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Effect of sorption moisture content of heavy concrete on radon emanation

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Abstract. This article provides the results of a study to determine the emanation coefficient of artificial construction material, namely heavy concrete produced by semi-dry pressing, depending on equilibrium sorption moisture content achieved in the desorption stage. We measured radon volumetric activity in a sealed chamber at different air relative humidity, determined the specific activity of naturally occurring radionuclides (NORs), and calculated the specific effective activity of NORs. Then we obtained mathematical models of the dependence of the emanation ratio on the degree of pores filling with water, and it was determined that the emanation coefficient increases by almost 50 % in the range of relative humidity typical for residential and public buildings, which necessitates the numerical control of this parameter in order to accurately assess the dose load on the population.

1. Introduction

Currently, it has been determined that the main contribution to the population exposure is made directly by natural radionuclides [1], and radon and its decay products make the largest contribution to the total population exposure dose. Due to internal exposure, radon can cause oncological diseases, including lung cancer.

The main radioactive nuclides contained in rocks are radium, thorium, and potassium. Radioactivity of ready-made construction materials is caused by naturally occurring radionuclides (NORs) in original raw materials and is characterized by the value of the specific effective activity (A_{eff}). Radioactivity of construction materials depends on the type and place of extraction of mineral raw materials, and the content of NORs in industrial waste which is used to improve the properties and characteristics of the material or to reduce the cost of its manufacture. In this case, the values of gamma activity and emanation are determined by the composition of building mixes [2, 3, 4, 5, 6].

Such radioactive emanations as radon (Rn) and thoron (Tn) are exhaled from building envelopes and accumulate in the indoor air. Due to the short half-life, thoron makes an insignificant contribution to the total dose load, as only a small fraction of thoron exhales from construction materials into the air of the room and soil under the building. Concentration of radon is largely dependent on the ventilation mode, and low air exchange in the room leads to quite high levels of radiation. Thus, the man-made altered radiation background is formed in building spaces.

It is vital to understand the processes of generation and migration of radon and the factors that influence these processes, to ensure radiation safety and to take appropriate measures to reduce dose commitment on the population.

Radon emanation is a complex process involving the release of radon atom from the solid phase, diffusion in liquid and gas media, adsorption on the walls of cracks and capillaries of materials [7].

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The emanation coefficient E of the material is composed of emanating due to the effect of the recoil of radon atoms E_r and emanating due to diffusion E_D:

$$E = E_r + E_D$$

Emanating due to the recoil effect is practically independent of the human environment. Due to the energy released during the decay of radium, a radon atom acquires momentum and shifts by a distance equal to the range of the atomic recoil in the medium. At the expense of the recoil effect, the emanation atoms, formed in the surface layer with a thickness less than the range of atomic recoil, exhale to the media surrounding the sample [8].

After exhaling from the substance grains, radon atoms enter the pore space of the material. Radon can diffuse in pore voids, and some fraction of atoms decays directly in the body of the material, while the other part emanates from the building structure into the room air and decays.

Emanating depends on mineral composition, uniformity of radium-226 (^{226}Ra) distribution in the material, characteristics of pores (sizes, quantity, and type), specific surface area and grain size distribution of the test sample.

The diffusion component of emanation depends on the internal structure of the material and external environmental conditions, such as temperature, humidity, atmospheric pressure, and air mobility near [9].

One of the main external factors that can significantly affect the amount of radon exhaled under normal conditions is humidity. This fact is confirmed by the studies of emanation of rocks and soils.

In article [10], the authors summarize the basic theories about the influence of humidity on emanating of rocks from the insignificant influence of humidity [11, 8] to a stable relationship between these characteristics. An experiment within the research showed that an increase in moisture content for most of the studied samples resulted in a rise in the value of the emanation coefficient. Upon reaching a degree of pores filling with water close to complete water saturation, some samples tended to decrease their emanation, while others remained passive to the external impact of moisture. The authors in [12] studied emanating of radioactive samples into air and water. It turned out that for most of the samples, emanation into water is 1.1 ... 2.5 times more than into the air. Based on the studies conducted under the laboratory conditions, the authors in [10] concluded that there are rocks with a different emanation response to changes in humidity, which causes a discrepancy of the existing data.

