

Article

^{222}Rn Exhalation Rates from Some Granite and Marble Used in Korea: Preliminary Study

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Abstract: The objective of this study was to establish a test method for assessing radon exhalation rates from building materials considering radon related environmental policy and research in Korea. This method was established in consideration of cost-effectiveness based on the International Standards Organization (ISO) method and the closed chamber method, which is an evaluation method for the emission of hazardous chemical substances from building materials in Korea. The assessment of radon exhalation rates from five types each of granite and marble used in the construction industry in Korea gave mean radon exhalation rates of 0.497 ± 0.467 Bq/m²·h from granite and 0.193 ± 0.113 Bq/m²·h from marble, indicating higher radon exhalation rates from granite. These results are consistent with those of a previous study, indicating that granites are more likely to show higher radon exhalation rates than marbles.

Keywords: ^{222}Rn ; exhalation rates; building material; granite; marble



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1. Introduction

More than 50% of the radiation dose received by the general public is due to radon and its progeny [1], which have been considered as one of the main causes of lung cancer [2–4]. Although this radon is mainly emitted in soil or rocks, radon from building materials is another potential source of radon in indoor air [5–10]. Building materials are any materials produced to permanently assemble buildings, and many studies report that building materials contain natural radionuclides, which can determine significant exposure to gamma rays and contribute to indoor radon concentrations [11]. In particular, the indoor contribution of radiation emitted from building materials is estimated to be up to 30% [6], and the indoor contribution of radon has been reported to be 2 to 5% [12].

The European Union (EU) has already implemented provisions and directives since 1989 to control radiation emitted from building materials, taking into account the health effects of radiation exposure (or radon exposure) in the indoor environment. In particular, the European Commission (EC) announced the “ALARA (As Low As Reasonably Achievable) Principle” in 1999 and set the ultimate goal of radiation control in building materials to limit radiation exposure from substances with elevated or high levels of natural radionuclides [13]. The radiation activity index (I), introduced for this purpose, consists of radium, thorium, and potassium as influencing factors, and is generally used as a screening tool to limit exposure to gamma rays from building materials.

As radon was detected in beds and bathroom shelves has been reported through the media recently in Korea, the public's interest in radon in the indoor environment is increasing day by day. Especially, as interest in radon emitted from stone has risen

since a high concentration of radon was detected on a bathroom shelf made of stone, the Ministry of Environment (MOE) of Korea announced the “Guidelines for Reduction and Management of Radon in Building Materials (2019)” in cooperation with relevant government ministries (the Ministry of Land, Infrastructure and Transport (MOLIT) and the Nuclear Safety Committee). The key to this guideline is to control radon contamination in indoor air caused by building materials such as stone by introducing a standard value based on the radiation activity index.

However, the radiation activity index considers only the external exposure caused by direct exposure to gamma rays, and therefore, in order to consider the internal exposure by inhalation of radon and thoron and their short-lived decay products, it is necessary to evaluate radon emitted from building materials [13]. In addition, the activity of naturally occurring radionuclides expressed as a radioactivity concentration index, cannot be satisfactorily predictive of indoor radon concentrations due to radon emitted from soil or building materials [14], it is important to evaluate radon exhalation rates from building materials used in residential environments to assess indoor radon concentrations and sources contributing to health hazards [15]. When materials with relatively high radioactivity, such as imported granite, ceramic, and marble materials, are used as building materials, potential health problems may arise [10], and most of these studies report that it is important to directly evaluate radon of material surfaces [9,16–26]. Radon exhalation rates data generated for various building material samples are useful for validating diffusion-based models for predicting radon flux and concentrations in residential environments [27,28]. There has been a steady increase in interest in radon exhalation rates of building materials over the past few years, and many publications have been published [15]. However, there is currently no standard method for evaluating and managing radon exhalation rates from building materials, including stone materials, worldwide.

