Transactions of the Korean Nuclear Society Spring Meeting
Jeju, Korea, May 23-24, 2019

Assessment of Soil Water Content Parameter on Radon and Thoron Exhalation Rate

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1. Introduction

Trace amounts of ^{238}U and ^{232}Th which decay to radon ^{222}Rn and thoron ^{220}Rn can be found on earth and common building materials.

In the decay process, radionuclides with half-lives less than one year will reestablish equilibrium conditions with their longer-lived parent radionuclides within several years. For this reason, at processing sites what was once a single, long decay series (for example the series for uranium-238) may be present as several smaller decay series headed by the longer-lived decay products of the original series (that is, headed by ^{238}U, ^{234}U, ^{230}Th, ^{226}Ra, and ^{210}Pb in the case of uranium-238). Each of these sub-series can be considered to represent a new, separate decay series [5]. Understanding the physical and chemical processes associated with materials containing uranium, thorium, and radium is important when addressing associated radiological risks.

A part of radon, which is produced mainly on the surface layer of the minerals can eject out of the grains and may emanate into the interstitial space between them. These radon atoms exist in gaseous form and spread into the pore space driven by diffusion and advection. They are dominantly transported by carrier fluids, whereas the radon migration depends upon the fluid flow characteristics of the soil. Some radon gas will eventually migrate upwards to the soil/air interface and exhales out into the atmosphere. The characterization of this transfer process is crucial for the understanding of the following fate of the radioactive rare gas: either as trace substance in atmospheric dispersion or as accumulating contaminant in the indoor environment. This importance made it to an intense subject of investigations.

Radon and its short-lived decay products in the atmosphere are the most important contributors to human exposure from natural sources [4]. Radon escapes from the ground into the air. In the open air, the amount of radon gas is very small and does not pose a health risk. However, soil gas containing radon can enter enclosed spaces whenever it finds an opening or a path. In enclosed spaces like residential homes, schools, office buildings and especially underground mines, radon can sometimes accumulate to relatively high levels and become a health hazard.

Megumi and Mamuro reported on the relationship between the thoron exhalation rate and the soil moisture content by way of an indoor experiment and in situ measurement, respectively. Furthermore, it was reported that the radon exhalation rate was changed by the effect of rain on the soil.

In Korea, some traditional houses still use soil as the main building material to be used as their wall and some daily tools. In this case, radiological consideration on radon and thoron from soil become important. Moreover, it has been proven by many researches that radon has a high contribution on indoor radiation exposure.

In addition, by simultaneous measurement, both types of exhalation rate data can be closely compared to each other. Consequently, we expect that the reliability of data for the radon and thoron exhalation rates is improved.

2. Methods and Results

In conducting this research, theoretical assessment and experimental measurement of radon and thoron exhalation rate are done to give a broader perspective and better understanding on both characteristics.

2.1 Sample Preparation

Before the measurements, soil samples of Daegu were sieved to get 3 different grain size soils (<1 mm, $1 \leq x < 2$ mm, $2 \leq x < 4$ mm) and packed in a closed plastic bag. Then the samples were dried in a temperature-controlled furnace (oven) at 110 C for 24 h to ensure complete removal of moisture.

Before starting the measurement, each sample was kept at the desired measurement conditions for 3-5 days to reach a state of equilibrium between it and the chamber environment.

Later, the samples were placed one at a time in a $29 \times 31.5 \times 48.5 cm^3$ accumulation chamber under a controlled atmosphere.

The water content, θ, is calculated by weighing the dry soil and determining it as zero water content w_0. If we add certain amount of water with w_w weight to the soil, then the water content can be adjusted as

$$\theta = \frac{w_w}{w_0 + w_w} \tag{1}$$

2.2 Measurement System

The ^{222}Rn and ^{220}Rn exhalation rates from surface soil have been determined by using an accumulation chamber equipped with a solid-state alpha particle detector of RAD7. Then, the activity concentrations of parent radionuclides (^{226}Ra and ^{224}Ra) in soil samples collected from the same site have been determined in the laboratory by using HPGe detector.

The accumulation chamber was connected to the RAD7 using hose.

