New gamma method for the radon exhalation measurement from building materials

Ahmed Ali Salim Awhida, Predrag Ujić, Igor Čeliković, Dušan Nikezić, Katarina Karadžić and Boris Lončar

***Abstract*— In the era of the energy saving policy increases the importance of the measurement of radon exhalation from building materials. We present a new method of the radon exhalation measurement using only aHPGe detector or any other gamma spectrometer. 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. 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.**

***Index Terms*— radon exhalation; building materials; radon diffusion coefficient; emanation.**

I. INTRODUCTION

Radon is the greatest source of the public exposure to radiation, whereas more than 50% of the radiation dose received by the general population is due to the exposure to radon. Radon is the second main cause of the lung cancer, responsible for up to 14% of lung cancer death [1]. 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 [2,3].

In order to quantify and to regulate the exposure to gamma radiation originating from radionuclides in building materials the Activity Concentration Index I_γ (often corrected by “alpha index” I_α) has been proposed [4]:

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

where C_{Ra}, C_{Th} and C_K are the ^{226}Ra, ^{232}Th and ^{40}K specific activities, respectively. However, there is a possibility that the dose of inhabitants received from the radon exhaled from building materials can exceed the dose received from the radium content in the same building material [5].

We propose a new method of radon exhalation measurements using a gamma detector, whereby this method provides the values of the emanation coefficient and the diffusion length.

II. THEORY

The method is described on the case of a cylindrical sample with sealed lateral side and one base is considered (see Figure 1). The sample was left for a period of 10 half-lives of ^{222}Rn in order to achieve the equilibrium state. 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.

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

where, D - the radon diffusion coefficient, λ- the decay constant of the radon, ρ- the density, p- porosity of the material. As the equilibrium is achieved, it is $\partial\varphi/\partial t=0$.

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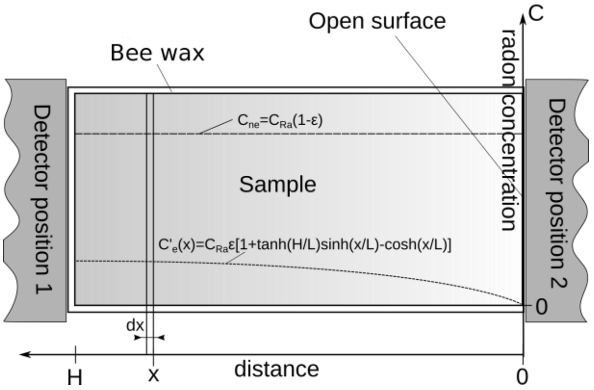

Fig. 1 Scheme of the measurement setup showing the position of the sample and two possible positions of the detector. Radon concentrations (emanated and non-emanated), defined by equations (1) and (3), are also presented. The detector positions are shown as referent to the coordinate system referred to the sample. In reality the detector is at the same position, while the sample is turned upside down between measurements.

The general solution of the equation (2) is the following [6], whereby the solution is normalized by *p/ρ*:

$$\mathrm{C}_{\mathrm{e}}'(\mathrm{x}) = \mathrm{C}_{\mathrm{Ra}}\varepsilon\left[\tanh(H/L)\sinh(x/L) - \cosh(x/L) + 1\right] \quad (3)$$

where L=√D/L is the radon diffusion length, H- the height of the sample.

The total radon concentration is:

$$C_{tot}(x) = C_e'(x) + C_{ne} = \mathrm{C}_{\mathrm{Ra}}\left[1 + \varepsilon\tanh(H/L)\sinh(x/L) - \varepsilon\cosh(x/L)\right] \quad (4)$$

In the case of open sample where radon exhales at one side of the sample, the homogeneity is obviously not fulfilled (see Figure 1). Thus, the estimated radon concentration using a gamma ray *i* (i.e. chosen energy) of certain radon progeny is:

$$C_i = \mathrm{C}_{\mathrm{Ra}}\int_0^H [1 + \varepsilon\tanh(H/L)\sinh(x/L) - \varepsilon\cosh(x/L)]\eta_i(H-x)dx \quad (5)$$

where $\eta_i(H-x)$ is a relative contribution to the counting rate of the layer of the sample of thickness *dx* at distance *(H-x)* from a detector at the position 1 see Figure 1. $\eta_i(x)$is determined by a semi-empirical method using the EFFTRAN package [7]. The EFFTRAN package was used to transfer calibration coefficient of a standard to layers of finite thickness *Δx* at different distances from the detector (see Figure 1) and these values were interpolated with a polynomial of tenth order - $\eta_{\Delta x}^{(i)}$ which is normalised to 1:

$$A\int_0^H \eta_{\Delta x}^{(i)}(x)dx = 1 \Rightarrow A = \frac{1}{\int_0^H \eta_{\Delta x}^{(i)}(x)dx}$$