Therefore, this study established a standard method for evaluating radon exhalation rates in domestic building materials through a review of previous studies and the evaluation method of emission of pollutants from building materials currently applied in Korea. Furthermore, using the established method, radon exhalation rates were evaluated for 10 types of granite and marble, which are used for decorative use in the domestic building environment, and which have recently become a problem due to radon emission. This study was the first to establish a method for evaluating radon exhalation rates from building materials in Korea and was conducted to set the foundation for radon management in domestic building materials through scientific and reliable data.

2. Materials and Methods

2.1. Samples

Granite and marble are rich in radium and thorium and are known to cause an increase in indoor radon concentration when used as interior decoration [29], and they were reported as the cause of the increase in indoor radon through the media in Korea, and it emerged as a social and environmental problem. Therefore, this study selected 5 types of granite (S1~S5) and 5 types of marble (S6~S10), which are used in the domestic construction industry in Korea as the target building materials for study with the help of the Stone Works Council.

Because radon concentrations are largely affected by moisture, the radon concentrations of dry samples are generally higher than those of wet or damp samples [10]. Therefore, this study conducted the tests after drying the target stone materials in a 105 °C oven for 24 h to remove water content. The size of the samples was set to a common dimension of 160 × 160 mm.

2.2. Measurements of Radon Exhalation Rate

2.2.1. Establishment of Method

Since there is no standard test method for the evaluation of radon exhalation rates from building materials in both domestic and foreign countries, this study preferentially

established a method for evaluating radon exhalation rates from building materials that could be applied in Korea based on previous studies. The assessment methods for radon exhalation from building materials are classified into four main types: (1) closed chamber method; (2) purge and trap method; (3) radon exhalation measurement from surfaces method; (4) in situ radon exhalation measurement [30]. Among them, the closed chamber method, which is based on the principle of wrapping or covering the sample, is usually applied for measurement of radon exhalation rates [31,32]. This method is suitable as the closed space minimizes the impact of the surrounding environment, however, if the radon concentration increases inside the chamber, a reduction in the exhalation rate may occur due to radioactive equilibrium. Furthermore, when short-term measurements are performed at a low concentration, the slope of concentration increases; thus, considering a radioactive equilibrium may render the results clearer. Nevertheless, the closed chamber method is accepted as a test method as its applicability is high compared to the other methods, especially when the cost-effectiveness is considered [30].

On the other hand, the International Standard Organization (ISO) 11665 standard test version stipulates the measurement and application method for radon and its short-lived decay products. ISO 11665-9 is the method which determines the free radon exhalation rates from the mineral-based building material, and measures radon exhaled from the sample of building material by collecting it in a container through constant nitrogen gas flow [33]. This method uses a container to collect the radon gas released together with nitrogen gas in an environment with a relative humidity of 50% and temperature of 20 °C, and measures the concentration of the gases. Unlike the closed chamber method, this method prevents the decrease in the exhalation rate that occurs when the radon concentration increases inside the chamber but instead has to maintain a low flow rate to prevent the occurrence of back-diffusion. This method suffers from a drawback, in that underestimation during measurement may occur if the adequate flow rate is not met. Furthermore, the test conditions are different from the actual normal conditions because dehumidified nitrogen gas is used. The test method is also difficult to apply in the field because of problems such as technical difficulties and high operating costs [30].

At present, the measurement of indoor air pollutants released from building materials are assessed in South Korea according to the “Indoor Air Quality Testing Standard”. It is a method for assessing the unit exhalation rates of volatile organic compounds and formaldehyde from building materials using a small emission-test chamber under conditions of constant temperature, relative humidity, and air ventilation frequency, and is a version of the closed chamber method. Accordingly, based on the small chamber method and considering the radon exhalation rates assessment method’s applicability in Korea and its economic feasibility, this study established an assessment method for radon exhalation rates from building materials. This was done by combining the closed chamber method with the ISO method to mitigate the disadvantages of the closed chamber method.

