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Control of radon emanation at determination of activity concentration index for building materials

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HIGHLIGHTS

- No need to seal the sample when determining activity concentration index of building materials.
- Rapid and precise methods to determine radium specific activity C_{Ra} and emanation coefficient k.
- The improved method for controlling radon leakage μ from the measuring container.
- The dependence of the underestimation of C_{Ra} on k and μ is given.
- For reliable sealing the sample, a standard polyterephthalate flask is recommended.

ARTICLE INFO

ABSTRACT

Radon emanation is a well-known source of uncertainty at determination of activity concentration index for building materials and NORM. To reduce this uncertainty both radon emanation coefficient of the sample material and radon leakage from the measuring container should be considered. Two different methods (the rapid method and precise method, which requires to keep the sample sealed for at least three weeks), which are based on the use of only gamma-ray spectrometer, are proposed for testing radon emanation. Both methods also allow simultaneous determination of the specific activity of radium and an assessment of radon exhalation rate from the surface of the finished building products. In addition, the method for determination of radon leakage from the container is proposed. The reliability of the sealing methods of containers is compared and standard PET-flasks with a high radon-tightness are offered. The criteria to control radon leakage and radon emanation coefficient are given for three types of test samples: (a) building materials, (b) natural materials of mineral origin (sands, clays, rocks etc.), and (c) artificial materials, such as absorbents, filters, radioactive wastes. A simplified screening test to control activity concentration index for building materials without a need to keep the sample sealed before the measurement is proposed.

1. Introduction

The activity concentration index (ACI) is one of the main characteristics of the radiation quality of building materials and NORM (Naturally Occurring Radioactive Materials), the value of which is limited, according to Council [1]. The quantity ACI is determined by the following equation [10]:

$$I = C_{Ra}/300 + C_{Th}/200 + C_K/3000, \tag{1}$$

where C_{Ra}, C_{Th} and C_K are specific activities of ^{226}Ra, ^{232}Th and ^{40}K, respectively, in Bq kg^{-1}.

The measurements of specific activity (or activity concentration) of ^{226}Ra, ^{232}Th and ^{40}K are usually conducted using gamma-ray spectrometers. It is important to clarify that measurements of the specific activity of ^{226}Ra and ^{232}Th are performed by detecting the gamma-rays of the short-lived progeny of radon (^{222}Rn) and thoron (^{220}Rn), respectively, under the condition that a radioactive equilibrium in the relevant natural decay chains is provided. To ensure this important condition, the samples must be stored before the measurement in a hermetically sealed state for at least 3 weeks to prevent leakage of radon isotopes. Otherwise,

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radon isotopes' release from the sample results in uncontrolled underestimation of the measured activity concentration index. For example, the underestimation of radium activity concentration is 10–40%, that approximately corresponds to a typical range of radon emanation coefficient for building materials and most types of soils [8,11,7,3]. In case of highly radon-emanating materials, such as finely dispersed rock of clay type with a radon emanation coefficient of more than 40% and even reaching 70% [11,3], the measurement result of radium activity concentration may be underestimated by 2–3 times. And even greater underestimation of the measurement result, exceeding the order of magnitude, can be observed in the case of testing artificial adsorbents. For example, we have samples of quartz sand used for water purification at the treatment plant in Ryazan (Russia) for many years, in which the emanation coefficients of radon and thoron reach extremely high values – of 96% and 48%, respectively.

The measurement problem is caused by the fact that reliable radon-tight reusable measuring containers are lacking. Moreover, there is still no justified criterion of allowable radon leakage at sealing of the samples. Currently the standard tests include the determination of the tightness of the sealed measurement beaker [2] by using a source of extremely high radon activity. In our opinion, this test and applied criteria are complicated and can be supplemented or replaced by a more available test. Moreover, including this complex test in normative part of the future standard for measurements of ACI would seriously limit its application, because only a few gamma laboratories have the required resources to carry out the standard test. Obviously, the ultimative solution for this problem would be to make a standard sealed beaker, which can be used multiple times.

According to [13], the worldwide average values of natural activity concentrations in soil are: 32 (1000) Bq/kg for ^{226}Ra, 45 (3 6 0) Bq/kg for ^{232}Th and 412 (3200) Bq/kg for ^{40}K (the maximum measured values are shown in parentheses). Consequently, for the most building materials of the mineral origin, such as concrete, ceramics, gypsum, natural stone etc., the average ACI level, according to (1), is less than a half of the reference value. For materials used in bulk amounts ACI should not exceed 1 that corresponds to an annual effective dose for building occupants of less than 1 mSv y^{-1} Council [1].