The authors in [13] studied the effect of humidity of uranium tailings in the form of sand on the change in the emanation rate. The effect of increasing the emanation coefficient with rising humidity was attributed to the accumulation of radon atoms in pores of the material when the pore space is filled with water, which prevents the introduction of recoil atoms into the crystal lattice of neighbouring grains.

Rising of radon emanation at the increase in the moisture content of the material was also calculated using the modified Monte Carlo program TRIM [14]. In this case, the distribution of moisture in a porous medium is discussed at the level of the capillary theory. It is noted that the emanation of the material quickly reaches the value of emanation in a saturated state when humidity ranges from 10 % to 30 %. Upon reaching 30 %, the pore surface forms a thin film which impedes the incorporation of the recoil radon atom into another part of the pore wall.

The data obtained by the authors in [15] are worth considering. The amount of the maternal isotope of radium-226 in concrete samples increased in two different ways. The first method was to add radium bromide. In the second method, the amount of radium in concrete was increased due to the enrichment of uranium ore. The authors observed a strong dependence of the radiation dose on the enrichment method. For a sample enriched by uranium ore, radon emanation was about ten times less, and the authors observed a pronounced dependence of the radon release on the water content.

The authors of [16] obtained the emanation coefficients for granites, used in construction, for a dry, natural, and wet state, calculated the speed of emanating as well as alpha-equivalent dose. Results of the test showed that even with a slight increase in moisture content, the emanation coefficient increased significantly. Thus, the authors attribute the resulted data to the possibility of trapping radon atoms in the pore space by water.

Production of construction materials utilizes mixtures of natural raw materials which undergo significant changes in the process of technological conversions. The emanation of such materials can vary significantly [17]. Burnt materials such as ceramic bricks, tiles, expanded clay or unhydrated cement demonstrate the least radon release, unlike sand, gravel, hydrated cement, cement concrete, and mortars which show the greatest one.

For instance, as a result of hydration and hardening of hydraulically active cement and lignite coal fly ash, emanating of artificial stones from these materials increases by almost 10 times [2, 3]. Article [3] demonstrates the possibility of analysing the specific effective activity and emanating ability of samples of cement concretes and mortars based on the data on the radioactivity of their components taking into account chemically bound water, change in emanation as a result of cement hydration and time of hardening of the binding. The analysis has revealed that over time, the emanation of cement and cement-ash samples decreases [3, 18].

In general, it is obvious that the effect of humidity on the materials emanation is mostly considered by works connected, mainly, to the rocks and soils representing objects that are different in composition and structure from multi-component building materials obtained as a result of technological conversion (including the usage of hydraulically active artificial binders, such as cement) to ensure the required performance properties. Thus, the question of studying the emanation of building materials under operating conditions of various types of premises is relevant and requires further study in order to ensure radiation safety of the population.

In this research, we set the goal to establish the influence of relative humidity on the emanation of radon from heavy concrete made by semi-dry pressing. During the study, patterns of the formation of the number of emanations were revealed and a mathematical model was proposed.

2. Methods

The paper presents results aimed at identifying changes in the emanation coefficient of heavy concrete samples depending on the relative humidity of the ambient air and the sorption moisture of the samples. Samples were tested in the radiation monitoring laboratory of Siberian Federal University.

To determine the parameters of the radon emanation process from finished construction products, samples of heavy concrete with a total weight of 9.9 kg, a volume of 0.00423 m^3, and an estimated average density ρ = 2340 kg/m^3 in the dry state were selected. Concrete composition per 1m^3: CEM I 42.5 GOST 31108-2016 – 520 kg; sand and gravel (aggregate), grain size up to 10 mm – 1810 kg; water – 230 kg. The porosity of the material was 10 %, while the moisture content by volume with complete water saturation of the heavy concrete sample was 9.49 %.

