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RADON EXHALATION RATES OF SOME GRANITES USED IN SERBIA

by

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In order to address concern about radon exhalation in building material, radon exhalation rate was determined for different granites available on Serbian market. Radon exhalation rate, along with mass exhalation rate and effective radium content were determined by closed chamber method and active continuous radon measurement technique. For this research, special chambers were made and tested for back diffusion and leakage, and the radon concentrations measured were included in the calculation of radon exhalation. The radon exhalation rate ranged from 0.161 Bq/m²h to 0.576 Bq/m²h, the mass exhalation rate from 0.167 Bq/kgh to 0.678 Bq/kgh, while the effective radium content was found to be from 12.37 Bq/kg to 50.23 Bq/kg. The results indicate that the granites used in Serbia have a low level of radon exhalation.

Key words: ^{222}Rn, indoor radon, radon exhalation, radon chamber, granite

INTRODUCTION

Radon (^{222}Rn) is naturally existing radioactive gas with a half-life of 3.83 d, which emanates from soil as well as from building materials, into outdoor or indoor air. Radon decays into series of short-life progeny (^{218}Po, ^{214}Pb, ^{214}Bi, and ^{214}Po), which could be inhaled into the human respiratory tract and impose a significant health hazard for the population. There is a considerable public concern about radon exhalation from building materials, especially those used for interior decoration such as granite tiles that are considered as an important source that contributes to indoor radon concentration through exhalation from walls and floors [1].

Radon gas is formed inside building materials by decay of the parent nuclide ^{226}Ra. However, it is not possible to determine the radon exhalation rate simply from the activity concentration of ^{226}Ra. Instead, one must measure radon exhalation rates directly from the surface of the material, especially for granites [2, 3].Various methods have been developed for measuring the radon exhalation rate from building materials, such as the accumulation method, first proposed by the Lawrence Berkeley Laboratory, the charcoal method, and the solid state nuclear track detector method [4-9]. In addition, the closed chamber method, essentially a kind of accumulation method, which takes into account the leakage of the accumulation chamber and the back diffusion rate, is widely used for measurement of radon exhalation rate from building materials [10]. For application of this method it is necessary to conduct continuous monitoring of radon concentration inside the chamber.

Radon exhalation rates from building materials vary widely and some building materials may contribute significantly to indoor radon levels. Although many data are available, especially for granites commonly used in home decoration, large variability has made the existing data insufficient to conclude which types of granites have higher radon exhalation rate than others. Even though such variation of radioactivity in natural materials cannot be reduced by conducting more and intensive measurements, it is of interest to assess radon contribution from building materials commonly available in the Serbian market, and to verify whether granite materials used in Serbian homes have any difference in radon exhalation characteristics compared to the reported data [11-14].

THEORETICAL APPROACH

The radon exhalation rate E_0 is defined as the quantity of radon activity liberated from the surface area of building materials per unit time [Bqm^{-2}h^{-1}].

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Using this value and the area of the indoor surface, radon emanated per unit time could easily be calculated and used to estimate the radon concentration in the indoor environment.

Among various techniques for determining radon exhalation rate, a closed chamber technique was chosen for this research and a special glass chamber was made for this purpose. The sample under study was sealed inside the chamber until saturation time occurred.

As the radon concentration around the sample grows, radon atoms are diffusing back into the material due to porous nature of the samples thus lowering the equilibrium radon concentration inside the accumulation chamber. This phenomenon is known as back diffusion, defined through back diffusion coefficient λ_b [h^{-1}], which causes an underestimate of true radon exhalation rate. However, if the volume of sample is 10 % or less, according to the volume of the chamber, the back diffusion effect can be neglected [1, 15, 16]. The chamber was built in this manner and back diffusion was checked for each tested sample. The initial growth of radon concentration was not registered and thus back diffusion was neglected (fig. 1.)

However, another factor that could also underestimate the true exhalation rate is leakage. The chamber leakage is described by the rate of ventilation, λ_v [h^{-1}], and in this research, it was determined through radon decay curves. After measuring background radiation, the chamber without a sample was sealed and radon decay was monitored. Through the mass balance, the chamber leakage is calculated as [1, 12]

$$\lambda_v = \frac{M_I - M_L}{C_b - C_0} \qquad (1)$$

where M_I and M_L are initial slopes of ideal decay curves of radon concentration inside the chamber, in the case of no leakage and in the case with leakage, respectively (fig. 2). C_b is the initial concentration of radon inside the chamber in leakage measurement ($C_b = 16$ Bq/m^3), and C_0 – the background radon concentration in the laboratory ($C_0 = 12$ Bq/m^3).