If the activity concentration index in the material is low, it has no sense to seal and store a sample for a long time, because the ACI-value would be obviously low as well. However, in other cases both hermetical sealing and storing the samples for a period of at least three weeks before the test are needed. In this regard, it would be appropriate to develop a quantitative criterion of the need to seal and keep the samples, if the ACI is relatively low. In case of samples with a high radon emanation, the ACI can be determined in a rapid mode based on periodic measurements of accumulating activity after sealing the sample using only a gamma-ray spectrometer. However, an algorithm of the evaluation of the result uncertainty does not exist and has to be developed. In addition, thoron emanation should be considered.

This research will contribute to solve the above-listed problems, and will offer the most optimal methods and approaches to account for radon emanation and leakage at controlling the activity concentration index for building materials and NORM.

2. Theoretical background

Radon emanation from the test sample is a source of uncertainty at determining the activity concentration index for building materials by the formula (1). To reduce this uncertainty of ACI the radon release from the measuring container into the environment should be considered. This radon release, besides the radium activity concentration, depends on the two factors: (a) coefficient of radon emanation from the sample material and (b) radon leakage from the measuring container. The process of radon transport from a sample does not play a role, since all the produced free radon is released from the test sample, because it is in a crushed/granular form.

Radon emanation coefficient k (rel) is determined through the following equations [10]:

$$k = C_{Rn}/C_{Ra} = 1 - C_{Ra}^*/C_{Ra}, C_{Ra} \geqslant C_{Ra}^*, \qquad (2)$$

where

C_{Rn} – specific activity of free radon in the material (no flow of radon), Bqkg^{-1};

C_{Ra}– specific activity of Ra-226 in the material ($C_{Ra} = C_{Rn}+ C_{Ra}^*$), Bqkg^{-1};

C_{Ra}^*– specific activity of bound radon in the material, Bqkg^{-1}.

If the sample material is crushed (or its dimensions do not exceed a few centimeters) and stored in a dispersed state outside of the container (under room conditions) at least for a few hours, it contains only bound radon, because free radon is removed from the sample naturally by diffusion. In this case, we consider that the sample is in "deemanated" state.

After sealing of the container with the deemanated sample there is an increase of the total radon concentration $C(t)$ increases due to the accumulation of free radon $C_{Rn}(t)$, according to Fig. 1 and equation

$$C(t) = C_{Ra}^* + C_{Rn}(t). \qquad (3)$$

The function of free radon accumulation $C_{Rn}(t)$ is determined by solving the differential equation, which describes the process of accumulation, decay and leakage of free radon:

$$\frac{\partial C_{Rn}(t)}{\partial t} = \lambda \cdot k \cdot C_{Ra} - \lambda \cdot C_{Rn}(t) \cdot (1 + \mu), \qquad (4)$$

where

λ – the radon decay constant equal 2.09·10^{-6} s^{-1} (or 0.00755 h^{-1});

μ – the radon leakage from the container expressed in parts of λ, rel.

It is important to note that often in the literature, for example [6], the parameter of ($\mu \cdot \lambda$) instead of the radon leakage rate is defined as the air exchange rate in the container. However, such
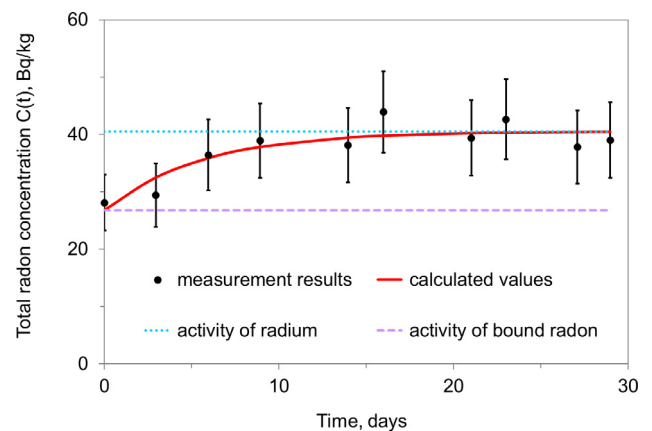

Fig. 1. The example of radon accumulation in the initially deemanated and then sealed sample with the following characteristics: C_{Ra}^* = 26.8 ± 0.7 Bq/kg, C_{Ra} = 40.9 ± 1.3 Bq/kg, k = 0.34 ± 0.09.

a definition is wrong, since air exchange with the external medium does not occur in a sealed container (including measuring devices connected in a closed loop). In the sealed container, the leakage of activity occurs solely due to the radon diffusion through the material of seal and/or container walls (and/or connecting tubes, etc.). In addition, we would like to draw attention to the fact that radon is an inert gas; therefore, it possesses much more leakage than the basic components of air (nitrogen and oxygen). In this regard, the high air-tightness of the container is not sufficient for the radon-tightness.

