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A method for measuring effective radon diffusion coefficients in radon barriers by using modified Lucas cells

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Abstract

Radon proof barriers are used for lowering of radon transport from the soil into the house and the determination of the radon diffusion coefficient is an important parameter to be determined in order to design the minimal thickness of the radon proof insulation. A method has been developed in our laboratory by using modified Lucas cells connected to a radon source and tightly closed onto the top by the tested membranes whose radon diffusion coefficients are being measured. Solving the time-dependent differential equation for radon diffusion in the membrane for well-defined experimental conditions the effective radon diffusion coefficient of the insulating material can be evaluated by comparing the radon concentration decrease in the cell for the first hours with the well-known radioactive decay. First results obtained in several preliminary tests carried out with a parafilm M barrier and two polyethylene membranes are shown in this paper.

Keywords: Radon diffusion coefficient; Radon barriers; Modified Lucas cells

1. Introduction

The increasing interest of people and authorities in reduction of radon gas in houses generates also the question of what kind of insulating materials can be considered as radon proof barriers and if the conventional waterproof insulation creates an effective barrier against the infiltration of radon. To solve this, the measurement of the radon diffussion coefficient is absolutely necessary. The standard method for this determination is based on the evaluation of radon flux through tested material exposed to high radon concentration and placed between two containers by measuring time-varying or steady-state radon concentrations in both sides of the material (Nielson et al., 1982). For this, one of the containers is connected to a radon source which produces there radon concentrations as high as 100 MBq m^{-3} and the other where the radon concentration is periodically evaluated. Nevertheless, in order to minimize uncertainties in the determination of the radon diffusion coefficient with this method the study is normally performed under steady-state conditions. Then, the measurement of the radon concentrations is made after a certain time called "relaxation" time, which depends on the diffusion coefficient and thickness of tested material and can be, normally, more than 15 days. Additional problems appear concerning the maintenance of a constant value for the radon concentration in one of the two containers used in the experiment during this long period time. In this paper we propose a new method for the measurement of the radon diffussion coefficient showing some results achieved in our laboratory.

2. Methods

The method outlined in this paper for evaluating effective radon diffusion coefficients of radon proof membranes is based on the principle of following the decrease in time of the radon concentration initially brought into a modified Lucas cell of 1.1 l capacity (Quindós et al., 1991), previously calibrated in efficiency, comparing this decrease with

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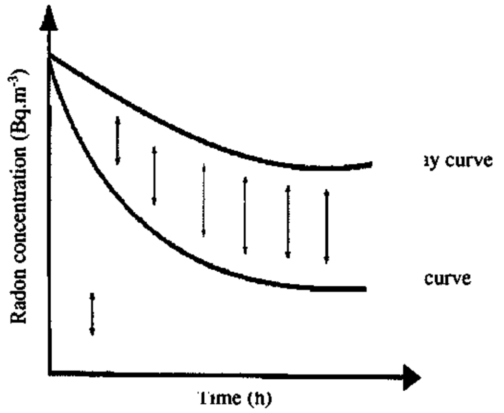

Fig. 1. Principle of the method for diffusion coefficient measurement.

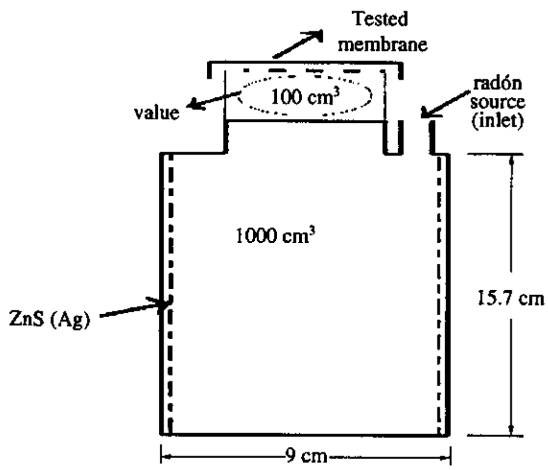

Fig. 2. Experimental device for testing insulating material.

the known curve of radioactive decay for radon as shown in Fig. 1.

In a more general way, the proposed method can be applied to determine at very low cost and quickly, without final steady-state conditions being required, the radon effective diffusion coefficient of any kind of material in which radon diffusion be independent on porosity and moisture content.

The device used is connected to a radon source and tightly closed onto the top by the test insulating material as shown in Fig. 2.

Assuming no radon production in the tested material and no leakage in the cell, the time-dependent one-dimensional differential equation for radon diffusion in a membrane of area S and thickness d placed onto the top of the cell of volume V to be solved is

$$\frac{\partial C(z,t)}{\partial t} = D\frac{\partial^2 C(z,t)}{\partial z^2} - \lambda C(z,t) \tag{1}$$

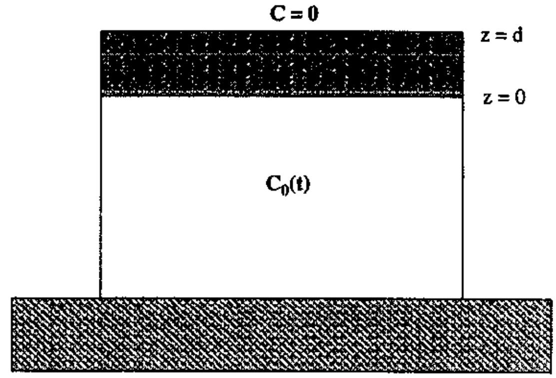

Fig. 3. Schematic set up of radon concentrations in both sides of the insulating material of d mm thick.

with boundary conditions for the schematic set up shown in Fig. 3

$$C(d,t) = 0,$$

$$\frac{dC_0(t)}{dt} = \frac{DS}{V}\frac{dC(z,t)}{dz}\bigg|_{z=d} - \lambda C_0(t), C(0,t) = \beta C_0(t), \tag{2}$$

$$C(z,0) = \begin{cases} \beta C_0(0), & z = 0, \\ 0, & z > 0, \end{cases}$$

where λ is the radon decay constant, $C_0(0)$ the initial inner radon concentration in the cell, and β and D the radon adsorption and diffusion coefficients in the membrane, respectively, which are characteristic of each material and basically depend on temperature.