Since in case of no leakage, decay curve takes the simple form: $C_I(t) = C_b e^{-\lambda t}$, the initial slope M_I can
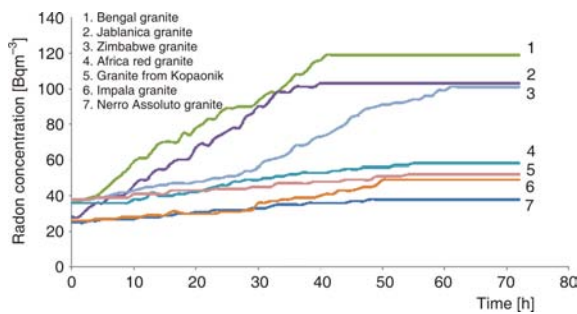

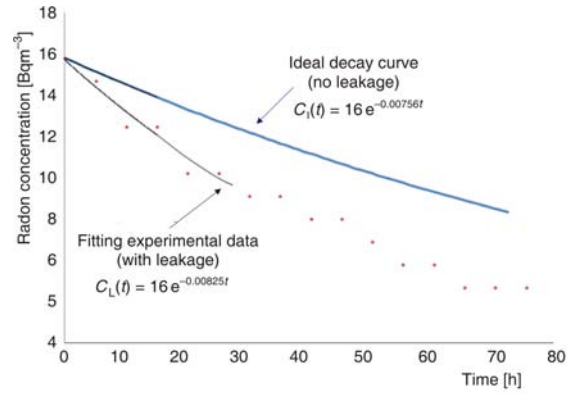**Figure 1. Radon concentration growth in time for different types of granites**

Figure 2. The radon decay curves (experimental and ideal)

be calculated directly as $M_I = -C_b\lambda = -0.121$ Bq/m^{3}h, where the value of radon decay constant λ is used $\lambda = 2.1 \cdot 10^{-6}$ s^{-1} = 0.00756 h^{-1}.

The initial slope of the curve for decay with leakage is found to be $M_L = -0.132$ Bq/m^{3}h. By substituting these values into eq. 1, the leakage rate is found to be: $\lambda_v = 0.0028$ h^{-1}. This result is consistent with the previously published values of leakage coefficients ranging from 0.002 h^{-1} to 0.033 h^{-1} for the close chambers with volumes from 0.013 m^3 to 1.5 m^3 [12, 16].

By solving radon mass transfer equation with neglected back diffusion, radon exhalation rate can be estimated as [1]

$$E_0 = [C_\infty(\lambda + \lambda_v) - \lambda_v C_0]\frac{V}{A} \qquad (2)$$

where C_∞ is the equilibrium radon concentration inside the chamber, V – the volume of the chamber, and A – the surface of the sample.

The mass exhalation rate E_m [Bqkg^{-1}h^{-1}], which is often the criteria for evaluation of materials used in building of apartment houses, and the effective radium content Ra$_{eff}$ [Bqkg^{-1}], are given by [1]

$$E_m = E_0 \frac{A}{m} \qquad (3)$$

and

$$\text{Ra}_{eff} = \frac{E_m}{\lambda^*} \qquad (4)$$

where λ^* is the total removal constant of radon transport process with neglected back diffusion, $\lambda^* = \lambda + \lambda_v$.

