American Journal of Applied Sciences 9 (10): 1653-1659, 2012
ISSN 1546-9239

An Overview on Studying ^{222}Rn Exhalation Rates using Passive Technique Solid-State Nuclear Track Detectors

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Abstract: Problem statement: Uranium is a radiotoxic element found in trace quantities in alomost all natural accurring materials like soil, rock. Radon an inert radioactive gas whose predecessor in uranium, is emitted from soil beneath the house and from building materials. Accurate knowledge of exhalation rate plays an important role in characterization of the radon source strength in some building materials and soil. It is a useful quantity to compare the relative importance of different sample of building materials and soil. **Approach:** This study provides an overview of measurements of radon exhalation rates for selected samples in Egypt were carried out using passive measuring techniques were measured by Can Technique using LR-115 type II plastic track detectors. **Results:** The radon concentration varies from 2.44-29 k Bq m^{-3} and the corresponding values of surface exhalation rates from 4.16-26.24 Bq m^{-2}. h the radium content 226Ra results in all samples under test in increasing order of magnitude. From the results it can be noticed that The lowest value of 226Ra is 7 Bq kg^{-1} in Sand sample, while the highest value is 85 Bq kg^{-1} Ordinary Cement. **Conclusion:** All the values of radium content in all samples under test were found to be quite lower than the permissible value of 370 Bq kg^{-1} recommended by Organization for Economic Cooperation and Development.

Key words: Solid-State Nuclear Track Detector (SSND), building materials, exhalation rates, radium content, radon exhalation, cooperation and development, organization for economic

INTRODUCTION

Exhalation of ^{222}Rn, α-radioactive inert gas, is associated with the presence of ^{226}Ra and its ultimate precursor uranium in the earth crust. Although these elements occur in virtually all types of rocks and soils, their concentration varies with specific sites and geological materials. The half-life (τ1/2) of ^{222}Rn is 3.82 days. Being a noble gas ^{222}Rn can move large distances through rocks and soils. Radon can diffuse through rocks and soil, can move from one place to the other and can leak out in the atmosphere from the soil. So the distribution of radon in soils has been related to geological controls in terms of its generation and migration; uranium content in bed rocks and soils influence production and the soil characteristics (including the soil moisture and permeability) control the transportation of radon (Bajawa *et al*., 2009).

Henshaw *et al*. (1990), has claimed that indoor radon exposure is associated with the risk of leukaemia and certain other cancers, such as malenoma and cancers of the kidney and prostate. If Uranium rich material lies close to the surface of the earth there can be high radon exposure hazards (Fahmia *et al*., 2008; Abdelzaher, 2011; El-Zaher, 2011; Archer *et al*., 1973; Sevc *et al*., 1976; UNSCEAR, 1993). The rate at which radon escapes or emanates from solid into the surrounding air is known as radon exhalation rate of the solid. This may be measured by either per unit mass or per unit surface area of the solid.

Among the different techniques available for radon measurements, the method based on the use of solid state nuclear tracks detectors (SSNTD) is probably the most widely applied for long term radon measurements. The solid state nuclear track detector have been used for determining the radon emanation in a limestone cave (Oufni and Misdaq, 2001) also for determination radon activity and uranium content in different geological samples (Oufni, 2003; Oufni *et al*., 2005; Amin and Eissa, 2007). Since uranium and radium present in the soil, rocks and building materials are the main sources of indoor radon. Solid state nuclear track detectors have become an important tool in every investigation of the presence of radon gas. In this work, we describe a method based on using LR-115 type II for the study of radon exhalation rate and radium content on selected building materials and soil samples used locally in Egypt.

Theoretical approach: The passive measuring techniques ''Can Technique'' employing a Solid-State Nuclear Track Detector (SSNTD), a simple and efficient

method to assess radon exhalation rates (Abu-Jarad *et al*., 1980; Samuelsson and Pettersson, 1984; Ramola and Choubey, 2004; Prasad *et al*., 2008), besides being relatively inexpensive, the technique provides quite reliable measurements.