The Eq. (3), considering (2) and the solution (4), is transformed into

$$C(t) = C_{Ra}^* \cdot \left[1 + \frac{k}{(1-k) \cdot (1+\mu)} \cdot \left(1 - e^{-(1+\mu) \cdot \lambda \cdot t} \right) \right] \quad (5)$$

According to Fig. 1 and formula (5), in the first measurement (before sealing) of deemanated sample $C(t=0) = C_{Ra}^*$, and after storage in the sealed state $C(t \to \infty) = C_{Ra}^*/(1-k) = C_{Ra}$. Thus, without a need in prolonged sealed storage of the sample (for at least 3 weeks) the values of k and C_{Ra} can be easily determined from the results of periodic measurements of the C_j-values in the initial storage period of the sealed container (e.g., during the first 4–10 days), using a gamma-ray spectrometer. In this case the value of C_j, which is usually the measurement result of ^{226}Ra by gamma-ray spectrometers, is the specific activity of gamma-emitting short-lived daughter products of free and bound radon (^{214}Pb and ^{214}Bi), which is equal to the total radon activity concentration in the sample at the time of measurement.

In this method (called in further **Method 1**) determination of the values of k and C_{Ra}^* is performed in the rapid mode based on the approximation of the measured C_j-values by the calculated function $C(t)$ in the period of accumulation of a free radon in the sample (Fig. 1), if the number of measurements (j) is not less than 3, which is a minimum number of measurements. The approximation is performed by the method of least squares - searching the minimum values of the function:

$$F = \sum_j [C_j - C(t_j)]^2. \quad (6)$$

Minimum values of the function F correspond to the points of its extremum, which are the desired values of k and C_{Ra}^*, obtained from the solution of the system with two differential equations:

$$\begin{cases} \frac{\partial F}{\partial k} = 0 \\ \frac{\partial F}{\partial C_{Ra}^*} = 0 \end{cases}. \quad (7)$$

Annex 1 contains more detailed algorithm for calculating the values of k and C_{Ra}.

In the Method 1, the uncertainty of the k-value is determined by the formula

$$\Delta k = \sqrt{\sum_{j=1}^n \left(\frac{\partial F_k}{\partial C_j} \right)^2 \cdot (\Delta C_j)^2}, \quad (8)$$

where

F_k – the function expressing the value of k, obtained from the solution of the system (7);
ΔC_j – the uncertainty of the j-th measurement result of radon activity concentration in the sample, Bq kg^{-1}.

Besides, the value of Δk can be determined using the simulation based on the estimation of the range of k-values, at which the calculated function of $C(t)$ does not exceed the aggregate confidence intervals of all the measured values of C_j.

The following steps improve the accuracy of Method 1:

(a) increase the number of measurements in a gamma-ray spectrometer during the accumulation of radon activity, and/or
(b) increase the duration of the specific activity measurement, especially making the first measurement longer (or by repeating the measurements of the sample in its deemanated state).

It should be taken into account that the accuracy of the first measurement result significantly affects the uncertainty of radon emanation coefficient in the sample material.

An alternative method of determining the radon emanation coefficient (called in further as **Method 2**) is the accumulation of radon until reaching the radioactive equilibrium with radium. This method requires executing two series of the measurements of the sample activity by also a gamma-ray spectrometer. In the first series the periodic measurements of the sample in its deemanated state are carried out (the sample is loaded into the container, immediately measured and unloaded; the next measurement is carried out not earlier than one day, etc.). Then, after storing the sample in a sealed state at least for three weeks the second series of measurements is performed. In each of these two series at least 3 measurements are carried out.

