A Simple Method for Determining the Diffusion Coefficient of Radon in Concrete Samples Using Charcoal

A. Tsapalov, T. Maslov, K. Kovler
National Building Research Institute – Faculty of Civil and Environmental Engineering, Technion – Israel Institute of Technology, Haifa, Israel

Keywords: radon, exhalation, diffusion coefficient, concrete, activated charcoal.

INTRODUCTION

Radon is one of the most dangerous carcinogens that causes cancer. For this reason, in many countries of the world, the indoor radon is limited. For example, in Israel, the reference level of the radon concentration is 200 Bq/m³ for homes and 500 Bq/ m³ for working places.

The main source of indoor radon is soil, so building materials and structures in the underground part of the building, such as concrete in foundation slab, should prevent the transport of soil radon into the building. However, due to aging and deterioration of concrete in time, its permeability to radon increases. An important characteristic of material permeability is radon diffusion coefficient, which is often used to evaluate quality of radon barrier materials. In addition, radon diffusion coefficient can be used to indicate concrete performance in terms of its durability, similarly to diffusion coefficients of chloride ion, air, oxygen, carbon dioxide and water, which cause detrimental effects in concrete. The advantage of testing diffusion of radon is that it does not interact chemically with concrete constituents, because is a noble gas.

Over the past few decades, many different methods and approaches have been published to determine the diffusion coefficient of radon in building materials. Two international ISO standards are among them, they differ fundamentally in the test modes: one of them [1] is based on the non-stationary mode, and another [2] – on the stationary one. The main advantage of the non-stationary method is the short duration of the test, not exceeding 24 hours, but this method uses a powerful radon source and non-standard measuring equipment, as well as a complex mathematical apparatus [3]. The duration of tests in the stationary mode is significantly longer (from 2 to 4 weeks). At the same time, a radon source of low activity and standard radon devices can be used [2]. The experimental setup in the stationary measurement methods usually includes two sealed chambers, one of them contains a radon source. The test sample is located between these two chambers.

The main novelty of our approach is the use of only one chamber containing a radon source. In addition, we propose to use very simple equipment based on radon adsorption in activated charcoal for measuring its activity, as well as a standard plastic tube for the manufacture of the chamber and concrete samples.

THEORY OF THE METHOD

The measurement scheme of the proposed method is shown in Fig.1a, and its principle is as follows. The test cylindrical sample with a sealed lateral surface and unsealed ends is hermetically mounted in a cylindrical chamber, the lower end of which is hermetically sealed. The source of radon is located inside the chamber.

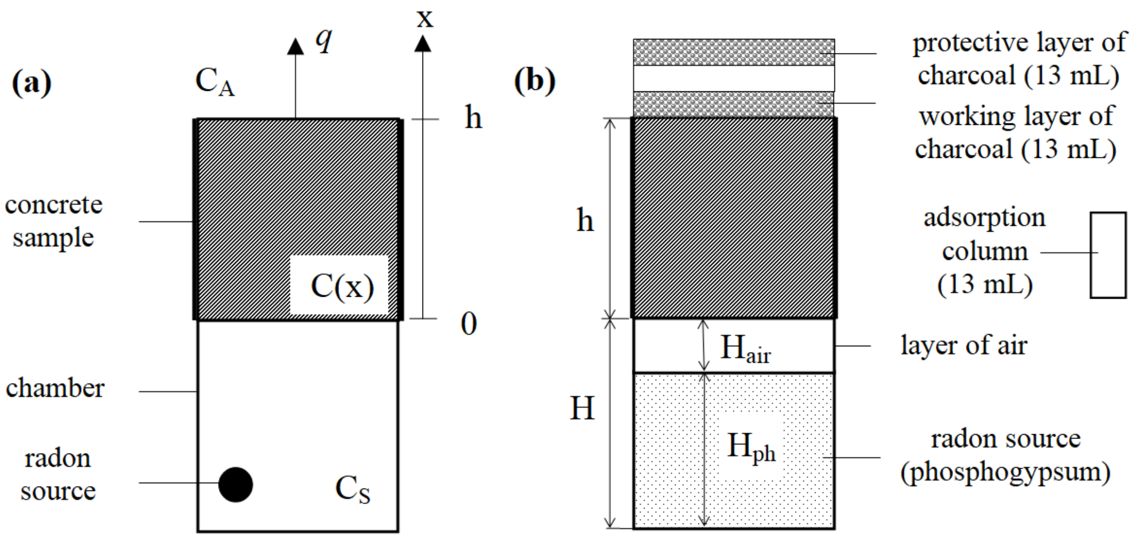

Fig. 1. Measurement scheme (a) and experimental setup (b)