Theory Consider a sealed cylindrical can fitted with a source of radon and a SSNTD dosimeter fixed at the top of the can as shown in Fig. 1. Assume that radon, thoron and their daughters are in radioactive equilibrium in the air volume of the can. For diffusion in air, it is expected that all daughters of interest will be deposited except ^{216}Po will be in air volume. Furthermore, ^{220}Rn and ^{216}Po are inhomogenously distributed in the air due to their short half lives. The ^{212}Po and ^{212}Bi formed by the decay of ^{216}Po will be preferentially inhomogenously deposited on the wall of the can. It is clear that the track density registered on the detector which is related to ^{222}Rn, as well as its plated-out daughters. The exhalation of radon from the sample surface represents the source of the number of radon atoms & N(t)' present in the air between the sample and SSNTD. The natural decay of radon provides the only removal mechanism. The rate of change of N(t) with time is therefore governed by the following differential Eq. 1:

$$\frac{dN(t)}{dt}=EA-\lambda N(t) \quad (1)$$

where N(t) is the total number of radon atoms present in the can at time t, E is the exhalation rate (Bq m^{-2} h^{-1}), A is cross-sectional area of the can (the surface area of sample from which the exhalation takes place) and λ is the decay constant of radon (h^{-1}). The solution of Eq. 1 with initial condition N(0)=0 is Eq. 2:

$$N(t)=\frac{EA}{\lambda}[1-e^{-\lambda t}] \quad (2)$$

If V is the volume of air (m^3), The activity concentration of radon C(t). in the air volume of the can as a function of time t can be given by the following relation Eq. 3 (Morawska, 1989; Chen *et al*., 1993):

$$C(t)=\frac{EA}{\lambda V}[1-e^{-\lambda t}] \quad (3)$$

Since the solid state nuclear track detector measures the total number of alpha-disintegration in unit volume of the cylindrical can during the exposure time t. Therefore, the measured track density rate recorded on the SSNTD is proportional to the radon concentration and is given by Eq. 4:

$$\frac{d\rho(t)}{dt}=KC(t) \quad (4)$$

where ρ(t) is the track density and K is the calibration coefficient (α-tracks cm^{-2}day^{-1}/Bq m^{-3}). Using Eq. 3 in (4), we derive Eq. 5:

$$\rho=K\int_0^T C(t)dt=\frac{KAE}{\lambda V}\int_0^T (1-e^{-\lambda t})dt \quad (5)$$

with initial condition ρ(0)=0, the solution is given by Eq. 6:

$$\rho=K\frac{AE}{\lambda V}[t-\frac{1}{\lambda}(1-e^{-\lambda t})] \quad (6)$$

The radon exhalation rate in terms of area is calculated from the Eq. 7:

$$E_A=\frac{CV\lambda}{AT_{eff}} \quad (7)$$

where, T_{eff} is the effective exposure time which is related with the actual exposure time t and decay constant λ for ^{222}Rn with the relation Eq. 8:

$$T_{eff}=t-\frac{1}{\lambda}(1-e^{-\lambda t}) \quad (8)$$

And E_A is the radon exhalation rate expressed in Bq m^{-2}h^{-1}, C represents the integrated radon exposure (Bq.m^{-3}.h), V is the effective volume of the can, t is the exposure time in hours (h), λ is the decay constant for radon (h^{-1}) and *A* is cross-sectional area of the can (m^2). The radon exhalation rate in terms of mass is calculated from the expression Eq. 9:

$$E_M=\frac{CV\lambda}{MT_{eff}} \quad (9)$$

Were E_M is the radon exhalation rate in terms of mass (Bq kg^{-1}hr^{-1}) and M is the mass of sample All the quantities on the right-hand side of Eq. (7,9) are known except C(t). We have experimentally determined the value of C(t) using LR-115 type II based in radon dosimeter. Putting the value of C(t) in Eq. (7,9), exhalation rate was determined.

