RADON EXHALATION RATES FROM COMMON BUILDING MATERIALS IN INDIA
Effect of Back Diffusion

by

Amit KUMAR* [*] *and Rishi Pal CHAUHAN

Department of Physics, National Institute of Technology, Kurukshetra

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A radon exhalation study for building materials was carried out by *closed accumulator technique* **using plastic track detector LR-115 type-II, taking into account the effect of back diffusin. The back diffusion of radon into the materials causes an underestimate of free exhalation rates. The results showed that radon exhalation rates of soil, sand, brick powder, and crasher were found to be high as compared to rice husk ash, wall putty, and plaster of Paris. The radon exhalation rates from building materials varied from 0.45 0.07 mBq/kgh to 1.55 0.2 mBq/kgh and 3.4 0.7 mBq/m^2h to 28.6 3.8 mBq/m^2h as measured without considering back diffusion. The radon exhalation rates of building materials oblivious of back diffusion varied from 4.3 0.8 mBq/m^2h to 44.1 5.9 mBq/m^2h. The radon exhalation rates from building materials can be used for estimation of radon wall flux and indoor radon concentration. Thus, it is necessary to make correction in the measured exhalation rates by back diffusion.**

Key words: radon exhalation, back diffusion, accumulator technique

INTRODUCTION

Many radiological measurements were performed to reveal a significant association between the uranium content of soil and high indoor radon concentration [1]. Indoor radon and its decay products contribute 55 % of the total radiation dose from the natural source to the population [2, 3]. This contribution can be much higher in radon prone areas, the natural dose exposure being 5-10 times higher. Many studies have demonstrated the evidence of occurrence of lung cancer caused by radon even at low levels of radon in residential buildings. But the efforts of the various agencies working to reduce the number of lung cancers related to radon exposures are stymied in some countries due to very high radon exposure [4]. The approach of indoor radon studies in connection with radon gas which originates from soil and building materials and is, due to many factors, linked to the underlying geological formations and building structure, shows results of increased levels of radon inside houses [5, 6].

Soil and building materials used for the house construction are the main sources of ^{222}Rn entry into indoor environment [7, 8]. The grains of the bed rock of the Earth crust which contain the ores of uranium and thorium and their decay products in secular equilibrium produce radon isotopes (^{219}Rn, ^{220}Rn, and ^{222}Rn). The ^{222}Rn is of utmost significance for the radiation dose due to very long half life of 3.824 days while the contribution of ^{220}Rn and ^{219}Rn is neglected due to their very short half life. The quantity of radon in the Earth's atmosphere is very small and amounts to about $4\ 10^{-7}$ % by weight [9]. The radon emitted in the decay series of uranium and thorium causes further decay to produce the daughter elements like polonium, lead and bismuth, which are more hazardous than radon itself because these elements are in a solid state, while radon is a gas. Some of these decay products gets attached to aerosol particles to form nanoparticles of 0.5-200 nm in size, while some remain unattached and float in the air. If these nanoparticles are inhaled by humans through the nose and mouth, they get deposited in the respiratory tract by impaction and diffusion, and give alpha doses to the lung, causing damage of cells and tissues and hence, ultimately leading to the initiation of cancer. Radon is the second most leading cause of cancer, after smoking, and is considered responsible for 3 to 14 % of lung cancer deaths [3].

The radon exhalation rates of building materials thus depend upon the radioactive content, radon emanation rates and radon diffusion length in the materials

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[*] Corresponding author; e-mail: amit.vera@gmail.com

matrix. Both, active and passive techniques can be used to find radon exhalation rates. Accumulator technique has limitations in the context of the accuracy of derived radon flux because of back diffusion and interference of thoron in radon measurement. When a material is sealed in an accumulator, radon concentration first increases linearly with time and attains a saturation value. The radon flux from the sample is directly proportional to the concentration gradient. As the radon concentration in the air above the sample goes on increasing, it causes a decrease in radon flux. This decrease in the radon flux can be quantified in term of back diffusion coefficient (α), which relates the two type of radon exhalation rates namely free and bound radon exhalation rates. Thus, the presence of radon, back diffusion causes an underestimate of the free exhalation rates.