In Method 2 the value of k is determined by formula (2), wherein

$$C_{Ra}^* = \frac{1}{n} \cdot \sum_{i=1}^n C_{Ra}^*(i) \text{ and } C_{Ra} = \frac{1}{m} \cdot \sum_{j=1}^m C_{Ra}(j), \quad (9)$$

where

$C_{Ra}^*(i)$ – the result of the i-measurement in the first series, Bq kg^{-1};
n – the number of measurements in the first series;
$C_{Ra}(j)$ – the result of the j-measurement in the second series, Bq kg^{-1};
m – the number of measurements in the second series.

The value of k is accepted zero, if the result of the calculation by the formula (2) is negative.

In Method 2 the uncertainty of radon emanation coefficient Δk (rel) is determined in accordance with the recommendations [4] by the following formula:

$$\Delta k = k_c \cdot \frac{C_{Ra}^*}{C_{Ra}} \cdot \sqrt{ \left(\frac{\Delta C_{Ra}^*}{C_{Ra}^*} \right)^2 + \left(\frac{\Delta C_{Ra}}{C_{Ra}} \right)^2 + \delta_{fr}^2 }, \quad (10)$$

where $\Delta C_{Ra}^* = \sqrt{ \dfrac{\sum_{i=1}^n [C_{Ra}^* - C_{Ra}^*(i)]^2}{n \cdot (n-1)} }$,

$$\Delta C_{Ra} = \sqrt{ \frac{\sum_{j=1}^m [C_{Ra} - C_{Ra}(j)]^2}{m \cdot (m-1)} } \quad (11)$$

k_c – the coverage factor equal to 2 (provided the value of the confidence level of result is at least 95%);
δ_{fr} – the relative uncertainty caused by the loss of free radon, which is equal to 0.01.

To improve the accuracy of Method 2 the following steps could be undertaken:

(a) increase the number of the measurements in a gamma-ray spectrometer up to 5–7 in each (or at least one) of the two series, and/or

Fig. 2. The samples of phosphogypsum in deemanated state.

(b) increase the duration of sample specific activity measurement up to 2 h.

The Method 2 can be also used to determine the thoron emanation coefficient, but the sample of the test material in this case must be crushed to a powder consistency. Deemanation of such sample is carried out by keeping a powder at a layer not thicker than 2 mm during at least two days. The second series of measurements is performed in at least two days after sealing the sample (or after the accumulation of ^{222}Rn).

3. Equipment and materials

A homogeneous phosphogypsum with a high specific activity of radium, but a very low content of thorium and potassium, served as an experimental material. The samples of phosphogypsum in deemanated state are shown in Fig. 2.

Three different 1-liter Marinelli beakers and five identical 125-mL flasks served as measuring containers, as shown in Fig. 3.

R – the prototype of special radon-tight Marinelli beaker;
BS – the Marinelli beaker commonly used in Israel and European countries;
LA – the Marinelli beaker of the type LA-130G-E (GA-MA & ASSOCIATES Inc. USA);
F – the standard of PET-flask (125 mL) made of polyethylene terephthalate, according to Specification TU 9464–007-95202676-2011 (Russia), with a one-component lid of the type 28 BPF used in the pharmaceutical industry (μ < 0.03).

The reliability of sealing in the "R" container is provided by a rubber gasket in the lid.

For sealing of the "BS" container, a wide insulating tape of the Scotch type was used. The container was tightly and thoroughly glued by three layers (without wrinkles) along the perimeter of the joint between the container and the lid.

The "LA" container was closed by the plastic lid without additional sealing measures, as is.

The flasks "F" provide a very reliable sealing because of the radon leakage not exceeding 3% with respect to radon decay constant [9]. Determination of radon leakage from "F" containers was carried out experimentally, according to Annex 2.
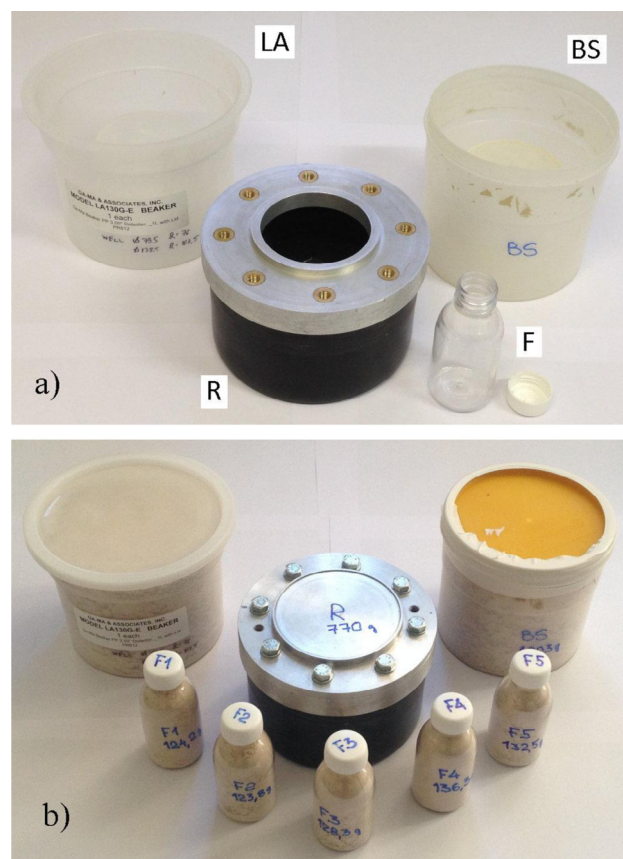