By using Laplace transforms, it can be easily proved that for $V/Sd \approx 10^3$ or higher, as in the experimental conditions used in our tests with $S = 1.2566 \times 10^{-3}$ m^2 and thickness $d \approx 10^{-4}$–10^{-3} m, the decrease in time of the radon concentration in the cell is accurately describe by

$$C_0(t) = C_0(0)\mathrm{e}^{-(1+D_eS/\lambda Vd)\lambda t}, \tag{3}$$

where $C_0(t)$ is the radon concentration in the cell at any time t, and $D_e = \beta D$ the effective radon diffusion coefficient in the membrane, sometimes also called "radon permeability" (Nilsson, 1999).

Hence, choosing the suitable values for V, S and d to fulfill the aforementioned condition and assuring the applicability of Eq. (3), and once the initial radon concentration in the cell and its decrease after a time interval Δt have been measured, the effective diffusion coefficient for radon in the tested membrane can be easily evaluated as

$$D_e = \frac{Vd}{S\Delta t}\ln\left[\frac{C_0(0)}{C_0(\Delta t)}\mathrm{e}^{-\lambda\Delta t}\right]. \tag{4}$$

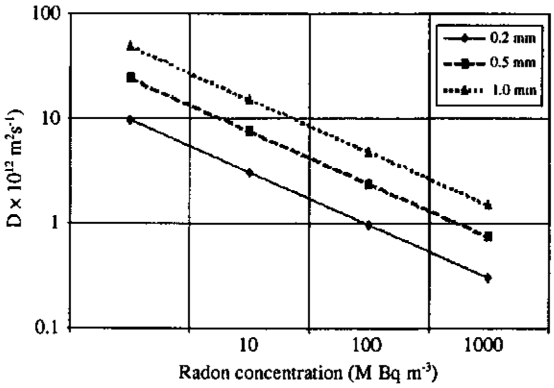

Fig. 4. Detection limit for different experimental conditions.

The minimum effective radon diffusion coefficient which can be measured by this method, calculated on the assumptions that Poisson counting statistics may be approximated to a normal distribution and radon concentration after the selected interval time Δt, $C(\Delta t)$, is significantly less than the initial concentration corrected for decay, $C_0 e^{-\lambda \Delta t}$, at a 95% confidence level ($p = 0.05$), is shown in Fig. 4 as a function of the initial radon concentration in the cell for different membrane thickness in the experimental conditions used in our laboratory corresponding to Δt=12 h, V=1.1×10^{-3} m^3 and $S = 1.2566 \times 10^{-3}$ m^2. It can be seen that for initial radon concentrations of the order 10^2 MBq m^{-3} the minimum measurable effective radon-diffusion coefficient ranges between 5×10^{-13} and 5×10^{-12} m^2 s^{-1} for membranes 0.2 –1 mm thick. Since the effective radon diffusion coefficient in the most commonly supplied radon proof membranes exceeds 10^{-13} m^2 s^{-1}, it can be concluded that the proposed method provides a simple, quick and inexpensive way to evaluate radon diffusion coefficient for most insulating materials used as radon barriers.

3. Results

In order to experimentally check the applicability of the proposed method, several preliminary tests have been conducted in our laboratory with three different barrier films. The first, 0.127 mm thick, from SPI Supplies, Parafilm M, Chicago, Illinois, which is made basically as a mixture of polymers and waxes. The other two were membranes of low, 650 kg m^{-3}, and high density, 950 kg m^{-3}, polyethylene (1.6–0.6 mm thick, respectively) kindly donated by Dr. Martin Jiranek from Czech Technical University, Prague, Czech Republic. Using initial radon concentrations in the cell ranging between 10^5 and 10^7 Bq/m^{-3}, average effective radon-diffusion coefficients at 18°C and 60% relative humidity of $(6.2 \pm 1.1) \times 10^{-10}$, $(3.2 \pm 0.6) \times 10^{-10}$ and $(1.2 \pm 0.2) \times 10^{-10}$ m^2 s^{-1} were estimated for the Parafilm M foil and the low- and high-density polyethylene membranes, respectively. The last two values are in good agreement with those found by Jiránek and Hülka (2001) for the radon diffusion coefficients, on the order of 10^{-11} m^2 s^{-1}, assuming radon adsorption coefficients $\beta \sim 10^1$ for both low- and high-density polyethylene membranes.

Likewise, tests of improved experimental devices based on the same principle of using, modified Lucas cells are also ongoing with the aim of separately measuring both the radon adsorption coefficient and the radon diffusion coefficient in radon barriers. A wide collection of membranes are now being processing in order to compare the values resulting from our method, with those evaluated from other measurements techniques (Hülka and Jiránek, 1996; Cozmuta and Van der Graaf, 1999).

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