting and optimising of the radon concentrations in the chambers, as a function of source activity and chamber volumes, allowing the choice of the optimal detector type, based on its specifications: detection limit, measurement range and accuracy.

The concentrations \(C_1\) and \(C_2\) may be measured by a variety of detectors, such as electronic continuous radon monitors (CRM) or passive detectors such as alpha track—solid state nuclear track detector (AT), electret (EL), activated charcoal (AC) canister or liquid scintillator AC vials (LS). The CRM, AT or EL detectors measure the radon concentration without perturbing it and therefore may be used in both chambers. An AC AC or LS detector may be used only in chamber 2. When setting it in chamber 1 it would adsorb most of the radon released by the source and will lead to a very low concentration in chamber 2, measurable with large uncertainty or below the detection limit of 5–10 Bq m\(^{-1}\), estimate the maximal concentration \(C_1 = A_S/S\) and use a detector with an adequate range. For example, when using an EL detector in chamber 1 one should ensure \(C_1\) below about 30 000 Bq m\(^{-1}\), its maximal range in LLT configuration for a 21 d exposure. When measuring \(C_2\) one should use a larger source activity (\(A_S\) equal to a few thousand Bq) to produce a larger activity in chamber 2, at least a few hundred Bq m\(^{-1}\), to ensure a precise measurement. One may use AT detectors in both chambers, which have a larger measurement range up to about 100 000–150 000 Bq m\(^{-1}\) using CR39 plastic and higher using polycarbonate films. Finally, one may choose the chamber volume in order to fit the expected concentration to the EL or AT detector measurement range. The measurement range of the AC or LS detectors is much larger. The activity of the AC detector may be measured by HPGe (or NaI) spectrometry and that of the LS detector by beta spectrometry from the detection limit of a few tenth of 1 Bq up to several thousands Bq, higher than attained in chamber 2 using the radon sources mentioned above.

Eqs (4)–(7) are transcendental and thus the diffusion length \(L\) needs to be calculated by iterations. For this purpose, one inputs the values of the setup parameters \(V_1, V_2, \ell\) and \(S_M\) and those of the measured quantities \(C_1, C_2, A_S\) and \(A_M\) and varies the value of the diffusion length \(L\) till there is agreement between the calculated and the measured values. Since the setup parameters may be determined with much lower uncertainty as compared with the measured quantities, the theoretical diffusion length measurement uncertainty is determined, essentially, by the uncertainty of the measured quantities. In the actual measurement, there are also systematic errors due to changes in the environmental conditions, which may influence the source activity, for example possible radon leaks from the chambers and around the partition sealing which may influence the radon concentrations, etc. which need to be minimised by careful setup construction and checking.

In the present measurement, \(L\) from Eq. (5) was estimated by measuring the radon concentration in chamber 2 by a single AC canister. The uncertainty in the diffusion length \(L\) measurement result may be calculated numerically from Eq. (5) by varying \(A_S\) and \(C_2\) in the range of their measurement uncertainties of about 10 \%, resulting in a theoretical diffusion length uncertainty of about 15 \%. In the measurements here, the standard deviation of the diffusion length measurement results was found to be about 25 \%, dominated by systematic uncertainties. Therefore, the experimental uncertainty as standard deviation of a series of measurements done for the same partition material was reported.

**MEASUREMENT OF THE RADON CONCENTRATION BY AN AC CANISTER**

Normally, AC detectors, AC canisters or LS vials, are used for short-term (2–7 d) radon concentration measurements in rooms under closed conditions. They work by adsorbing a very small radon quantity as compared with the total radon quantity in the room and thereby do not change its concentration. For this reason they are considered ‘passive’ like the AT and EL detectors which measure the radon concentration without disturbing it. However, measuring the radon concentration in a small chamber by an AC detector requires a special approach.