Figure 1 illustrated the method used in this study to evaluate radon exhalation rates from building materials. Based on the small chamber method, a 20 L chamber was used to maintain the temperature and relative humidity (RH) inside the chamber at 25 °C and 10% or less, respectively, while the test period was set to 99 h, thereby overcoming the limitation of the conventional method that required extended hours. Furthermore, nitrogen gas, which is proposed as the carrier gas in ISO 11665-9, was used as the radon carrier gas in this study to minimize the error by increasing the background concentration, which could occur in the case of applying regular air as the carrier gas. It was assumed that the back-diffusion effect, which could be produced by the carrier gas, would not occur because the volume of the chamber was ten times larger than the volume of the sample [9,18,34].

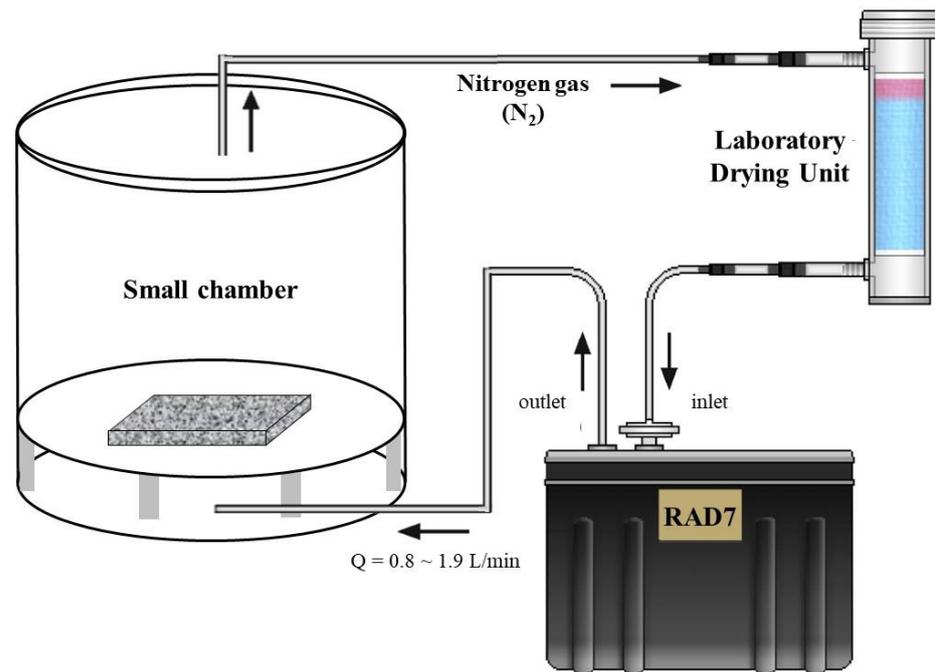


Figure 1. The experiment system used to assess ^{222}Rn exhalation rates from building materials.

To assess the radon exhaled from the building materials, the following procedure was performed. First, prior to the start of the test, a prepared sample was placed in the chamber and it was sealed. The radon was removed from inside of the chamber by injecting nitrogen for about 5 min. In addition, RAD7 (DURRIDGE Company Inc., Billerica, MA, USA), a continuous radon detector, was connected such that the nitrogen gas could be circulated in the closed chamber with a flow rate of 0.8–1.9 L/min. Then, the radon concentration in the chamber was measured at 1 h intervals.

This study prevented the back-diffusion by using a 20 L small chamber, which was ten times larger than the volume of the test sample and also reduced the test time. Furthermore, to eliminate the effect of background concentration and prevent underestimation due to radioactive equilibrium, nitrogen gas was used as the carrier gas. This enabled the air inside the closed chamber to be circulated, and hence mitigated the limitation of the conventional closed chamber method. The method established in this study has the advantage of being easy to perform and cost-effective, and it is suitable as a standard test method in Korea since it can be applied to the emission test method of building materials currently implemented in Korea.