Problems encountered in exhalation rate:

Back-diffusion: In a closed chamber which contains a sample, ^{222}Rn concentration increases with the passage of

time from zero to its maximum value. However, after reaching its maximum value, back diffusion of radon also take place which reduces the 222Rn concentration

It is very important to keep the activity concentration in the chamber air low, compared to the activity concentration in the pore air of the sample. Almost immediately after enclosing a sample in a container or attaching an accumulator to an exhaling surface, the radon released from the exhaling material has a significant probability of diffusing back to the sample. Closed-chamber methods of small chamber volume compared to the pore volume of the sample are highly susceptible to such phenomena. The theoretical and experimental work reported by Samuelsson and Petterson (1984); Samuelsson and Erlandsson (1988) and Samuelsson (1990). Suggests that choosing a chamber free volume 10 times larger than the pore volume of the sample, may acceptably minimize the back-diffusion effect.

In this study the radon exhalation rate may be effected by the back diffusion process, because the ratio of the cup volume to the sample volume is about three. Therefore, the process of the back diffusion has to be taken into consideration. Back diffusion parameter (β) can be defined by Hafez *et al*. (2001) Eq. 10:

$$\beta=\frac{\lambda PV_s}{V}P=1-\frac{V_d}{V_w} \quad (10)$$

where, V is the effective volume inside the container,Vs is the volume of soil sample, p is the porosity of soil porous material, V_w and V_d are the volumes of wet and dry sample, respectively.

The exhalation rate of radon in samples (E_A and E_M) and the effective exposure time (T_{eff}) was corrected with Back diffusion parameter (β) as follows Eq. 11-13:

$$E_A=\frac{CV(\lambda+\beta)}{AT_{eff}} \quad (11)$$

$$E_M=\frac{CV(\lambda+\beta)}{MT_{eff}} \quad (12)$$

$$T_{eff}=t-\frac{1}{(\lambda+\beta)}(1-e^{-(\lambda+\beta)t}) \quad (13)$$

Radium content calculation: Since the half-life of ^{226}Ra is 1620 years and that of ^{222}Rn is 3.82 days, it is reasonable to assume that an effective equilibrium (about 98%) for radium-radon members of the decay series is reached in about three weeks. Once the radioactive equilibrium is established, one may use the radon alpha analysis for the determination of steady state activity concentration of radium. The activity concentration of radon begins to increase with time T, after the closing of the can, according to the relation Eq. 14:

$$C_{Rn}=C_{Ra}(1-e^{-\lambda t}) \quad (14)$$

where, C_{Ra} is the effective radium content of the sample. Since a plastic track detector measures the time-integrated value of the above expression i.e., the total number of alpha disintegrations in unit volume of the can with a sensitivity K during the exposure time T, hence the track density observed is given by Eq. 15:

$$\rho=K\,C_{Ra}\,T_{eff} \quad (15)$$

where T_{eff} denotes, by definition, the effective exposure time, Referring to Fig. 1 it is clear that the "effective radium content" of the solid sample can be calculated using the formula Eq. 16:

$$C_{Radium}=\frac{\rho hA}{k_{Rn}T_eM} \quad (16)$$

where, M is the mass of the soil sample in kg, A is the area of cross-section of the can in m^2; h is the distance between the detector and top of the solid sample in meter.