EXPERIMENTAL SETUP

For the purpose of this experiment, a special closed chamber was created (fig. 3). The chamber was designed entirely out of thick glass and it consists of three independent compartments, providing measurements with three samples at the same time, thus increasing the research efficiency. Each compartment having volume of 0.45 m^3 is tightly sealed and isolated

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Table 1. Radon exhalation rates from various granite samples

Source	Mass [kg]	Exhalation rate [$\mathrm{Bqm^{-2}h^{-1}}$]		Mass exhalation rate [$\mathrm{Bqkg^{-1}h^{-1}}$]		Effective radium content [$\mathrm{Bqkg^{-1}}$]	
Nerro Assoluto	0.867	0.161	0.024	0.167	0.025	12.42	1.86
Africa red	1.345	0.249	0.037	0.167	0.025	12.37	1.85
Granite from Kopaonik	0.723	0.215	0.032	0.266	0.039	19.71	2.95
Impala granite	0.821	0.217	0.032	0.238	0.035	17.63	2.64
Granite from Jablanica	0.950	0.493	0.074	0.467	0.070	34.65	5.19
Zimbabwe	0.623	0.469	0.070	0.678	0.101	50.23	7.53
Bengal	0.928	0.576	0.086	0.564	0.084	41.78	6.26

Figure 3. Chamber assembly for radon exhalation measurements

from the environment. Samples, from which radon gas emanates freely, are located at the bottom of each compartment.

Active, continuous, short-term measurements were performed by using SafetySiren radon detector (Family Safety Products Inc.) which is certified by Environmental Protection Agency. It is important to mention that the measuring error of this device is 15 %, which will be outlined in the results.

Seven days before the experiment chamber was opened and the detectors were placed in order to determine background radon concentration, along with concentration in the chamber, which varied for each set of measurements in the range from 12 Bq/m³ to 32 Bq/m³. This procedure was applied for each set of samples.

The samples were then dried in oven at 110 ºC in order to eliminate moisture content and then put into compartments until saturation time occurred (5-7 days). Radon concentration was monitored hourly via Webcam and Chronolapse software, since it was crucial to observe if sudden radon fluctuations occurred. Chronolapse is a tool for creating time lapses and stop motions. It can take screenshots or webcam captures (or both simultaneously) at user defined intervals and each change of radon concentration on the detectors was noted.

RESULTS AND DISCUSION

Table 1 shows the radon exhalation rates and effective radium content of the granite samples available at the Serbian market. Calculations are based on radon concentrations measurements and the application of eqs. (2-4).

The results for the exhalation rates ranged from 0.161 Bq/m²h for Nerro Assoluto black granite to 0.576 Bq/m²h for Bengal granite. The highest mass exhalation rate was found to be 0.678 Bq/kgh for Zimbabwe granite, while the lowest was established to be 0.167 Bq/kgh both for Nerro Assoluto and Africa red granite. The highest radium effective content was found to be 50.23 Bq/kg in Zimbabwe granite while the lowest one was 12.42 Bq/kg in Nerro Assoluto and 12.37 Bq/kg in Africa red granite.

Measurements of the radon exhalation rate from 33 samples of granite available on the Canadian market for interior home decoration [13] and 32 samples of granite mainly from Saudi Arabia, but also imported partly from other countries [17], produced similar results to each other: from non detectable values to almost 11 Bq/m²h, with an avarage of less than 2 Bq/m²h. Moreover, the effective radium content ranged from 13.3 Bq/kg to 225 Bq/kg [17]. Based on these data and the values given in tab. 1, we conclude that granites used in Serbia have low level of radioactivity, but with considerable variation from sample to sample.

CONCLUSION

Closed chamber specially designed, with three independent compartments and low leakage of radon, enabled the active and continuous measurements of radon exhalation using commercially available Safety Siren detectors. The results obtained for the seven granite samples sold on Serbian market show a low level of radon exhalation and an effective content of radium, but also significant differences between the samples. Hence, using this simple and quick measuring technique the characterization of construction materials on radon emanation can be performed successfully.

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AUTHOR CONTRIBUTIONS

The experiments and calculations were carried out by M. D. Nikolić. The results were analysed and manuscript was prepared jointly by both authors.