Numerous data have been published in literature without considering the back diffusion effect of radon from building materials and soil. The corrective measure of radon exhalation rates of soil was introduced by Rehman *et al*. [10, 11] and reported that the back diffusion of radon causes an underestimate of free exhalation rate of soil. Thus it is necessary to correct the radon exhalation rates data by considering the back diffusion rates so that the proper selection of the building materials may be made for construction purposes.

MATERIALS AND METHODS

The measurement of radon exhalation rates of building materials was carried out by "Sealed Accumulator technique" the exhalation rate depends upon the material and its amount, as well as on the geometry and dimensions of the accumulator [12-15]. The building materials under study were dried and sieved through a 100-mesh sieve and 100 g was placed in the accumulators (diameter 7.0 cm and height 13.5 cm). LR-115 type II plastic track detector (3 cm ×3 cm) was fixed at the top inside of the accumulator so that the sensitive lower surface of the detector may be freely exposed to radon [16, 17]. The height of the samples (1.2-1.5 cm) inside the accumulator was selected so that the tracks formed on LR-115 detector may be due to radon only. While due to the short half life of thoron and the range of alpha particles, thoron and its decay products do not form tracks on SSNTD (LR-115). Radon and its daughters reach an equilibrium concentration after 4 hours and thus, the equilibrium activity of the emergent radon could be obtained from the geometry of the accumulator and the time of exposure. After the exposure for 100 days, the detectors were etched in 2.5 N NaOH at 60 °C for a period of 90 minutes in a constant temperature water bath. The tracks densities from the etched track detectors were then determined using spark counter. The etched track detector first pre-sparked at 900 V twice and then tracks were counted at 500 V thrice and then mean values were used for calculation. From the track density, the radon activity was obtained, using the calibration factor of 0.056 tr. $\text{cm}^{-2}\text{d}^{-1}/\text{Bqm}^{-3}$ obtained from an earlier calibration experiment [15]. The radon exhalation rates of building materials with back diffusion were calculated using the relations [12-15]

$$E_{\rm A} \quad \frac{CV\lambda}{A\ \ t\ \ \ \frac{1}{\lambda}(\mathrm{e}^{\ \ \lambda t}\ \ \ 1)} \tag{1}$$

$$E_{\rm M} \quad \frac{CV\lambda}{M\ \ t\ \ \ \frac{1}{\lambda}(\mathrm{e}^{\ \ \lambda t}\ \ \ 1)} \tag{2}$$

where C is the equilibrium radon activity inside the accumulator, V and A are the volume and the area of cross-section of the accumulator, M is the mass of the sample, and λ – the radon decay constant and, t – the time of exposure.

In the past, many investigators used the sealed accumulator technique employing a solid-state nuclear track detector (SSNTD) for measurements of radon exhalation rates from various construction materials [16-19], since this method is being inexpensive and giving obtains reliable measurements [18]. But this technique did not take into account the impact of back diffusion. After the accumulator is sealed, the radon activity inside the accumulator increases with time and eventually, reaches a saturation value. Thus the radon concentration difference in the pore space and the air decrease with time, causing a decrease in the radon flux from sample surface. This effect was incorporated in term of a quantity called back diffusion. This causes a decrease in the radon flux from the sample inside the accumulator causing an error in estimation of radon exhalation rates. The radon exhalation inside the accumulator, for a short exposure time is assumed to be free from back diffusion and is called free exhalation rate, while the exhalation rate for a long time is bound by back diffusion and is called bound exhalation rate. The parameters which affect the radon exhalation are porosity and height of the sample inside the accumulator. The correction for radon exhalation rates due to back diffusion was considered by Rehman *et al*. [10, 11]. The free F_0 and bound F exhalation rates are given by

$$F_0(t) \quad \frac{C(t)(\varepsilon\lambda Y_0 A \quad \lambda V)}{A(1 \quad \mathrm{e}^{\ [\lambda\ \ (\varepsilon\lambda Y_0 A)/V]t})} \tag{3}$$

$$F(t) \quad \frac{C(t)(\varepsilon\lambda Y_0 A \quad \lambda V)}{A(1 \quad \mathrm{e}^{\ [\lambda\ \ (\varepsilon\lambda Y_0 A)/V]t})} \quad \varepsilon\lambda Y_0 C \tag{4}$$

where ε is the porosity of the sample measured by water absorption method, λ – the radon decay constant and Y_0 – the height of the sample in the accumulator. A is the area of cross-section of the accumulator. In eq. (4) the term on the left hand side represents the total