2.2.2. Calculation of Radon Exhalation Rate

In this study, the radon exhalation rates from building materials were calculated based on the method used in a previous study [14]. Equation (1) calculates the change in radon concentrations in the chamber with time that is caused by radon exhaled from the building materials.

$$C = C_0 \cdot e^{(-\lambda \cdot t)} + \frac{E \cdot (1 - e^{-\lambda \cdot t})}{\lambda \cdot V}, \quad (1)$$

where E is the radon exhalation rate (Bq/h), λ is the damping constant of ^{222}Rn (/h), C_0 is the initial radon concentration in the chamber (Bq/m³), C is the equilibrium concentration of radon (Bq/m³), t is time (h), and V is the total volume of the analytical system (m³).

Equation (2) summarizes the above equation for calculating the radon exhalation rate E , and finally, Equation (3) calculates the amount of radon exhaled from a building material per hour.

$$E_{222} = \frac{(C - C_0 \cdot e^{-\lambda_{222} \cdot t})}{1 - e^{-\lambda_{222} \cdot t}} \cdot \lambda_{222} \cdot V, \quad (2)$$

$$\therefore E_{222} = (C_m + \lambda_{222} \cdot C_0) \cdot V, \quad (3)$$

where E_{222} is the radon exhalation rate (Bq/h), λ_{222} is the damping constant of constant of ^{222}Rn (/h), C_0 is the initial radon concentration in the chamber (Bq/m³), C_m is the slope by the least square method of the radon growth curve (Bq/m³/h), and V is the total volume of the analytical system (m³)

The amount of radon exhaled per hour from a unit area (Bq/m²·h) can be calculated by dividing the radon exhalation rates calculated in Equation (3) by the area of the building material used in the test.

3. Results and Discussion

Table 1 shows the assessment results of radon exhalation rates from the building materials. The average radon exhalation is 0.497 ± 0.467 Bq/m²·h from the five granite samples and 0.193 ± 0.113 Bq/m²·h from the five marble samples, which indicates that in general the radon exhalation rates of granites are higher. Granite building material S2 shows the highest radon exhalation rates at 1.247 Bq/m²·h, while marble S10 shows the lowest exhalation rates at 0.072 Bq/m²·h. However, there is no statistically significant difference between the radon exhalation rates of granites and marbles ($p > 0.05$) and the large variance was observed between the samples.

Table 1. The results of radon exhalation rates from building materials.

	Granite (N = 5)		Marble (N = 5)		<i>p</i> -Value
	Mean	S.D.	Mean	S.D.	
E_{222} (Bq/m ² ·h)	0.497	0.467	0.193	0.113	>0.05

At present, few studies have assessed the amount of radon exhaled from stone materials, such as granites and marbles. Furthermore, because there is no standardized test method, existing studies have applied different methods and it is difficult to compare the results directly. However, some previous studies were performed using a similar method as this study. Dabayneh [35] conducted the assessment of radon exhalation rates for building materials used in Palestine, such as granites and marbles, and reported that the mean radon exhalation rates from granites was 0.146 Bq/m²·h and from marbles was 0.127 Bq/m²·h, indicating that the radon exhalation rates from granites was slightly higher than that from marbles. According to Chen, Rahman, and Atiya [9], the radon exhalation rates from four types of granite used in Canada varied from 0.004 Bq/m²·h (0.1 Bq/m²·d) to 0.017 Bq/m²·h (0.4 Bq/m²·d), while the mean radon exhalation rate from 33 types of granite was 1.75 Bq/m²·h (42 Bq/m²·d). Therefore, the radon exhalation rates from granites are higher-similar to the result of Dabayneh [35]. Furthermore, according to a study performed in Iraq, the radon exhalation rate is approximately 1.24 Bq/m²·h from marbles and 2.3 Bq/m²·h from granites, again showing that the radon exhalation from granites is higher [36]. An assessment of radon exhalation from building materials in Libya gave a mean radon exhalation rate of 1.751 Bq/m²·h from six types of granite and 0.593 Bq/m²·h from two types of marbles, demonstrating that the radon exhalation rates from granites were approximately three times higher than that from marbles [10]. Sabbarese, Ambrosino, D’Onofrio, and Roca [2] assessed the radon exhalation rates of two types of marble in Southern Italy. The results showed that $(47.1 \pm 5.1) \times 10^{-2}$ and $(111.2 \pm 3.9) \times 10^{-2}$ Bq/m²·h, which was slightly higher than that of this study.