2.1. Determining the specific activity of radionuclides and the specific effective activity

We determined the NORs specific activity in the test material using PROGRESS gamma-spectrometer following the procedure [19]. The PROGRESS software package is designed to analyse spectrograms of a standard radionuclide composition: ^{40}K, ^{137}Cs, ^{232}Th, ^{226}Ra in equilibrium with daughter products.

A pre-weighed crushed sample with grain sizes of 0.5-1 mm was placed in a standard measuring cell and sealed. To establish a radioactive equilibrium between radon and its daughter products, the sample was aged for 14 days in the same state. The content of basic radionuclides in the test sample was measured on a gamma spectrometer three times with a 180-minute exposure.

The specific effective activity of natural radionuclides was determined by [20]

$$A_{eff} = A_{Ra} + 1.3A_{Th} + 0.09A_K \quad (1)$$

2.2. Determining the radon concentration in the air of a sealed container

AlphaGUARD Radon monitor PQ2000 was used for measuring. The method is based on direct measurement of the volumetric activity of radon in the air of a sealed container under the modes of passive sampling of the air and pumping it through an ionization chamber.

The emanation of radon from concrete into the air of a sealed container was analysed on the samples of various sorption moisture. The test samples were placed into a sealed container of 0.05 m^3, where a switched-on radon monitor was installed (Fig. 1).

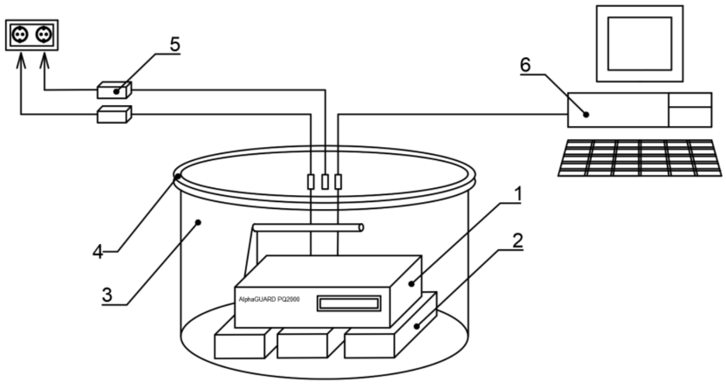

Figure 1. General Installation Diagram:
1 – AlphaGUARD radon monitor with a power supply;
2 – test samples; 3 – sealed steel emanation container; 4 – steel locking ring; 5 – fan power unit;
6 – personal computer.

The volumetric activity measurements were recorded automatically for at least 15 days with a 1-hour interval. The radiometer simultaneously recorded radon concentration, temperature, relative humidity, and barometric pressure in the chamber air. The information on radon concentration in the container was recorded using a PC and AlphaEXPERT software.

The change in the concentration of radon in the air of the sealed container can be described as [9]:

$$\frac{dC}{dt} = \frac{G}{V} - \lambda \cdot C \tag{2}$$

where G is an emanation rate of radon from a sample, Bq/sec; V is a container volume, m^3; C is *a* concentration of radon, Bq/m^3*;* λ is a radon disintegration constant, 0.00755 h^{-1}; t is exposure time, hrs.

The solution of the equation (2) is:

$$C(t) = \frac{G}{\lambda \cdot V} \cdot \left(1 - e^{-\lambda \cdot t}\right) + C_0 \cdot e^{-\lambda \cdot t} \tag{3}$$

where C_0 is an initial concentration of radon, Bq/sec.

Formula 3 can be given as:

$$C(t) = C_{\max} \cdot \left(1 - e^{-\lambda \cdot t}\right) + C_0 \cdot e^{-\lambda \cdot t} \tag{4}$$

where $C_{\max}$ is a maximum concentration of radon in the air of the container, Bq/m^3.

The emanation coefficient during the experiment was determined as:

$$E(t) = \frac{C(t) \cdot V}{\left(1 - e^{-\lambda \cdot t}\right) \cdot M \cdot A_{Ra}} \tag{5}$$

where $C(t)$ is a current concentration of radon, Bq/m^3; V is free volume inside of the container, m^3; t is exposure time, hrs; M is a sample mass, kg; A_{Ra} is a radium specific activity, Bq/kg.