Table 1. Radon mass and surface exhalation rates of building construction materials

No.	Sample	Porosity	Mass exhalation rate E_m [mBqkg^{-1} h^{-1}]	Surface exhalation rate E_A [mBqm^{-2} h^{-1}]
1	Cement	0.56	0.8 ± 0.14	13.8 ± 2.4
2	Soil	0.42	1.48 0.12	26.5 0.2
3	Sand	0.41	1.1 0.02	20.2 0.6
4	Rice husk ash	0.35	0.45 0.07	3.4 0.7
5	Plaster of Paris	0.57	0.7 0.30	10.2 3.9
6	Wall putty	0.65	0.45 0.07	8.7 1.1
7	Brick powder	0.63	1.55 0.2	28.6 3.8
8	10 mm aggregate	0.55	0.85 0.07	15.2 0.9
9	Crasher	0.45	1.35 0.07	24.8 1.2

Table 2. Free and bound radon exhalation rates of building materials samples

No.	Sample	Free exhalation rates (F_0) [mBqm^{-2}h^{-1}]	Bound exhalation rates (F) [mBqm^{-2}h^{-1}]
1	Cement	20.4 3.7	13.1 2.3
2	Soil	35.6 0.2	25.1 0.2
3	Sand	26.9 0.8	19.1 0.6
4	Rice husk ash	4.3 0.8	3.3 0.6
5	Plaster of Paris	15.2 5.9	9.6 3.7
6	Wall putty	13.7 1.7	8.3 0.9
7	Brick powder	44.1 5.9	27.1 3.6
8	10 mm aggregate	22.2 1.4	14.3 0.8
9	Crasher	34 1.6	23.4 1.1

exhalation rates of radon from building materials with the back diffusion effect. The first term on the right hand side represents the exhalation rates free from back diffusion while the second term represents the correction for back diffusion.

RESULTS AND DISCUSSION

Using the relations (1) and (2) from the previous chapter, the radon exhalation rates of some common building construction materials were calculated from the measured track densities and are shown in tab. 1. The radon exhalation rates of soil, sand, brick powder and crasher were found to be high as compared to rice husk ash, wall putty and plaster of Paris. The radon mass exhalation rates of building materials varied from 0.45 0.07 mBq/kgh to 1.55 0.2 mBq/kgh, whereas surface exhalation rates varied from 3.4 0.7 mBq/m^{2}h to 28.6 3.8 mBq/m^{2}h. The free and bound exhalation rates, and effective dose equivalent from these building materials using eqs. 3 and 4 are listed in tab. 2.

The free radon surface exhalation rate of building materials varied from 4.3 0.8 mBq/m^{2}h to 44.1 5.9 mBq/m^{2}h with an average of 24 mBq/m^{2}h and bound exhalation rates varied from 3.3 0.6 mBq/m^{2}h to 27.1 3.6 mBq/m^{2}h with an average of 15.9 mBq/m^{2}h. The comparison of exhalation rates measured by relations 2, 3, and 4 is shown in fig. 1

The free radon surface exhalation rates measured by the relation (3) are higher than those of the bound exhalation using the relation (2) and the relation