The system was a closed loop in which the gas was circulated continuously with the flow rate generated by an external pump with the same flow rate as the RAD7 pump. The concentrations of radon and thoron released from each specimen inside the chamber were allowed to build up with time and were measured by the RAD7 in a certain cycle time.

To maintain relative humidity of the system, a desiccant is connected between RAD7 inlet and the accumulation chamber.

The schematic system of radon and thoron exhalation measurement is shown in Fig. 1.

RAD7 can assess radon and thoron concentration profile by examining sample continuously within time until radon and thoron considered to reach their saturated region. This method can be used also to check a leakage from the accumulation chamber. The figure of radon and thoron concentration as a function of time in an accumulation chamber is shown by Fig 2.

To get higher concentration of radon and thoron within shorter time, the measurement test is done by using specific soil sample that has high radon concentration from Takandeang, Indonesia.

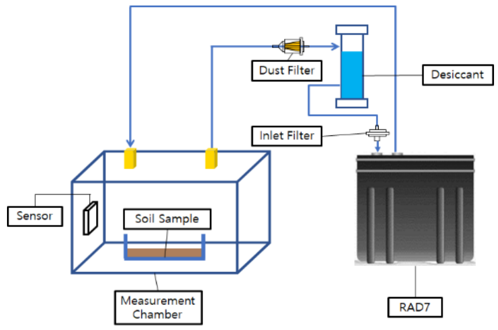

Fig. 1. Schematic system of exhalation measurement.

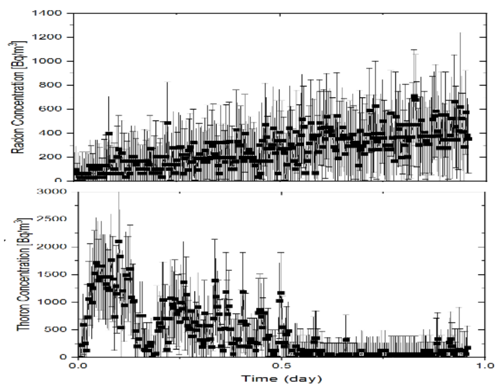

Fig. 2. Radon and thoron concentration as a function of time in an accumulation chamber due to exhalation from soil sample.

2.3 Result

One of important parameter that influences radon and thoron exhalation rates and has been a research interest of many researchers is water content.

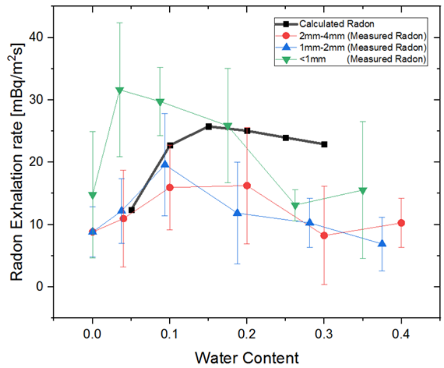

Fig. 3. Relationship between radon exhalation rate and water content

Extracting from the graph, it seems that radon exhalation is increasing until become maximum at 10-20 % water content and start to decrease above this value. The result is in line with Schumann research that obtained 15-20% as the maximum radon exhalation rates. Schumann said that at these soil moisture levels, pore water exists in thin coatings on soil grains that absorb some of the recoil energy of radon atoms as they escape, preventing them from burying themselves in adjacent soil grains and thus increasing the rates.

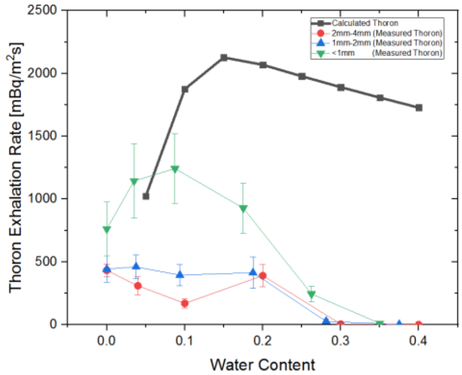

Fig. 4. Relationship between thoron exhalation rate and water content

In case of thoron, the measured thoron exhalation rates give similar trend as the calculated value. However, the measurement result is below the calculated value. This possibly caused by trapped thoron inside soil and decayed before measured by the system.