MATERIALS AND METHODS

Passive integrating methods of radon measurement by Solid-State Nuclear Track Detectors (SSNTDs): A passive method (can-technique) using LR-115 type II (Kodak-Path6, France) plastic detector, as a solid state nuclear-track detector was developed for measurements of radon exhalation rate of different samples materials, in which the samples of interest is enclosed in a sealed can (Abu-Jarad *et al*., 1980; Somogyi *et al*., 1986; Samuelsson and Pettersson, 1984; Ramola and Choubey, 2004). The cellulose nitrate LR-115 (12 µm thickness), is a very useful detector for the direct registration of alpha particles, the sensitive surface of the detector faced to the samples, the experimental arrangement is shown in Fig. 1. The tracks detected by these plastic detectors are not directly visible and have to be enlarged by adequate chemical processing. Different samples of soil, sand and some building materials were collected from several quarries and commercial companies in

Egypt. All samples were dried in a temperature controlled furnace (oven) at a temperature 100±0.1°C for 24 h to ensure that moisture is completely removed. All samples were crushed to a fine powder form, the crushed samples were then sieved through a small mesh size to remove the larger grains size and render them more homogenous. About 200 g of sample was placed in a plastic Can of size 10 cm in height and 7.0 cm in diameter. A piece of detector of size 2×2 cm was fixed on the top of inner surface of the can, in such a way, that it is sensitive surface always facing the soil sample. The Can is sealed air tight with adhesive tape and kept for exposure of about 90 days. During exposure period, the sensitive side of the detector always faced the sample and is exposed freely to the emergent radon from the sample in the can so that it could record alpha particles resulting from the decay of radon in the remaining volume of the can. Radon and its daughters reach equilibrium in about 4 weeks (Sonkawade *et al*., 2008) and hence the equilibrium activity of emergent radon could be obtained from the geometry of the can and time of the exposure.

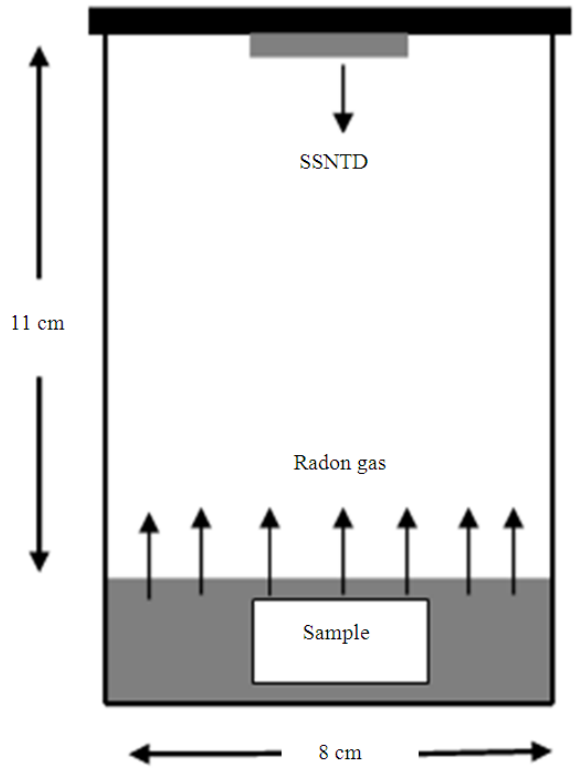

Fig. 1: Experimental set-up for the measurement of radon exhalation

Following exposure, the the LR-115 films (SSNTDs) are chemically etched in 2.5 N NaOH solution at 60°C for 1.5 h. After this chemical treatment, these SSNTDs were washed, dried and scanned using an Olympus optical microscope with a magnification of 300X. The laboratory's Image Analysis System includes a microscope and a camera and counts the number of alpha tracks on 0.5 cm^2 of the detector. On receipt of a new batch of detectors, each sheet is sampled for quality assurance testing: 10% of the unexposed detectors are etched. The subsequent counting determines the background track density (tracks per square centimeter or tr cm^{-2}). If the background track density of all detectors from a sheet is less than 50 tr cm^{-2}, the sheet is released for use. The radon track density ρ_{Rn} (in tr.cm^{-2}) is related to the radon activity concentration C_{Rn} (in Bq.m^{-3}) and the exposure time T by formula Eq. 17 (Somogyi, 1986):

$$C_{Rn}\ (Bq.m^{-3}) = \rho_{Rn} / K_{Rn}.\ T \qquad (17)$$

where, K_{Rn} is the calibration factor of LR-115 plastic track detector (0.033 track.cm^{-2}.d^{-1}/Bq.m^{-3}) with an uncertainty of a bout ±10%, which is calibrated in pervious study (Abd-Elzaher, 1997). The value of K_{Rn} will depend on the height and radius of the measuring can (Somogyi, 1990).