REFERENCES

[1] Abo-Elmagd, M., Radon Exhalation Rates Corrected for Leakage and Back Diffusion – Evaluation of Radon Chambers and Radon Sources with Application to Ceramic Tile, *Journal of Radiation Research and Applied Sciences, 7* (2014), 4, pp. 390-398

[2] Ackers, J. G., Direct Measurement of Radon Exhalation from Surfaces, *Radiat Prot Dosim, 7* (1984), 1-4, pp. 199-201

[3] Mustonen, R., Natural Radioactivity in and Radon Exhalation from Finnish Building Materials, *Health Phys., 46* (1984), 6, pp. 1195-1203

[4] Ingersoll, G., A Survey of Radionuclide Contents and Radon Emanation Rates in Building Materials Used in the US, *Health Phys., 45* (1983), 2, pp. 363-368

[5] Ingersoll, G. , Stitt, D., Zapalac, H., A Fast and Accurate Method for Measuring Radon Exhalation Rate from Building Materials, *Health Phys., 45* (1983), 2, pp. 550-552

[6] Iimoto, T., *et al*, Development of a Technique for the Measurement of the Radon Exhalation Rate Using an Activated Charcoal Collector, *J. Environ. Radioac., 99* (2008), 4, pp. 587-595

[7] Abu-Jarad, F., Fremlin, J. H., Bull, R., A Study of Radon Emitted from Building Materials Using Plastic-Track Detectors, *Phys. Med. Biol., 25* (1980), 4, pp. 683-694

[8] Petropoulos, N. P., Anagnostakis, M. J., Simopoulos, S. E., Building Materials Radon Exhalation Rate: ERRICCA Intercomparison Exercise Results, *Science of The Total Environment, 272* (2001), 1-3, pp. 109-118

[9] Sahoo, B. K., *et al.*, Radon Exhalation Studies in an Indian Uranium Tailings Pile, *Radiation Measurements, 45* (2010), 2, pp. 237-241

[10] Samuelsson, C., The Closed-Can Exhalation Method for Measuring Radon, *J. Res. Natl. Inst. Stand. Technol., 95* (1990), 2, pp. 167-169

[11] Arafa, W., Specific Activity and Hazards of Granite Samples Collected from the Eastern Desert of Egypt, *J. Environ. Radioactivity, 75* (2004), 3, pp. 315-327

[12] Chao, C. Y. H., *et al.*, Determination of Radon Emanation and Back Diffusion Characteristics of Building Materials in Small Chamber Tests., *Build Environ, 32* (1997), 4, pp. 355-362

[13] Chen, J., Rahman, N. M., Atiya, I. A., Radon Exhalation from Building Materials for Decorative Use, *J. Environ. Radioactivity, 101* (2010), 4, pp. 317-322

[14] Keller, G., Hoffmann, B., Feigenspan, T., Radon Permeability and Radon Exhalation of Building Materials, *Sci. Total Environ., 272* (2001), 1-3, pp. 85-89

[15] Abo-Elmagd, M., Soliman, H. A., Daif, M. M., Radon Effective Dose from TENORM Waste Associated with Petroleum Industries, *Radiat Prot Dosim, 136* (2009), 3, pp. 209-215

[16] Ujić, P., *et al.*, Internal Exposure from Building Materials Exhaling ^{222}Rn and ^{220}Rn as Compared to External Exposure Due to Their Natural Radioactivity Content., *Appl. Radiat. Isot., 68* (2010), 1, pp. 201-206

[17] Al-Jarallah, M., Radon Exhalation from Granites Used in Saudi Arabia, *J. Environ. Radioactivity, 53* (2001), 1, pp. 91-98

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ЈАЧИНА ЕКСХАЛАЦИЈЕ РАДОНА ИЗ ГРАНИТА КОЈИ СЕ КОРИСТЕ У СРБИЈИ

У намери да се обрати пажња на ексхалацију радона из грађевинског материјала, одређена је јачина ексхалације радона различитих гранита који су расположиви на тржишту у Србији. Јачина ексхалације радона, заједно са масеном јачином ексхалације и ефективним радијумским садржајем, одређена је методом затворене коморе и поступком активног непрекидног мерења радона. За ово истраживање начињене су посебне коморе, проверене на повратну дифузију и цурење, а измерене вредности концентрације радона употребљене су у прорачуну ексхалације. Јачина ексхалације радона износила је од 0.161 Bq/m²h до 0.576 Bq/m²h, масена јачина ексхалације од 0.167 Bq/kgh до 0.678 Bq/kgh, док је ефективни садржај радијума био од 12.37 Bq/kg до 50.23 Bq/kg. Резултати указују да су гранити коришћени у Србији са ниским нивоом ексхалације радона.

Кључне речи: ^{222}Rn, радон у затвореном простору, ексхалација радона, радонска комора, гранит