The emanation was calculated by the formula in [21]:

$$E = \frac{C_{Rn} \cdot V}{A_{Ra} \cdot M} \tag{6}$$

where C_{Rn} is an equilibrium concentration of radon, Bq/m^3, in the sealed container.

At the end of the experiment, the sorption moisture of the samples was determined as:

$$W_s = \frac{m_s - m_d}{m_d} \tag{7}$$

In this formula, m_d and m_s are masses of the samples in dry and in equilibrium states with maximum air humidity achieved during the experiment. We determined open porosity + by sequential moistening, immersing the sample in water, first at 1/3, then at 2/3 of its height. At the last third stage, the water level exceeded the top surface of the samples by 50 mm. The duration of the last stage was determined by the time at which the results of two consecutive weightings differed by no more than 0.1 %.

The value of total water saturation was calculated as:

$$W_f = \frac{m_f - m_d}{m_d} \qquad (8)$$

The degree of filling of open pores with water was determined by:

$$\eta = \frac{W_s}{W_f} = \frac{m_s - m_d}{m_f - m_d} \qquad (9)$$

In these formulae, m_f is a mass of the samples in a water-saturated state, kg.

3. Results and Discussion

Following the results of gamma-ray spectrometry (Table 1), we obtained the average specific activities of radionuclides of radium, thorium and potassium, and calculated the specific effective activity using formula (1).

Table 1. The results of gamma-ray spectrometry.

^{226}Ra, Bq/kg	^{232}Th, Bq/kg	^{40}K, Bq/kg	A_{eff}, Bq/kg
19.1±0.92	20.8±0.03	338±15.5	76.6±1.67

The rated value of the specific effective activity did not exceed the maximum permissible value for the materials used in construction and restoration of residential and public buildings set at the level of 370 Bq/kg [20].

Fig. 2 shows a representative curve of growth of the radon concentration in the air of the sealed container. As it can be seen in the flowchart below, after 150–200 hours from the beginning of the experiment, the air relative humidity (RH) in the container becomes constant, which indicates a frozen equilibrium between the sorption moisture content of the samples and the air relative humidity.

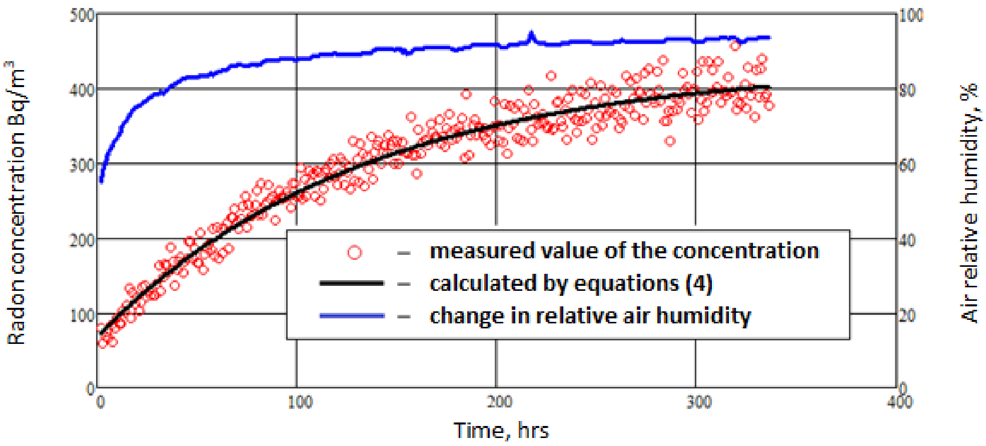

Figure 2. Accumulation of radon and change in air relative humidity in the sealed container.

The statistical analysis of the parameters which was obtained by the least square method is given in Table 2.

Table 2. Analysis of parameters C_{max} and C_0.