2.4 Calculation and Assessment

To assess the contribution of water content from soil as a building material and estimate the dose caused by radon and thoron, specific calculation and scenario was made and the result will be presented in Table 3.

After it is released by decay of a radium, radon atom can remain embedded in the same grain, embedded in adjacent grain, or released into a pore space. The transfer process of radon atom inside materials is influenced by some parameters. One important parameter is emanation coefficient. Typically, emanation coefficient of soils is about 0.22 [10]. Further researches have been done and range of emanation coefficient of soil has been scientifically agreed, as shown in Table 1. Considering the characteristic of soil, we apply 0.21 of emanation coefficient to be used in the scenario.

Table 1: Typical values of radon emanation coefficient for different soil textures

Soil types	Emanation coefficient
Sandy	0.14
Sandy loam	**0.21**
Loam	0.24
Silty loam	0.25
Clay	0.28

Theoretically, radon emanation coefficient can be calculated by the following formula:

$$E = \frac{Exh\ S}{\lambda M C_{Ra}} \quad (2)$$

where *Exh* is the radon exhalation rate from sample, λ is the decay constant, *M* is the dry mass of the sample, C_{Ra} is activity concentration of radium, and *S* is the sample surface area (by assuming the room size as 4 x 4 x 3 m, and taking 40% of total surface area that exhale radon and thoron, then it would be 32 m^2). For the scenario, we will use some variation on air exchange rate from 0.2 to 1.2 h^{-1} to see the effect of radon and thoron exposure in the indoor environment.

The measured concentration and other characteristic data of radon and thoron is shown in Table 2.

Table 2: Characteristic data of radon and thoron; and measured concentration of radium

Parameter	Value
^{226}Ra content of soil	61.5 Bq kg^{-1}
^{224}Ra content of soil	67.17 Bq kg^{-1}
Dry bulk density	882.5 kg m^{-3}
Porosity	0.3
Decay constant of radon	2.1×10^{-6} s^{-1}
Decay constant of thoron	0.012 s^{-1}

The dry bulk density, ρ_b was calculated by using the ratio between the dry sample mass and a total volume of sample in a container.

$$\rho_b = \frac{M}{V} \quad (3)$$

A theoretical calculation of radon and thoron exhalation rates was performed in order to assess the validity of the experimental data. Radon exhalation *Exh* from soil radium concentration C_{Ra} was calculated using the following expression.

$$Exh = \sqrt{\lambda D_e}\,\rho_b E C_{Ra} \quad (4)$$

where ρ_s is the soil grain density (2700 kg m^{-3}), ε is porosity of experimental soil, and L is the diffusion length, which is equal to $(D_e/\lambda_{Rn})^{1/2}$.

The relationship between the effective diffusion coefficient and soil water content can be determined by:

$$D_e = D_o \varepsilon\ exp\{-6\theta\varepsilon - 6\theta^{14\varepsilon}\} \quad (5)$$

where, D_0 is the diffusion coefficient of radon in the atmosphere (1.1×10^{-5} m^2 s^{-1}) and θ was the soil water content.

In steady condition, radon and thoron concentration caused by exhalation in indoor environment is calculated by:

$$C_{Rn} = \frac{Exh \times A}{(\lambda + \lambda_w) \times V} \quad (6)$$

where A is the surface area of possible exhalation, in this scenario is assumed to be 32 m^2; λ_w is air removal rate due to exchange rate; and V is air volume of the room.

The annual effective dose from indoor exposure is calculated based on relationship among radon concentration (C_{Rn}), equilibrium factor (F, which is 0.4 for radon and 0.02 for thoron), and dwelling time in the room (T=7000 h). The formula can be written as follows:

$$Dose = C_{Rn} \times F \times T \times EEC \quad (7)$$

where EEC is equilibrium-equivalent concentration to effective dose conversion (9 nSv (Bq h m^{-3})$^{-1}$ for radon and 40 nSv (Bq h m^{-3})$^{-1}$ for thoron).

From Table 3, the behavior of radon and thoron in relationship with air exchange rate is observed. The increase of air exchange rate significantly decreases radon concentration. In a contrary, it doesn't affect to thoron. The short half-life of thoron has caused it to have bigger decay constant that minimize the effect of air exchange rate.