When set in a small chamber, the AC detectors strongly adsorb most of the radon and thereby lower significantly the radon concentration. At equilibrium, the source activity \(A_S\) (in Bq) is equal to the sum:

\[
A_S = A_{\text{air}} + A_{\text{AC}}
\]

(8)
between the activity in the air $A_{\text{air}}$ (in Bq) and in the charcoal $A_{\text{AC}}$ (in Bq). The effective volume of the charcoal $V_{\text{AC}}$ (in m$^3$) is defined by the relations:

$$C_{\text{air}} = \frac{A_{\text{air}}}{V_{\text{air}}} = \frac{A_{\text{AC}}}{V_{\text{AC}}}$$  \hspace{1cm} (9)

where $C_{\text{air}}$ (in Bq m$^{-1}$) is the radon concentration in the air and $V_{\text{air}}$ (in m$^3$) is the free (unoccupied by objects) air volume in the chamber. From Eqs (8) and (9), the fraction of the radon activity in the air is:

$$\frac{A_{\text{air}}}{A_{\text{S}}} = \frac{V_{\text{air}}/V_{\text{AC}}}{1 + (V_{\text{air}}/V_{\text{AC}})}$$ \hspace{1cm} (10)

and the fraction in the charcoal is:

$$\frac{A_{\text{AC}}}{A_{\text{S}}} = \frac{1}{1 + (V_{\text{air}}/V_{\text{AC}})}$$ \hspace{1cm} (11)

As an example, suppose that a radon source and an AC canister containing 70 g of charcoal in a small chamber of volume $V_{\text{air}} = 0.002$ m$^3$ is hermetically sealed. With the typical effective volume of the charcoal of about 0.002 m$^3$ per gram$^{[9]}$ the effective volume of canister is about $V_{\text{AC}} = 0.140$ m$^3$. From Eqs (10) and (11) it is estimated that 1% of the radon emitted by the source will be in the air and 99% will be in the canister. Formally, the charcoal ‘enlarges’ the geometrical volume of the chamber by its own effective volume. Therefore, when using Eqs (4)–(7) to estimate the radon diffusion length and the radon concentrations $C_1$ and $C_2$ are measured by an AC detector of effective volume $V_{\text{AC}}$, one should replace the chamber geometrical free air volumes $V_1$ by the sum $V_1 + V_{\text{AC}}$ and $V_2$ by the sum $V_2 + V_{\text{AC}}$, respectively.

In the present measurement, a commercially available 4-inch diffusion barrier AC canister containing 70 g of AC was used. Due to its large effective volume it collected most of the radon in the chamber and accumulated a relatively large activity which could be measured precisely. This allowed a precise measurement of the radon concentration in the chamber using Eq. (9). The canister activity was measured by an HPGe spectrometer, capable of measuring activities down to the detection limit of a few tenths of 1 Bq. If the canister activity is higher, above a few Bq, a less expensive NaI spectrometer would measure it with sufficient precision.

**RADON SOURCE PREPARATION**

As a radon source one may use a commercially available calibrated source or may prepare it in the laboratory. Radon sources may be easily prepared by using a $^{226}$Ra rich material with high radon emanation, like natural phosphate soil or phosphogypsum (PG). Several radon sources using different types of PG with masses between 0.2 and 4 kg have been prepared. For example, an open jar containing 1 kg of dry PG with $^{226}$Ra concentration of 4000 Bq kg$^{-1}$ and radon emanation of 10% provides a simple radon source with activity of 400 Bq. Its activity may be increased by adding water and homogenising the material. Depending on the PG type, the radon activity may increase up to a maximum of three to four times the dry source activity when the water content is about 30%. In order to ensure constant activity, it is necessary to control its weight or to seal the jar on top by a thin plastic membrane with high radon transmission and low water vapor transmission.

The radon source activity may be measured simply by sealing it hermetically in a small container together with EL, AT, AC, LS or CRM detectors for a short time of 3–4 d. As mentioned, after sealing the container, the radon concentration in the container starts growing and after 4 $T_{\text{Ra}}$ (15.4 d) it reaches practically the equilibrium value.