After installing the sample, the concentration of radon in the chamber increases due to the source. However, in the stationary mode, the concentration of radon in the chamber, as well as the distribution of the radon concentrations along the height of the sample, is stabilized over time due to the equality of the rates of radon release from the source and its runoff, which is due to the natural decay and diffusion of radon through the sample into ambient air. The stationary radon diffusion in a homogeneous sample is described by the well-known equation:

$$D \cdot \frac{d^2C(x)}{dx^2} - \lambda \cdot C(x) = 0 \qquad (1)$$

The boundary conditions on the lower and upper surfaces of the sample are as follows:

$$C(x = 0) = C_s \quad (2), \qquad C(x = h) = C_A = 0 \quad (3),$$

where D is the radon diffusion coefficient (m^2/s), $C(x)$ is the distribution of radon concentration in the sample (Bq/m^3), C_s and C_A are the radon concentrations in the chamber and the ambient air, respectively (Bq/m^3), λ is radon decay constant ($2.09 \cdot 10^{-6}$ 1/s), h is the height of the sample (m).

The solution of the boundary task (1) - (3) has the form:

$$C(x) = C_s \cdot sh\left[(h - x) \cdot \sqrt{\lambda/D}\right] / sh(h \cdot \sqrt{\lambda/D}) \qquad (4)$$

According to Fick's first law, the radon exhalation rate from the upper surface of the sample q (Bq/m^2/s) is described by equation:

$$q = -D \cdot \frac{dC(x)}{dx}\Big|_{x=h}, \qquad (5)$$

and the balance of radon in the chamber is described by the following equation:

$$Q - \lambda \cdot C_s \cdot V + S \cdot D \cdot \frac{dC(x)}{dx}\Big|_{x=0} = 0\ , \qquad (6)$$

where Q is the rate of radon release from the source, Bq/s, S is the sample (chamber) area, m^2, V is the free air volume in the chamber, m^3.

Then, considering (4)-(6),

$$q = Q \cdot \sqrt{\lambda \cdot D} / \left[\lambda \cdot V \cdot sh(h \cdot \sqrt{\lambda/D}) + S \cdot \sqrt{\lambda \cdot D} \cdot ch(h \cdot \sqrt{\lambda/D})\right] \qquad (7)$$

Thus, according to (7), the value of the radon diffusion coefficient can be obtained by measurements of the values of two parameters only: q and Q.

EQUIPMENT AND MATERIALS

Dry phosphogypsum [4] with the density about 1,000 kg/m^3 is used as a radon source. For the manufacture of the chamber and the casting of concrete samples, it is advisable to use a standard plastic tube with an external (internal) diameter of 75.0 (71.2) mm, then $S = 4.0 \cdot 10^{-3}$ m^2.

If 1.0 kg of phosphogypsum is used, then chamber height (Hph) should be at least 0.25 m, taking into account S and the density of phosphogypsum. It is advisable to increase the height of the chamber (H) to 0.34 m, so that there is a layer of air ($Hair = 0.09$ m) in order to reduce the uncertainty in estimating the value of V, which also depends on the porosity of the powder (phosphogypsum). If the porosity (p) of phosphogypsum is assumed to be 0.2, then $V \approx S \cdot (Hph \cdot p + Hair) \approx 0.0006$ m^3.

The proposed method for measuring the parameters q and Q, according to Fig. 1b is based on the ability of activated charcoal to adsorb radon. In order to measure q an activated charcoal layer (working layer) is located on the upper surface of the sample, which adsorbs radon released from the sample for a certain time (no more than 20 hours). The protective layer of charcoal prevents the adsorption of radon in the working layer from the surrounding space, which provides $C_A = 0$, according to the boundary condition (3).

Using this method, the value of q is determined by the formula (8) considering the decay of radon in the charcoal layer during the exposure period:

$$q = \lambda \cdot A \cdot exp(\lambda \cdot t_{keep})/S/[1 - \exp(\lambda \cdot t)]\,, \qquad \text{or} \qquad (8)$$

$$q = A \cdot exp(\lambda \cdot t_{keep})/(S \cdot t) \quad \text{if } t < 5\text{ h}\,,$$

where A is the activity of radon in the working layer of charcoal, Bq, t is the exposure period, s, t_{keep} is the time interval between the end of the exposure and the start of measurements, s.

The activity of radon adsorbed in charcoal is measured on a very simple, but at the same time, highly sensitive detector [5]. This detector allows to measure the parameter A at a level of 1.0 Bq (0.5 Bq) with a statistical uncertainty of about 15% (25%).

The value of Q is determined in the same way as q, but without a concrete sample, according to the Fig. 1b, and $Q = q \cdot S$.

RESULTS AND SIMULATION

Based on multiple measurements with an exposure period of 3 to 6 hours, the value of Q was determined to be $(2.2 \pm 0.2) \cdot 10^{-4}$ Bq/s, which is consistent with the data from [4] (the difference is not more than 20%).