Fig. 3. Type of measuring containers in (a) open and (b) close conditions with the samples of phosphogypsum.

Measurements of the specific activity of natural radionuclides (^{40}K, ^{226}Ra and ^{232}Th) in the samples were performed in the scintillation gamma-ray spectrometer with 2.5 × 2.5″ NaI(Tl) crystal, protected by the 50-mm lead shielding (Scientific and Producing company "DOZA", Russia).

The gamma-ray spectrometer was operated by the special software "Progress 5.1".

Fig. 4. Location of the flask on the detector of gamma-ray spectrometer.

Fig. 5. The example of measurement and calculation (using Method 1) of free radon build up in the initially deemanated and then sealed "F1" sample.

The calibration of the gamma-ray spectrometer was conducted only for "Marinelli" geometry with a special set of the granular measures of the specific activity of ^{40}K, ^{226}Ra and ^{232}Th (separately) using the measuring "R" container. It should be noted that the quality of the gamma-ray spectrometer calibration does not really matter, because the radon emanation coefficient k is a dimensionless quantity. Therefore, the measured values of the specific activities (including ^{226}Ra) could differ depending on the used measuring container, but the results of the processing of all the measurements was carried out only at one geometry – "Marinelli", including the "F" flasks, placed always on the detector in the certain position, as shown in Fig. 4.

4. Measurements and results

In the first stage of the study, the phosphogypsum samples were measured in deemanated state. For this purpose each sample of the crushed phosphogypsum was kept for about a day before the measurement in a dispersed state by a thin layer under room climatic conditions, as shown in Fig. 2. After the measurement in the appropriate container, the sample was removed and placed again by a thin layer. At this stage, three measurements with each of the 8 samples were conducted. The measurement duration of one sample was always 30 min at this and following stages.

After the third measurement in deemanated state the measuring container with the sample was closed (tightly as much as possible) by the lid, as shown in Fig. 3(b), for keeping the sample in a sealed state.

In the second stage of the study periodical measurements of the samples in the sealed state were carried out in the period from 3 to 7 days after their sealing. At this period, five measurements with each of the 8 samples were conducted.

The final measurements of the closed samples were conducted in the period from 25 to 26 days after their sealing, i.e. in the state of almost complete accumulation of free radon in the sealed sample. At this period, 3 measurements with each of 8 samples were carried out.

The results of the measurements in the gamma-ray spectrometer and processing experimental data are given in Fig. 5 and Table 1.

Table 1 shows the values of radon emanation coefficient for each of the test samples obtained by the two methods without considering the radon leakage. According to Table 1, the actual value of the radon emanation coefficient in phosphogypsum was
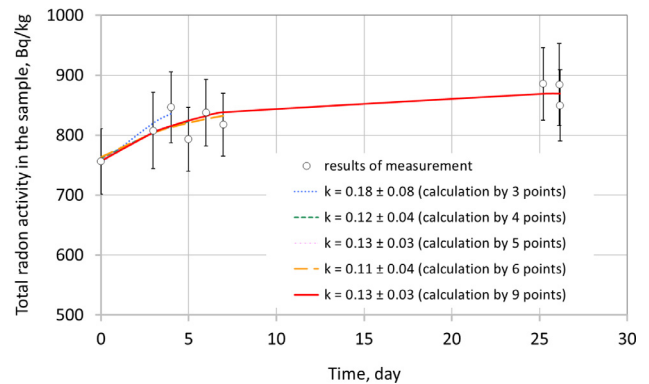
in the range of **0.14 ± 0.02**. This value was obtained using the sealed "F" flask containers.