When using an EL or AT detector, the short exposure is recommended to prevent the EL discharge or the AT saturation due to the high radon concentration, in the case of a strong source. The source activity $A_{\text{S}}$ may be measured as the product between the equilibrium radon concentration and the free (unoccupied by objects) air volume $V_{\text{air}}$ in the container. The EL and AT detectors measure the mean concentration over the exposure time. The equilibrium concentration may be calculated by extrapolating the measured mean concentration $C_{\text{air}}(t)$ at short time $t$ to the equilibrium value$^{[10]}$.

The source activity may be calculated as:

$$A_{\text{S}} = \frac{C_{\text{air}}(t) V_{\text{air}}}{1 - (1 - \exp(-\lambda_{\text{Rn}} t))/(\lambda_{\text{Rn}} t)}$$ \hspace{1cm} (12)

When using an AC canister, the short exposure is necessary only when the source is wet and open in order to prevent a large (more than 1–2 g) water accumulation in the canister which would, on the one hand, dry the source and reduce its activity, or on the other hand, wet the charcoal and reduce its effective volume. The canister activity may be measured by an HPGe spectrometer or by a less expensive NaI spectrometer. As mentioned, when the container volume is much smaller than the AC canister effective volume, the source activity $A_{\text{S}}$ is practically equal to the equilibrium canister activity, which may be calculated by a simple extrapolation from the activity $A_{\text{AC}}(t)$ measured at short time $t$:

$$A_{\text{S}} \approx \frac{A_{\text{AC}}(t)}{1 - \exp(-\lambda_{\text{Rn}} t)}$$ \hspace{1cm} (13)
When the container volume is comparable to or larger than the AC effective volume, Eq. (13) should be replaced by a more general formula (8).

MEASUREMENT OF THE CHARCOAL CANISTER EFFECTIVE VOLUME

The effective volume of the AC canister may be measured relatively simply, based on its definition in Eq. (9), by sealing it hermetically in a small container together with a passive (EL or AT) radon detector for a duration of at least 4 $T_{Rn} = 15.4 \text{ d}$ in order to reach the equilibrium state. One should use a dry radon source to prevent water accumulation in the canister. The effective volume may be evaluated from Eq. (9) as the ratio between the canister activity and radon concentration in the air $V_{AC} = A_{AC} / C_{air}$ measured at equilibrium. In this work, a commercially available diffusion barrier 4-inch AC canister containing 70 g of AC was used. An effective volume of 0.150 m$^3$ was measured.

EXPERIMENTAL SETUP

Figure 2 illustrates a typical experimental setup for the radon diffusion length measurement. The glass jar shown inside the larger plastic container is filled with a strong radon source in the form of PG powder. Wet PG in order to enhance the source activity was used. The source jar was sealed on top with the membrane to be measured using 1–2-cm thick regular plasticine clay (coloured). An improved apparatus would preferably seal the membrane using O-rings. The larger plastic (plexiglass) container was sealed from the atmosphere using O-rings. After closing the system, radon atoms from the source jar diffuse through the membrane into the larger container. The black EL detectors (long-term LLT configuration) measure the radon concentration in both chambers. In some of the measurements, AT detectors (rather than EL detectors, which have lower measurement range) in both chambers in order to measure the radon concentration and to compare it with the prediction of the theoretical model were included. A good agreement within 10–15 % was observed. The large 4-inch diffusion barrier AC canister measures the radon concentration in the larger container. As mentioned, the system was kept closed for at least 21 d.

The radon diffusion length in the membrane from Eq. (5) by using a radon source of known activity and by measuring the activity of the AC canister was measured. The methods for preparing the source and measuring its activity and for measuring the canister effective volume and activity were described above. Diffusion length measurements were done for three membrane types of different densities. Each membrane type was measured in three different laboratories, several times, using the same basic experimental setup shown in Figures 1 and 2 but with different parameters, i.e. with different volumes $V_1$ and $V_2$ and different source activities $A_S$ leading to different concentrations $C_1$ and $C_2$. The volumes $V_1$ and $V_2$ were chosen to be small to allow reaching high radon concentrations and therefore better measurement accuracy. Measurements were done with two basic setup configurations: (1) with the membrane covering the source container and (2) with the membrane covering the canister. The optimal configuration is the first, since it results in a higher canister activity and better measurement accuracy. In the second configuration, the canister activity is lower, but there is no need to estimate $V_2$ since it is practically equal to the canister effective volume $V_{AC}$.