Analysis parameter		Standard error, %	Ranges of limits values at 95% probability	
Symbol	Value, Bq/m³		Lower, Bq/m³	Upper, Bq/m³
C_{max}	432	1.82	428	435
C_0	67.9	2.84	62.3	73.5

Fig. 3 shows the result of calculating the emanation coefficient by equation (5) during 330-hour exposure. It is clearly seen that after 200–250 hours the ratio calculated by equation (5) becomes constant, i.e. we observe equilibrium between the activity of radon in the air of the sealed container and the activity of radon in the samples being studied.

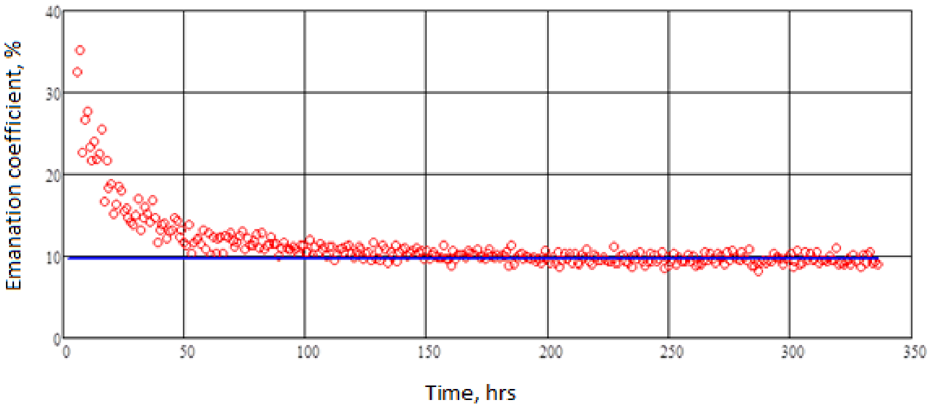

Figure 3. The analysis of emanating ability using equation (5).

The value of the emanation coefficient was respectively obtained for a series of experiments with different moisture contents of the test sample.

Fig. 4 presents the approximating curves and their analytical expressions, obtained and discussed below, as the best. The data clearly indicate that with an increase in the relative humidity of the air, the emanation coefficient increases by almost 2 times, however, in contrast to the value of sorption moisture content, which increases with the increasing *RH*, the rate of emanation increase declines.

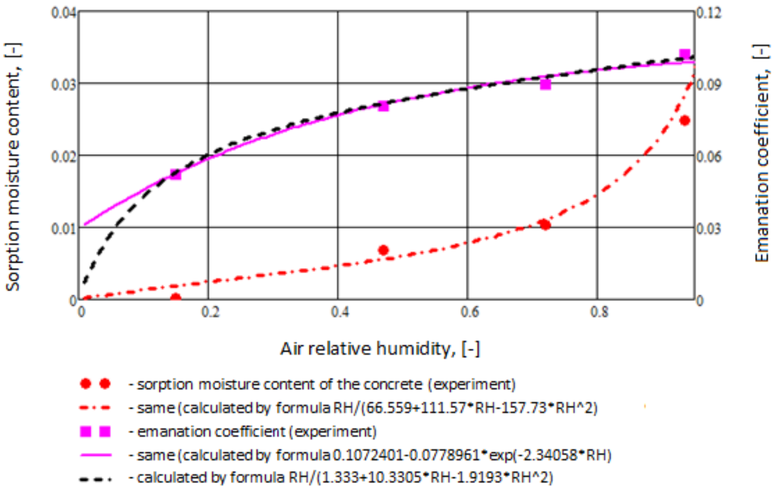

Figure 4. Change in the emanation coefficient of heavy concrete and its sorption moisture content in the stage of desorption depending on air relative humidity.

An analysis of the curves allows concluding that there is a close correlation between the processes of sorption-desorption of water vapor by the studied samples of semi-dry pressed heavy concrete and the emanation coefficient.

There are many semi-empirical models for calculating the equilibrium sorption moisture content of inorganic, organic, and biological materials depending on the air relative humidity and ambient temperature, for example, 15 models were analyzed in [22], the authors of [23] performed analysis for 24 models describing the sorption process for ceramic and silicate brick, autoclaved aerated concrete, cement-lime mortar, ordinary cement mortar and mortar modified with polypropylene fibres, to define the sorption model that most accurately describes the isotherms. The results of a similar work for wood for 27 mathematical models are given in [24].