According formula (2) and measured results by the gamma-ray spectrometer in geometry of "R" container the specific activity of ^{226}Ra in phosphogypsum (C_{Ra}) is **920 Bq kg^{-1}** at C_{Ra}^{*} = 790 Bq kg^{-1} and k = 0.14.

In addition, Table 1 shows the values of radon leakage from Marinelli-type containers, calculated using Eq. (5) at $t \rightarrow \infty$ and k = 0.14, and measured results of radium specific activity. In addition, radon leakage can be determined in accordance with Annex 2, as well as by formula (12).

$$\mu = \frac{k_F \cdot (1 - k_M)}{k_M \cdot (1 - k_F)} - 1, \tag{12}$$

where

k_M – the radon emanation coefficients determined by Marinelli-type container without considering the radon leakage, rel;

k_F– the average radon emanation coefficients determined by flask containers with a high radon-tightness (according Table 1, k_F =0.14) or a well-known value of the emanation coefficient in the reference material.

Formula (12) is obtained from Eq. (5) under the condition $t \rightarrow \infty$ also, at μ = 0 for Marinelli-type container, but using μ in another equation (where k_F is the actual value) of the system with the same values of C_{Ra} and C_{Ra}^{*} in both equations.

5. Discussion

Considering the small volume and mass of the phosphogypsum samples in the "F" flask (on average equal to 0.13 kg), we found that radium activity recalculated for the bigger sample in "Marinelli" container of 1 L, was approximately equivalent to the specific activity C_{Ra} = 120 Bq kg^{-1}. This value brings the activity concentration index closer to the threshold value, according to (1). At the level of such activity it is optimal to determine the most effective strategy to accelerate the determination of k and C_{Ra}, using the Table 1 data related only to the flasks. Analysis of these data shows that performing from 6 to 8 periodic measurements (including 3 measurements in deemanated state) during the first week after the sealing of the sample would be most optimum when using Method 1. It should be noted that the uncertainties of Methods 1 and 2 do not differ much, if the sample is also kept sealed for at least 20 days in Method 1.

From Table 1 it can be seen that the "LA" container has a high radon leakage (μ = 4 since the calculated value of k is very low),

Table 1
Radon emanation coefficients in the tested samples obtained by two methods without considering radon leakage, and calculated values of radon leakage from Marinelli-type containers

Number of measurements for Method 1 (duration of sealed keeping)	Sample code (mass, kg)								
	Flasks						Marinelli		
	F1 (0.124)	F2 (0.124)	F3 (0.128)	F4 (0.136)	F5 (0.133)	F(1-5)[**] (0.129)	BS (1.003)	R (0.770)	LA (1.234)
3+2[*] (4 day)	0.18±0.08	0.21±0.06	0.17±0.04	0.10±0.08	0.10±0.08	0.15±0.05	0.11±0.04	0.09±0.04	–
4+2[*] (5 day)	0.12±0.04	0.17±0.04	0.14±0.03	0.15±0.04	0.14±0.06	0.14±0.01	0.13±0.04	0.09±0.03	–
5+2[*] (6 day)	0.13±0.03	0.13±0.03	0.10±0.02	0.14±0.03	0.12±0.04	0.13±0.01	0.12±0.04	0.09±0.02	–
6+2[*] (7 day)	0.11±0.04	0.11±0.03	0.11±0.02	0.16±0.03	0.14±0.03	0.12±0.02	0.12±0.04	0.08±0.02	–
7+2[*] (25 day)	0.13±0.03	0.11±0.03	0.14±0.02	0.16±0.03	0.17±0.03	0.14±0.01	0.13±0.04	0.10±0.01	–
Method 2 (6 measurements – 3 before and 3 after sealing)	0.13±0.05	0.14±0.04	0.14±0.05	0.15±0.04	0.15±0.08	0.14±0.03	0.13±0.02	0.10±0.03	0.03±0.02
Radon leakage (μ)	**Less than 0.03**						**0.13**	**0.45**	**4**

