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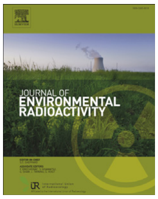

Novel method of measurement of radon exhalation from building materials

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ABSTRACT

In the era of the energy saving policy (i.e. more air tight doors and windows), the radon exhaled from building materials tends to increase its concentration in indoor air, which increases the importance of the measurement of radon exhalation from building materials. This manuscript presents a novel method of the radon exhalation measurement using only a HPGe detector or any other gamma spectrometer. Comparing it with the already used methods of radon exhalation measurements, this method provides the measurement of the emanation coefficient, the radon diffusion length and the radon exhalation rate, all within the same measurement, which additionally defines material's radon protective properties. Furthermore it does not necessitate additional equipment for radon or radon exhalation measurement, which simplifies measurement technique, and thus potentially facilitates introduction of legal obligation for radon exhalation determination in building materials.

1. Introduction

Radon (further on referring to ^{222}Rn) is the greatest source of the public exposure to radiation. More than 50% of the radiation dose received by the general population is a consequence of the exposure to radon i.e. to its progenies. Namely, the radon itself is a noble gas and the most of the inhaled radon in human lungs is also exhaled. However, the radon progenies are metals by its chemical nature and they attach to the surface of the lungs where its destructive damaging potential due to radioactive decay is the most prominent. As a short-lived and also alpha radioactive, ^{214}Po and ^{218}Po, attached to the lungs, give the epithelial layer of the bronchi a substantial radiation dose (BEIR VI, 1999; Ferlay et al., 2007; WHO, 2009). Ergo, the radon is responsible for between 3% and 14% of lung cancer death, being proved to be the second main cause of the lung cancer after smoking (WHO, 2009).

As a primordial radionuclide ^{238}U is present in the Earth's crust and consequently it is present in the building materials, as well. The ^{238}U is a parent radionuclide of the Uranium decay series which includes ^{226}Ra which is direct predecessor of ^{222}Rn. The ^{222}Rn can exhale from a building material and increase the indoor radon concentration. By the contribution to the indoor radon concentration the radon exhaling from the building material is at the second place, immediately after the radon originating from the soil or the bedrock where the building is constructed (Denman et al., 2007; Cosma et al., 2013).

In order to quantify and to regulate the exposure to gamma radiation originating from radionuclides in building materials the Activity Concentration Index I_γ (sometimes referenced as "gamma index") has been proposed (EC, 1999):

$$I_\gamma = \frac{C_{Ra}}{300\left(\mathrm{Bq\ kg^{-1}}\right)} + \frac{C_{Th}}{200\left(\mathrm{Bq\ kg^{-1}}\right)} + \frac{C_K}{3000\left(\mathrm{Bq\ kg^{-1}}\right)},$$

where C_{Ra}, C_{Th} and C_K are the ^{226}Ra, ^{232}Th and ^{40}K specific activities, respectively. The building materials should be restricted in their usage if their gamma index is higher than 1 ($I_\gamma > 1$) which corresponds to an effective annual dose to the inhabitants higher than 1 mSv (EC, 1999).

However, the "alpha index" I_α may be used to regulate the exposure to radon originating from radionuclides in building material (Nordic, 2000):

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$$I_\alpha = \frac{C_{Ra}}{200\left(\mathrm{Bq\ \ kg^{-1}}\right)}$$

The alpha index was proposed on the basis of the assumption that if ^{226}Ra concentration exceeds 200 Bq kg^{-1}, it is possible that the indoor radon concentration will exceed 200 Bq m^{-3} (Nordic, 2000).

There are certain indications that the internal dose received from the radon exhaled from building materials can exceed the external dose received from the radium content in the same building material (Petropoulos et al., 2002; Ujić et al., 2010). Nowadays, this issue becomes more relevant, whereas the energy saving policy (more tight windows and doors) will further decrease the average ventilation rate and increase the radon concentration in indoor air, consequently increasing the dose received from radon (Yarmoshenko et al., 2014). It was shown that the building construction characteristics (like wall materials and period of construction) has significant influence on mean of logarithmic indoor radon concentration (Yarmoshenko et al., 2016) which further implies the necessity to investigate the building materials regarding the radon exhalation issue (exhalation and also the diffusion length).

Due to all the above mentioned reasons the radon exhalation from building materials is still preoccupying interest of many researchers in different countries. Consequently, there are many recent publications dealing with this subject (Topçu et al., 2013; Kumar et al., 2014; Tan and Xiao, 2014; Feng and Lu, 2015; Morelli et al., 2015; Stajic and Nikezic, 2015; Saad et al., 2014), whereby these measurements use exclusively accumulation chamber method.