For practical purposes, any dependencies that best and most reliably describe experimentally observed values for a particular material are used for engineering analysis, and at the same time, based on the obtained graphic dependence, one of the six types of sorption isotherms accepted by the International Union of Pure and Applied Chemistry (IUPAC) is taken as the basis [25], paying attention to the behaviour of the curves in the entire range of humidity and, especially, under the humidity being close to maximum.

The change in emanation coefficient (E) and sorption moisture (Ws) depending on the relative humidity of the ambient air (RH) can be described with an appropriate degree of accuracy by formulae (11), (12) and (13).

The Hailwood – Horrobin (HH) model or the Dent model is most consistent with the experimental data on determining the processes of sorption–desorption of water vapour by wood-based materials [24].

$$W_s = \frac{RH}{A + B \cdot RH - C \cdot RH^2} \tag{11}$$

The team at the Technical University of Denmark has compiled a catalogue of sorption isotherms for more than a hundred materials used in construction [26], however, only one mathematical model was used to describe sorption and desorption.

$$W_s = A \cdot \exp\left(\left(-\frac{1}{B}\right) \cdot \ln\left(1 - \frac{\ln(RH)}{C}\right)\right) \tag{12}$$

Here in this formula: A is a maximum hygroscopic content; RH is relative humidity; B and C are empirical coefficients.

A promising model for cement concretes and mortars is the model in [23, 27, 28]:

$$W_s = \frac{A \cdot RH}{(1 + B \cdot RH) \cdot (1 - C \cdot RH)} \tag{13}$$

Table 3 presents the results of the analysis using expressions (11–13).

Table 3. The values of the coefficients in the equations (11), (12), (13) and the correlation of the rated and experimental values (R^2).

Parameters	A	B	C	R^2	Formula
Sorption moisture, W_S	66.559	111.57	-157.73	0.9823	(11)
Same	0.047	0.961	0.107	0.9780	(12)
Same	1.5	2.592	0.915	0.9823	(13)
Emanation coefficient, E	1.333	10.331	-1.919	0.9881	(11)
Same	0.1033	0.077	36.83	0.9777	(12)
Same	0.75	7.93	0.182	0.9881	(13)

As can be seen from the table, equations (11), (12) and (13) describe the process of sorption of concrete and changes in emanating within the range of RH from 0.18 to 0.935 with a high degree of pair correlation (R^2) which may indicate an interconnection of these processes. However, all of them are compromised by the calculated value of E = 0 at RH = 0. A model free of this remark can be the one which takes into account the emanation coefficient values of dry materials.

The following expressions are proposed to describe the functional dependence of emanation coefficient on the relative humidity of the ambient air and the moisture content of the studied samples:

$$E = E_d + (E_{ms} - E_d) \cdot (1 - \exp(-k \cdot v)) \tag{14}$$

Or

$$E = E_{ms} - \left(E_{ms} - E_d\right) \cdot \exp\left(-k \cdot v\right) \quad (15)$$

where E_d and E_{ms} are the emanation coefficient of the samples correspondingly in a dry state and in a maximum saturated-by-sorption state at $RH \to 100$ % (RH = 93.5% in the experiment); k is empirical constant; v is a parameter numerically equal to the characteristic of the ambient humidity or the moisture equilibrium state of the material, for example, relative humidity RH, moisture content (by weight or volume) or degree of filling of pores with adsorbed water η.

When we substituted the values of relative humidity into formulae (14) and (15), coefficient of determination R^2 = 0.9796 turned out to be somewhat lower than for (11), (12) and (13). At the relative humidity RH = 0, however, $E \neq 0$.

An imperial formula for calculating an emanation coefficient for soils was proposed by [29]:

$$E = E_0 \cdot \left[1 + k_1 \cdot \left(1 - \exp\left(-k_2 \cdot s\right)\right)\right] \quad (16)$$

where: E_0 is emanation coefficient in the dry state; s is soil moisture, k_1 and k_2 are empirical coefficients.