MEASUREMENTS AND RESULTS

The radon diffusion in thin membranes as part of an international comparison exercise organised by the National Radiation Protection Institute of Prague, who provided sample membranes to all the participants, was measured. The conference report (11) describes in detail, for each participant, the experimental procedure, the used equations for diffusion length ($L$) or diffusion coefficient ($D$) determination and the measurement results.

All the participating laboratories received three types of polyethylene membranes: high density of 950 kg m$^{-3}$ (HDPE), low density of 750 kg m$^{-3}$ (LDPE) and very low density of 500 kg m$^{-3}$ (LDPE) with the same surface area of 78 cm$^2$ and thicknesses of 1.6, 1.0 and 1.1 mm, respectively. A number of identical samples from each membrane type were provided, as requested by the participants.

The measurements reported here were carried out by three laboratories in Israel which worked independently using the two setup configurations (1) and
(2) mentioned above. As radon chambers, a variety of containers were used, made of glass, plastic and metal, with volumes between 0.003 and 0.080 m³ and a variety of PG radon sources with activities between 200 and 800 Bq. The activity of the sources was measured before the diffusion length measurement by using EL detectors and AC canisters as described above. In all measurements, the diffusion length was evaluated from Eq. (5) by measuring the activity of a commercially available 4-inch diffusion barrier AC canister containing 70 g of charcoal shown in Figure 2. All measurements were conducted at normal laboratory temperature of about 25°C on the average. A package of instructions(8), describing the mathematical calculations and the laboratory procedures required for carrying out the measurements, were provided to all the participants to reduce the systematic errors. Our results were previously reported(12).

Table 1 shows the results for the three membrane densities and compares them with the results of the Czech group(11). The first row shows our measured diffusion length \( L \) as a mean over six measurements together with the standard deviation. The second row shows the diffusion coefficient \( D \) calculated from the first row by using Eq. (2). For comparison in the third row the mean measurement result for \( D \) by the Czech group is shown. The Czech measurements were also carried out by using radon diffusion from a source chamber, via the membrane, into a receiver chamber. The diffusion coefficient was determined by an iterative numerical solution of the diffusion equation(11). Our results are in good agreement with those of the Czech group and with most of the reported results in the international exercise(11).

The radon diffusion length (or the diffusion coefficient) is expected to increase with decreasing membrane density. This trend is shown by the results of the Czech group within their small measurement uncertainty. Our results also show this trend between the high density of 950 kg m\(^{-3}\) and the low density range of 750–500 kg m\(^{-3}\), however they cannot clearly resolve the decrease from 750 to 500 kg m\(^{-3}\) due to our higher 25 % measurement uncertainty. As mentioned, our measurement uncertainty was dominated by systematic errors. An improved experimental setup is expected to reduce them. A large spread of the measurement results from different groups was also reported in the international comparison exercise(11).

**CONCLUSIONS**

The purpose of this work was to set up a relatively simple method for measuring the radon diffusion length in thin membranes used for radon mitigation in buildings and to check its performance by participating in an international comparison exercise(11). A theoretical model and its application for constructing the experimental setup and for carrying out measurements using different radon sources and detectors were described. The authors apply it by a simple measurement method using a radon source made of PG and a commercially available 4-inch diffusion barrier AC canister as radon detector. Measurement results for three polyethylene membranes of different densities carried out independently by three laboratories using the same measurement method with different setup parameters was reported. Our results show the expected trend of increasing diffusion length from high-to-low membrane density, however, they cannot resolve it in the low-density range, within the experimental uncertainty. An improved experimental setup is expected to reduce the experimental uncertainty, dominated by systematic errors. The results are in good agreement with most of the reported results in the international comparison exercise(11).

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