RESULTS

The results of mass and surface exhalation rates of radon, the soil-gas radon concentration and the corresponding radium content in different samples are presented in Table 1. The average value of the back diffusion parameter β was found to be 0:16λ which is, in our opinion, not significant in these measurements.

The radon exhalation rate in terms of area is calculated from the Eq. 7:

$$E_A = \frac{CV\lambda}{AT_{eff}}$$

where, E_A is the radon exhalation rate expressed in Bq $m^{-2}h^{-1}$, C represents the integrated radon exposure (Bq.m^{-3}.h), V is the effective volume of the can in (5.526×10^{-4} m^3),T_{eff} is the exposure time in hours (2028 h), λ is the decay constant for radon (h^{-1}) and A is the area of the bottle (5.026×10^{-3} m^2). The radon exhalation rate in terms of mass is calculated by using Eq. 9:

$$E_M = \frac{CV\lambda}{MT_{eff}}$$

Table 1: Results of radon concentration, radon exhalation rate and radium contents in some sample of building material

Sample name	Sample code	Radon (kBq/m^3)	Concentration radium ontent (Bq/kg)	Exhalation rates E_M (Bq/m^2.h)	Exhalation rates E_A (Bq/m^2.h)
Ordinary cement 1	OC1	10.10	85	0.49	8.95
Ordinary cement 2	OC2	29.63	71	0.52	26.24
Ordinary cement 3	OCc3	24.75	76	0.36	21.92
Gypsum	GY	26.46	52	0.14	23.43
Limestone	LI	18.12	20	0.06	16.05
Clay Soil 1	CL1	6.97	9	0.10	6.17
Clay Soil 2	CL2	3.13	15	0.05	2.78
Clay Soil 3	Cl3	5.22	8	0.04	4.63
Sand 1	SA1	2.78	7	0.09	2.47
Sand 2	SA2	2.44	14	0.13	2.16
Sand 3	SA3	4.88	19		4.32
		6.62			5.86

Here E_M is the radon exhalation rate in terms of mass (Bq kg^{-1}h^{-1}) and M is the mass of soil sample (200 g).

The radium concentration in soil samples was calculated using the relation Eq. 18:

$$C_{Radium} = \frac{\rho h A}{k_{Rn} T_e M} \quad (18)$$

where, C_{Radium} is the effective radium content of soil sample (Bq kg^{-1}), M is the mass of soil sample (200 g), A is the area of cross-section of bottle (5.024×10^{-3} m^2), h is the distance between the detector and the top of the soil sample (0.1m), K is the calibration factor of LR-115 plastic track detector, and T_e is the effective exposure time.

DISCUSSION

The radon exhalation rate was measured for some different samples of building materials as reported in Table 1. These building materials are usually used in construction in some regions of Egypt. The values of exhalation rates of building materials vary from one sample to another. This variation is due to the content of uranium and radium and to the porosity of the building materials. The radon concentration varies from 2.44 to 29 kBq m $^{-3}$ and the corresponding values of surface exhalation rates from 4.16 to 26.24 Bq/m^2.h. The ordinary cement 1, demonstrates the highest mean exhalation rate (26.24 Bq/m^2.h) while sand 1 and Clay Soil 3samples with values of 2.44 Bq/m^2.h and 2.788 Bq/m^2.h respectively have the lowest values within the same order of magnitude. Radon concentration in dwellings depends partly on the type of building material. It appears clearly from the obtained values of radon concentration of ordinary cement1,2 and 3 and Gypsum, that they would present a higher indoor radon concentration if were used together for construction.