For clay, silt and sand, the values of k_1 and k_2 vary within a fairly narrow range – respectively from 1.53 to 1.85 and from 18.8 to 21.8. In a more recent work [21], formula (16) was used to calculate radon flux density from soil at k_1 = 1.85 and k_2 = 18.8.

Within a complex process of emanating, caused, on the one hand, by an inhibiting ability of the adsorbed water layer on the emanating surface, and on the other hand, facilitating the movement of radon atoms due to levelling the uneven inner surface of pores and capillaries, it seems relevant to determine the dependence of emanation on the degree of filling the pores with water $E(\eta)$. The results of experimental data and calculations are given in Fig. 5.

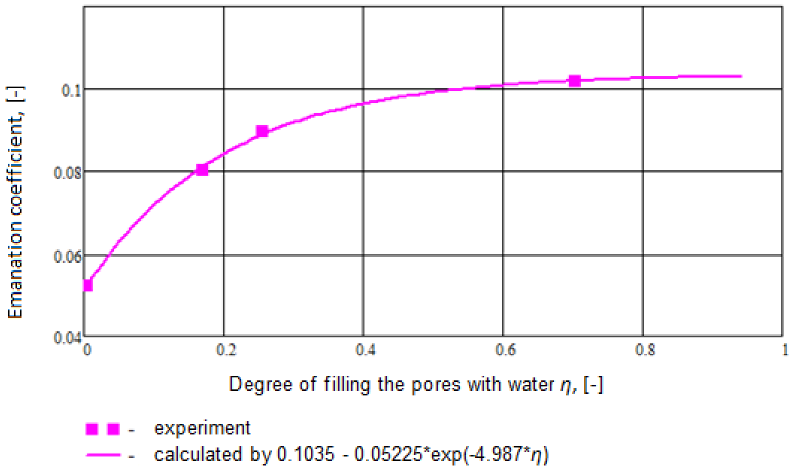

Figure 5. Effect of the degree of pores filling with water on the value of the emanation coefficient.

As we can see, a functional dependence of the change in the emanation of semi-dry pressed concrete and the degree of filling of open pores with water $E(\eta)$ can be represented by expressions similar to (14) and (15), but much more accurately, with R^2 = 0.9992.

A statistical analysis of the values of the coefficients calculated by the least square method is given in Table 4.

Table 4. The values of the coefficients in equations (14) and (15).

Indicators	Value
Dry emanation E_d	0.051±0.001
$E_{ms} - E_d$	0.052±0.002
Empirical coefficient k	4.99±0.34
Emanating at maximum sorption moistening E_{ms}	0.1035±0.0013

As can be seen from Table 4, the change in the emanation of semi-dry pressed heavy concrete is characterized by a coefficient value k = 4.99. The approximation of the experimental data by formulas (14) and (15) allows predicting the values of E_{ms} almost in the entire range of change in the relative humidity of the air where building structures enveloping rooms are used.

4. Conclusion

In contrast to the majority of previously published studies, where the analysis of the emanation process was performed on samples of rocks or soils, this work provides the results of the analysis of emanation of artificial building material.

1. We found a close correlation of the emanation coefficient of semi-dry pressed heavy concrete, the humidity of the ambient air, and the equilibrium sorption moisture of the material itself in the stage of desorption.

2. Empirical calculation formulas have been proposed for determining emanation coefficient depending on the air relative humidity and the moisture content of heavy concrete. The densest correlation is observed when determining the design parameter depending on the degree of filling of open pores, the value of which is estimated by the method of sequential immersion of the samples.

3. With an increase in the degree of filling of open pores with water from 0 to 70 %, the emanation coefficient increases by almost 2 times in the exponential dependence. In the range of relative humidity typical for residential and public buildings (20 ... 60 %), we observed almost 50 % increase of the emanation coefficient, which necessitates the numerical control of this parameter in order to accurately assess the dose load on the population.

Measurement of emanation coefficient of materials included in enveloping structures and interior finishings should be standardized taking into account moisture and age of samples.

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