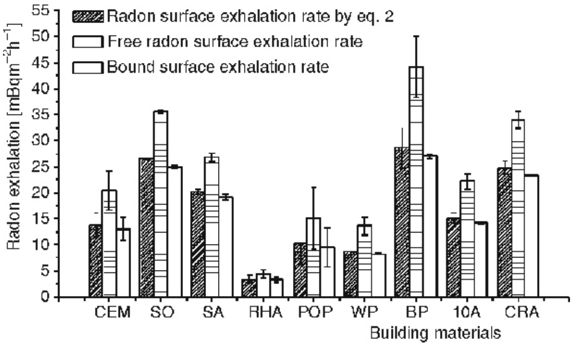

Figure 1. Comparison of radon surface exhalation rates using various relations

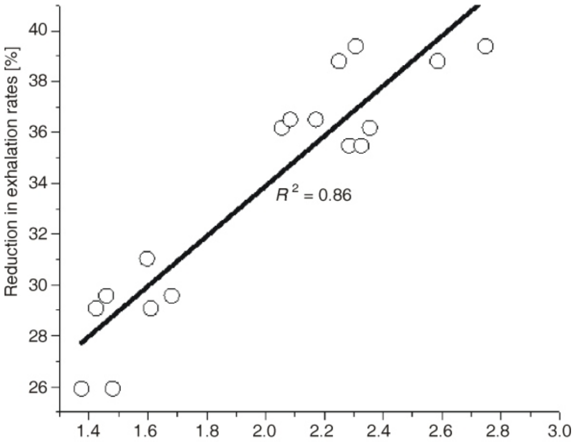

Figure 2. Percentage reduction of exhalation rates with back diffusion coefficient

(4) from all the building materials under study. The radon exhalation rates of building materials are affected by back diffusion which depends upon the porosity of the sample. The percent of reduction in the exhalation rates of the samples with the increase of back diffusion coefficient shows a good agreement (R^2 = 0.86) as shown in fig. 2.

CONCLUSIONS

The radon exhalation rates of soil, sand, brick powder, and crasher were found to be high as compared to rice husk ash, wall putty and plaster of Paris. The radon back diffusion rates from building materials increased with porosity of the sample and showed a good correlation with the percentage of reduction in the radon exhalation rates. The radon exhalation, radon activity and effective dose equivalent of the room, measured by considering the effect of back diffusion, was in good agreement with the conventional relation. While the free exhalation rate, radon activity and effective dose equivalent are 30 % to 40 % higher than bound exhalation rates, depending upon the porosity of a sample. The exhalation rates of building materials under study were found below the world wide average reported in UNSCEAR [20]. The radon exhalation rate along with the radium, thorium, and potassium contents of building materials, are important parameters used for selection of the building materials. The radon exhalation rates from building materials can be used for estimation of radon wall flux and indoor radon concentration. Thus, it is necessary to correct the measured exhalation rates due to back diffusion.

AUTHORS' CONTRIBUTIONS

The idea for the study was put forward by R. P. Chauhan and the measurement and calculations by A. Kumar. Both authors interpreted the data and prepared the manuscript.

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Амит КУМАР, Риши Пал ЧАУХАН

ЈАЧИНА ОСЛОБАЂАЊА РАДОНА ИЗ УОБИЧАЈЕНИХ ГРАЂЕВИНСКИХ МАТЕРИЈАЛА У ИНДИЈИ

Ефекат повратне дифузије

Употребом пластичних траг детектора LR-115 Тип-II и урачунавањем повратне дифузије, спроведено је испитивање ослобађања радона из грађевинског материјала „методом затворене акумулације”. Повратна дифузија радона доводи до потцењивања слободне јачине ексхалације. Резултати показују да су јачине ослобађања радона из земљишта, песка, праха, опеке и дробљеног материјала, више од пепела, зидног кита и париског гипса. Не узимајући у разматрање повратну дифузију, јачине ослобађања радона из грађевинског материјала су у опсегу од 0.45 0.07 mBq/kgh до 1.55 0.2 mBq/kgh и од 3.4 0.7 $\mathrm{mBq/m^2h}$ до 28.6 3.8 $\mathrm{mBq/m^2h}$. За грађевинске материјале без повратне дифузије, јачина ослобађања радона износи од 4.3 0.8 $\mathrm{mBq/m^2h}$ до 44.1 5.9 $\mathrm{mBq/m^2h}$. Јачине ослобађања радона из грађевинских материјала могу се користити за процену флукса радона из зида и концентрацију радона у затвореном простору. Стога је потребно извршити корекције у мерењима јачине ослобађања радона на повратну дифузију.

Кључне речи: ослобађање радона, повратна дифузија, техника акумулације