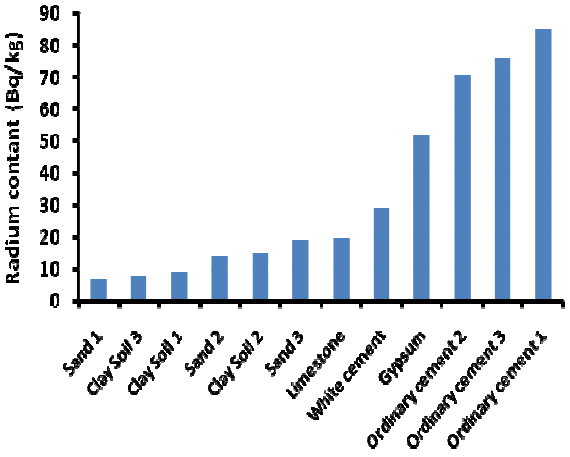

Fig. 2: Radium content (Bq/kg) for some used sample of building materials in increasing order of magnitude.

The radon exhalation rates shown in Table 1 were found to be consistent with data obtained by other investigators (Folkerts *et al*., 1984; Chen *et al*., 1993).

Figure 2 represent the values of radium content ^{226}Ra in all samples under test in increasing order of magnitude.

From the results it can be observed that the lowest value of ^{226}Ra is 7 Bq /kg calculated in Sand sample1, while the highest value is 85 Bq /kg calculated in Ordinary Cement 1. The high ^{226}Ra values can be rendered to the high concentration of the three radionuclides ^{226}Ra and ^{232}Th. The results shows that there are considerable variations in the ^{226}Ra of the different materials and also within the same type of material originating from different areas. This result is important from the point of view of selecting suitable materials for use in building and construction especially concerning those which have large variations in their activities.

Table 2: Shows values of radium contant (Bq kg^{-1}) for some building materials in different countries

Sample	Egypt(present work)	[1]Germany	[2]Australia	[3]Algeria	Zambia	[5]India
Gray cement	77.33	70	115.0	112	79.00	108.50
White cement	29.00	-	-	-	-	-
Sand	13.33	59	70.0	28	84.15	135.00
Gypsum	52.00	-	11.1	-	-	10.36

Krieger (1981); Beretka and Mathew (1985); Amrani and Tahtat (2001); Hayumba *et al*. (1995); Kumar *et al*. (1999)

The variation in radium equivalent activities may suggest that it is advisable to monitor the radioactivity levels of materials from a new source before adopting it for use as a building material (Kumar *et al*., 1999). The recommended maximum levels of radium equivalents for building materials to be used for homes is 370 Bq kg^{-1} and for industries is 370-740 Bq kg^{-1} (Oresegun and Babalola, 1988). The values of effective radium content are less than the permissible value of 370 $Bq.kg^{-1}$, as recommended by Organization for Economic Cooperation and Development (OECD, 1979).

Comparison of radium equivalent with results in other countries: Table 2 shows a comparison of Ra concentration equivalent in the present work with results obtained in other parts of the world. It can be seen that the values of radium content for cement and gypsum in the present work are comparable with those obtained in the countries listed in the table. For the sand radium content is much lower than the others, except for the Algerian one for sand.

CONCLUSION

The can technique using nitrate cellulose (LR-115 II) is a passive and convenient useful tool for determining the radon exhalation rates as well as the ^{226}Ra contents in some sample of building materials. The results obtained in this survey for local building materials and soil look similar to some available data for building materials and soils reported in literature. The used building materials should be characterized by lower radon concentration to avoid the health hazards. The obtained exhalation rate can be used to estimate the maximum radon concentration in various buildings. It is possible to establish a data base for all building materials available in a local market using this technique with low cost for a large-scale nation-wide indoor radon screening measurement.

ACKNOWLEDGEMENT

The reacherse are thankful to the residents of the study area for their cooperation during the fieldwork.

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