An important task is the selection of the optimal height (h) of the concrete sample. The determining criteria are two basic conditions: (i) the ability to more accurately find the difference between the values of D; for example, in the new (with lower D-value) and artificially aged (D is higher) concrete samples, and (ii) the value of q must be at least 15 mBq/(m^{2}s), so that $A > 1.0$ Bq for the exposure period no more than 5 hours (see above). To solve this task, modeling was carried out (Fig. 2) based on the obtained solution (7), taking into account the initial data and criteria presented before.

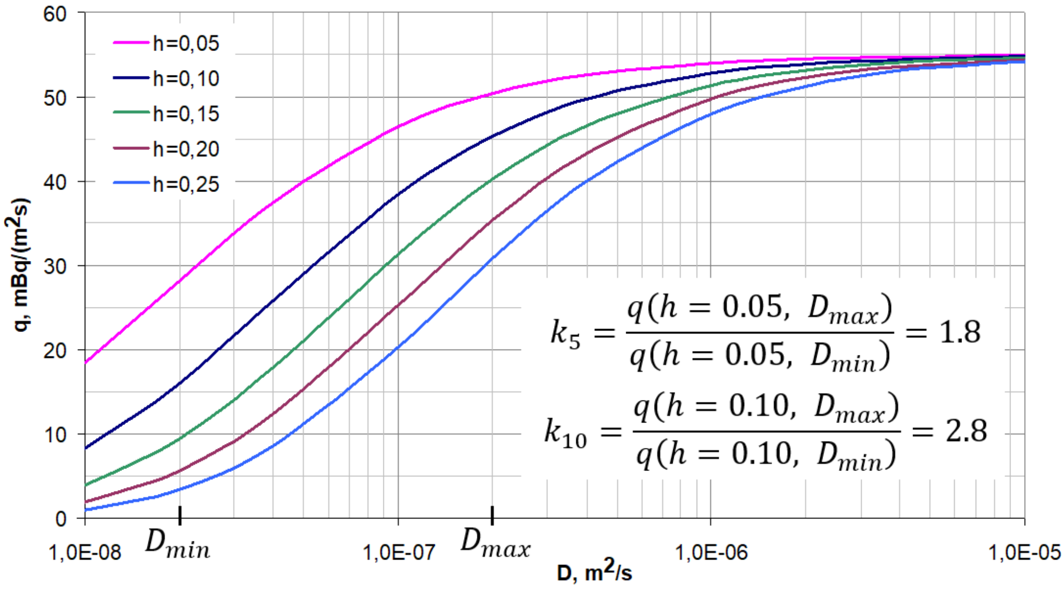

Fig.2. Dependence of the radon exhalation rate from the upper surface of the sample on the radon diffusion coefficient and height of the sample (results of simulation)

The expected range of *D* in new and aged concrete is about from $2{\cdot}10^{-8}$ (D_{min}) to $2{\cdot}10^{-7}$ (D_{max}) m^2/s. Then, considering the condition (ii), the height of sample should not exceed 0.10 m, according to Fig. 2. In other words, considering the condition (i), the sample height of 0.10 m is more preferable than 0.05 m, since a larger value of *h* provides greater sensitivity of the method ($k_{10} > k_5$) when comparing the diffusion rate of radon in the new and aged samples.

CONCLUSIONS

A simple method for determining the radon diffusion coefficient in concrete based on the radon adsorption in activated charcoal is proposed. The geometry of the experimental setup is scientifically justified considering the characteristics of the existing radon source and the proposed method for measuring the activity of radon adsorbed in activated charcoal.

REFERENCES

1. ISO/TS 11665-12:2018 Measurement of radioactivity in the environment — Air: radon 222 — Part 12: Determination of the diffusion coefficient in waterproof materials: membrane one-side activity concentration measurement method.
2. ISO/TS 11665-13:2017 Measurement of radioactivity in the environment — Air: radon 222 — Part 13: Determination of the diffusion coefficient in waterproof materials: membrane two-side activity concentration test method.
3. Tsapalov A., Gulabyants L., Livshits M., Kovler K., New method and installation for rapid determination of radon diffusion coefficient in various materials. *Journal of Environmental Radioactivity*, **130**, 2014, pp. 7-14.
4. Tsapalov A., Kovler K., Control of radon emanation at determination of activity concentration index for building materials. *Construction and Building Materials*, **160**, 2018, pp. 810-817.
5. Tsapalov A., Kovler K., Shpak M., Shafir E., Golumbic Y., Peri A., Ben-Zvi D., Baram-Tsabari A., Maslov T., Schrire O., A radon survey in Israel involving school children by means of the “RadonTest” online system. *NORM IX Symposium Proc.*, Denver, USA, 2019.