Even though the range of natural effective dose received by world population is 1 – 10 mSv y^{-1}[4], it is clear that radon is one of the main contributors. Combination between moist soil and less ventilation house can be the worst case that will significantly increase the effective dose. In case of thoron, even though relatively moist soil, around 15%, gives higher effective dose, it only contributes 0.65 mSv y^{-1} (with average air exchange rate, 0.5).

Table 3: Radon and thoron concentration and effective dose calculated for different water content, exhalation, and air exchange rate

Radon		Air exchange = 0 (h^{-1})	Air exchange = 0.2 (h^{-1})	Air exchange = 0.5 (h^{-1})	Air exchange = 1.2 (h^{-1})
Water content	Exh (Bq m^{-2} s^{-1})	C_{Rn} (Dose) (Bq m^{-3} (mSv y^{-1}))	C_{Rn} (Dose) (Bq m^{-3} (mSv y^{-1}))	C_{Rn} (Dose) (Bq m^{-3} (mSv y^{-1}))	C_{Rn} (Dose) (Bq m^{-3} (mSv y^{-1}))
0.05	0.0123	3933.16 (99.12)	143.26 (3.61)	58.58 (1.48)	24.62 (0.62)
0.1	0.0227	7213.01 (181.77)	262.72 (6.62)	107.44 (2.71)	45.16 (1.14)
0.15	0.0257	8178.77 (206.10)	297.90 (7.51)	121.82 (3.07)	51.20 (1.29)
0.2	0.0250	7954.38 (200.45)	289.72 (7.30)	118.48 (2.99)	49.80 (1.25)
0.25	0.0239	7605.87 (191.67)	277.03 (6.98)	113.29 (2.85)	47.62 (1.20)
0.3	0.0229	7271.19 (183.23)	264.84 (6.67)	108.30 (2.73)	45.52 (1.15)

Thoron		Air exchange=0 (h^{-1})	Air exchange=0.2 (h^{-1})	Air exchange=0.5 (h^{-1})	Air exchange=1.2 (h^{-1})
Water content	Exh (Bq m^{-2} s^{-1})	C_{Th} (Dose) (Bq m^{-3} (mSv y^{-1}))	C_{Th} (Dose) (Bq m^{-3} (mSv y^{-1}))	C_{Th} (Dose) (Bq m^{-3} (mSv y^{-1}))	C_{Th} (Dose) (Bq m^{-3} (mSv y^{-1}))
0.05	1.0229	56.83 (0.32)	56.57 (0.32)	56.18 (0.31)	55.29 (0.31)
0.1	1.8759	104.22 (0.58)	103.74 (0.58)	103.02 (0.58)	101.40 (0.57)
0.15	2.1270	118.17 (0.66)	117.63 (0.66)	116.82 (0.65)	114.98 (0.64)
0.2	2.0687	114.93 (0.64)	114.40 (0.64)	113.61 (0.64)	111.82 (0.63)
0.25	1.9780	109.89 (0.62)	109.39 (0.61)	108.64 (0.61)	106.92 (0.60)
0.3	1.8910	105.06 (0.59)	104.57 (0.59)	103.85 (0.58)	102.22 (0.57)

3. Conclusions

In this study, we conducted simultaneous laboratory measurements of the radon and thoron exhalation rates in the soil samples with the water content ranging from 5–35%. We obtained the following results. First, water content induced an increase in radon exhalation rates with the increase of water content up to around 10-20 % and then start decreasing above it. Secondly, since water content influence radon exhalation rate due to rain, soil-made building should consider good ventilation to anticipate it. Thirdly, the increase of air exchange rate significantly decreases radon concentration, while it has little effect on thoron.

Finally, even though thoron seems to give small risk, combination between moist soil and less ventilation house will significantly increase the annual effective dose caused by radon and thoron.

ACKNOWLEDGEMENT

One of the authors (Ahmad Ciptadi Syuryavin) would like to thank the Ministry of Research, Technology, and Higher Education (RISTEKDIKTI) of Indonesia for granting a PhD scholarship through its Research and Innovation in Science and Technology Project (RISET-Pro) Program. This work was also supported by KOREA HYDRO & NUCLEAR POWER CO., LTD (K-CLOUD Project 2017-TECH-08).

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