The above studies were similar to this study insofar as they assessed the radon exhalation rates from building materials by applying the closed chamber method or the sealed can technique, similar to the method used in this study. Dried samples were also used in four studies for better comparison [2,10,35,37]. The results of these studies are consistent with the results of this study, indicating that although the radon exhalation rates show large deviations between the respective construction materials, granites are more likely to show higher radon exhalation rates compared to marbles (Table 2).

Table 2. ^{222}Rn exhalation rates from building materials in different study.

Country	Method	E_{222} (Bq/m ² ·h)		Note	Reference
		Granite	Marble		
Korea	Closed chamber method + ISO method	0.497	0.193	Dried samples	Present study
Palestine	Can technique	0.146	0.127	Dried samples	Dabayneh [35]
Canada	Closed chamber method	1.750	0.010	-	Chen, Rahman, and Atiya [9]
Iraq	Sealed can technique	2.300	1.240	-	Najam, Tawfiq, and Mahmood [36]
Egypt	Sealed can technique	0.088	-	Dried samples	Shoeib and Thabayneh [37]
Libya	Closed chamber method	1.751	0.593	Dried samples	Saad, Al-Awami and Hussein [10]
Italy (Southern)	Sealed can technique		$(47.1 \pm 5.1) \times 10^{-2}$, $(111.2 \pm 3.9) \times 10^{-2}$	Dried samples	Sabbarese, Ambrosino, D'Onofrio and Roca [2]

Granite contains a high value of radium since it contains feldspar minerals that can capture uranium from circulating solutions [38], therefore, it is considered that granite showed higher radon emission than marble in this study and previous studies. Furthermore, it suggests that indoor radon concentration could be significantly increased according to granites use [39] since granite acts as a major source of radon in a house depending on its characteristics when used for large-scale tiling in an enclosed space [40].

According to studies of radon exhaled from domestic and foreign building materials, high concentrations of radon can be exhaled from stone materials, such as imported granite, ceramic, and marble or byproduct building materials [41]. However, although this study only assessed radon exhalation rates of some interior stone materials from among the many indoor decorative building materials that have become an issue nowadays in Korea, the most important thing in this study is that the results could help control the quality of domestic building materials. Therefore, further studies are required to construct a database of radon exhalation rates from building materials used in the Korean construction industry. Based on the results of the latter, a list of regulatory targets for building materials should be compiled and a management plan should be established.

4. Conclusions

In recent years, the occurrence of radon in residential environments has been repeatedly raised as a public health issue in Korea. Because the media has reported problems related to radon exhaled from stone materials, there is a growing interest in the radon emitted from building materials and a new assessment method to assist in its control is required. Therefore, this study was conducted as an effort to prepare a regulation-compliant assessment method for radon exhalation rates from building materials which would be suitable in the Korean contest.

This study established an assessment method for radon exhalation rates from building materials, which could be applied in Korea as a method to enable control of radon exhalation from building materials. The assessment steps are as follows:

Step 1. After placing the sample in the 20 L chamber and sealing it, nitrogen is injected for about 5 min, thereby removing the radon from inside of the chamber.

Step 2. The temperature and relative humidity in the chamber are maintained at 25 °C and 10% or less, respectively.

Step 3. The closed internal air is circulated with the flow rate of 0.8–1.9 L/min by connecting a continuous radon detector (RAD7).

Step 4. The test is performed for 99 h, and the radon concentration is measured at certain intervals.

Ten types of granite and marble were assessed in this study, and the result was a mean radon exhalation rate of 0.497 Bq/m²·h from granites and 0.193 Bq/m²·h from marbles. It was similar to the results of previous studies; the radon exhalation rates from granites were higher than that of marbles.

This paper strongly recommends that additional studies for standardization of the test methods of radon exhalation rates in domestic building materials and for assessment of radon exhalation rates from building materials distributed in Korea, including more types of stones, be continuously performed.

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