There are various methods developed for the radon exhalation measurements: the abovementioned accumulation chamber method (sometimes referred as “can” or “sealed can” method), the charcoal method and SSNTD (Solid State Nuclear Track Detector) method (Abu-Jarad et al., 1980; Ingersoll et al., 1983; Iimoto et al., 2008). Some of the methods for radon exhalation measurements are already incorporated into the ISO International Standards i.e. into ISO 11665 Measurement of radioactivity in the environment — Air: Radon-222 as Part 7: Accumulation method for estimating surface exhalation rate (ISO 11665–7:2012) and as Part 9: Test methods for exhalation rate of building materials (ISO 11665–9:2016).

In this manuscript we propose a new method of radon exhalation measurements which does not necessitate any equipment for a radon concentration measurement. Instead, the radon exhalation measurement is performed only by a gamma detector (spectrometer). Moreover, this method provides the values of the radon emanation coefficient, the radon diffusion coefficient and consequently the radon diffusion length, whereby the diffusion coefficient and the diffusion length characterise the building materials regarding their radon protective properties (Tsapalov and Kovler, 2016). Since the gamma spectrometry is the most common tool for radiological analysis of the environmental and other samples, many laboratories would not have to purchase radon equipment in order to estimate radon exhalation. Combined with the fact that only the radionuclide concentration in building materials are regulated, implementation of the proposed method could increase the perspectives to include the radon exhalation in national regulations.

2. Theory and methods

All the mentioned methods of radon exhalation measurement are measuring the exhaled radon directly. Herein described “gamma” method, the non-exhaled radon in the sample of building material is measured by gamma spectrometry, which then allows to estimate the exhaled radon.

A cylindrical sample with sealed lateral side and one base is considered (see Fig. 1). The sample was left for a period of 10 half-lives of ^{222}Rn (~38 days) in order to achieve the equilibrium state. The homogeneity of the sample is assumed. The radon in the building materials exists in two phases: non-emanated (positioned in the mineral phase of the sample) and emanated (positioned in the air phase of the sample) — see Fig. 1. The non-emanated part C_{ne} of the radon has a constant concentration in the sample:

$$C_{ne} = C_{Ra}(1-\varepsilon) \tag{1}$$

where C_{Ra} is the ^{226}Ra concentration in the sample given in Bq kg^{-1}, ε is the emanation coefficient given in non-dimensional unit. C_{ne} is expressed in Bq kg^{-1}. The emanated radon can diffuse through the air in the sample and its concentration C_e depends on the position on the axis of the sample.

$$D\frac{\partial^2 C_e(x)}{\partial x^2} - \lambda C_e(x) + \frac{C_{Ra}\rho\varepsilon}{p} = 0 \tag{2}$$

where D is the radon diffusion coefficient in the given material (m^2 s^{-1}), λ is the decay constant of the radon (s^{-1}), ρ is the density of the material (kg m^{-3}), p is the porosity of the material which defines the percentage of the air in a sample material. As the equilibrium is achieved the radon flux does not change with time,

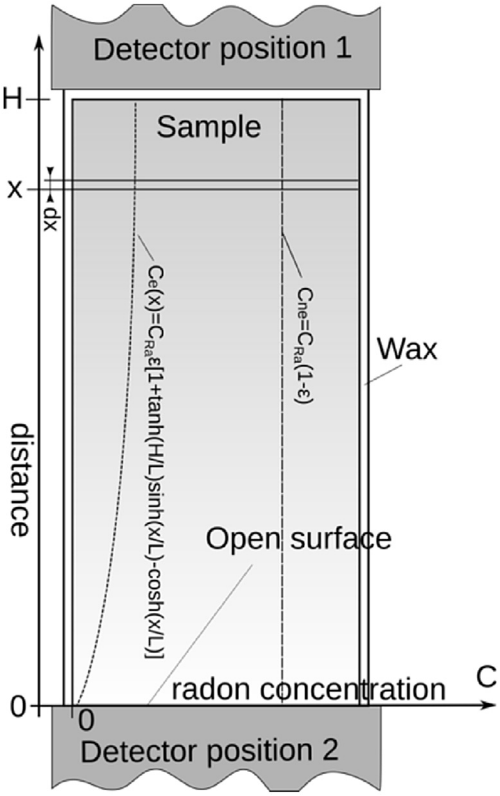

Fig. 1. Scheme of the measurement setup showing the position of the sample and two possible positions of the detector. Radon concentrations (emanated and non-emanated), defined by Eqs. (1) and (4), are also presented. The detector positions are shown as referent to the coordinate system referred to the sample. In reality the detector is at the same position, while the sample is turned upside down between measurements. The wax (beeswax) prevents radon diffusion through sealed sample sides, ensuring the radon exhales only through the open surface.