The final expression of radon concentration using a chosen radon progeny gamma ray *i* is:

$$C_i = \mathrm{C}_{\mathrm{Ra}}\frac{\int_0^H \left[1 + \varepsilon\tanh(H/L)\sinh(x/L) - \varepsilon\cosh(x/L)\right]\eta_{\Delta x}^{(i)}(H-x)dx}{\int_0^H \eta_{\Delta x}^{(i)}(x)dx} \quad (6)$$

or, if the detector is on the position 2:

$$C_i = \mathrm{C}_{\mathrm{Ra}}\frac{\int_0^H \left[1 + \varepsilon\tanh(H/L)\sinh(x/L) - \varepsilon\cosh(x/L)\right]\eta_{\Delta x}^{(i)}(x)dx}{\int_0^H \eta_{\Delta x}^{(i)}(x)dx} \quad (7)$$

There are two unknown variables in the equation (6): the diffusion length – L and the emanation coefficient – *ε*. The ^{226}Ra concentration – C_{Ra} can be measured directly, while for the estimation of the *L* and *ε*, two independent measurements are needed, one with the detector in position 1 (using eq. 6) and second with the detector at the position 2 using eq. 7. This system of two corresponding equations (6) and (7)with two unknown variables *L* and *ε*, has to be resolved numerically (for example, the Mathematica package can be used).

The radon flux can be described by the first Fick's law:

$$J(x) = -D\frac{\partial C(x)}{\partial x} \quad (8)$$

and the radon exhalation *E* is equal to the radon flux at the surface of the sample *E=J(0)*:

$$E = J(0) = C_{Ra}\varepsilon\frac{D}{L}\rho\tanh(H/L) \quad (9)$$

III. RESULTS

A cylindrical sample of 71 mm diameter and120 mm height made from 15 % sand, 45 % of travertine and 40 % of cement was prepared. The travertine from Niška Banja is chosen, as it was known that it has high ^{226}Ra content, which facilitates the testing of the method.

It was estimated that the radon diffusion length was L= 31 ± 3 cm, the emanation coefficient is estimated to be ε=0.45± 0.02. The measured ^{226}Ra concentration in the sample was 175±2 Bq kg^{-1}. The radon exhalation rate was estimated is estimated using the equation (9): 0.0326 ± 0.0014 Bq s^{-1} m^{-2}. The estimated exhalation rate of 0.0326 Bq s^{-1} m^{-2} is very high due to the high radium content in travertine, high emanation coefficient and long radon diffusion length.

This new gamma method is confirmed by measurement with an accumulation chamber of the 0.26 m × 0.275 m × 0.41 m volume and the RAD7 (Durridge Radon Instrumentation). The measured exhalation rate was E = 0.0311 ± 0.0002 Bq s^{-1} m^{-2}, which is in agreement with the proposed gamma method measurement.

The method was compared also to two other exhalation measurement methods. The measurement with accumulation chamber and the Landauer Radtrack solid state nuclear track detector gave the radon exhalation rate of 0.033 ± 0.004 Bq s^{-1} m^{-2}. The exhalation measurement with the charcoal canister gave 0.028 ± 0.004 Bq s^{-1} m^{-2}. The details of these methods intercomparison will be published elsewhere, however the new method showed very good agreement with other methods.

IV. DISCUSSION AND CONCLUSION

This novel method has certain restrictions regarding the ratio H/L. For instance, if the sample height H is much lower than the radon diffusion length L, there will be too small gradient of radon concentration inside the sample and consequently there will be very small difference between counting rates when the sample is measured from the open and from the sealed side. As the emanation factor get smaller (below 5%) it is more important that H should be chosen to be closer to the L. If C_{Ra} is low, the sample should be measured for longer period of time to attain certain precision of the measurement. However, the method still retains its applicability as method to make a selection of building materials regarding the radon exhalation.

A measurement by this method will provide the values of the emanation coefficient, the diffusion coefficient and consequently the diffusion length, which would otherwise require additional equipment and preparation.

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