[*] Additional measurements of the sample in deemanated state.
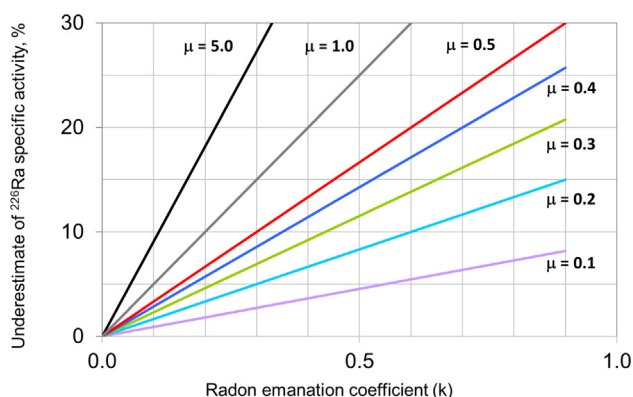[**] Average values for 5 flask samples.

Fig. 6. The dependence of the underestimate of radium activity concentration in the tested sample on radon emanation coefficient of the sample material and radon leakage from the test specimen container (in fractions of the radon decay constant).

so it cannot be uses for keeping the samples without special measures to ensure the tightness of the connection with the lid.

The "BS" container, on the contrary, has good tightness (μ = 0.13) at fairly simple way to insulate the joint with the lid using an adhesive tape (see more detailed description in the Section 3).

The "R" container has a noticeable radon leakage (μ = 0.45), accordingly the value of radon emanation coefficient (k_R = 0.10 by Table 1) understated by 40%, despite the complexity of the design and the seeming reliability.

However, despite the relatively high radon leakage from the "R" container, the measured value of the specific activity of ^{226}Ra in phosphogypsum (C_{Ra} = 880 Bq kg^{-1} without considering leakage of radon) using this "R" container is underestimated by not more than 5% (the actual value C_{Ra} = 920 Bq kg^{-1}) – due to a relatively low value of k in phosphogypsum. In this connection, it is worth

to analyze the dependence of the relative understatement (δ, %) of the measured specific activity of Ra-226 on radon emanation coefficient of the sample material and on radon leakage from the measuring container. This dependence is shown in Fig. 6 in the form of nomogram, which is built taking into account (5) based on the following relative difference:

$$\delta = 100 \cdot \left[\frac{C(t, k, \mu)}{C(t, k, \mu = 0)} - 1 \right] at\ t \rightarrow \infty. \tag{13}$$

Analysis of the data presented in Fig. 6 shows that even with significant radon leakage (e.g. μ = 0.45, as in the case of "R" container), the understatement of radium content does not exceed 20%, if radon emanation coefficient of the sample material does not exceed 0.6. However, such a very high coefficient of emanation is rarely found in the materials of the mineral origin [11]. For example, radon emanation coefficient for building materials does not exceed 0.3 [8,7]. In this case, according to Fig. 6, the value of δ does not exceed 30%, even if the measuring container with the test sample is left open ($\mu \sim 5$). Therefore, at the radioactivity control of building materials it has no sense to spend a time and efforts to store samples sealed - with the goal to reduce slightly an additional uncertainty caused by leakage and emanation of radon. In this regard, the activity concentration index for any building materials is recommended by test screening using the formula

$$I = 1.3 \cdot C_{Ra}/300 + C_{Th}/200 + C_K/3000, \tag{14}$$

without keeping the sample in a sealed state before measurements by a gamma-ray spectrometer. Here the C_{Ra}, C_{Th} and C_K are the upper limits of the confidence interval of the measured specific activities of ^{226}Ra, ^{232}Th and ^{40}K, respectively, in Bq kg^{-1}. If the result of test screening the building material sample without its sealing exceeds the reference level determined by the formula (14), we recommend keeping the sample sealed and using the formula (1).

If the sample is sealed, then condition δ < 10% must be satisfied, that can be achieved by limiting the value of μ depending on the

Table 2
The limit values of radon leakage (μ) from measuring containers depending on the type of the test of the sample material.

Type of a sample material	Maximum levels of radon leakage (μ)	Maximum value of radon emanation coefficient (k) (if its value for the sample material is unknown)
Building materials	0.5	0.3[*]
Natural soil materials of the mineral origin (sands, clays, loams, rocks etc.)	0.2	0.5[**]
Artificial materials, such as absorbent, filter materials, radioactive waste, etc.	0.1	1.0

[*] [8,7].
[**] [11,3].

emanation coefficient of a test sample material, as shown in Fig. 6. Moreover, the materials of the test samples are expediently ranked into three types, in each of which the radon emanation coefficient does not exceed a given level, according to Table 2.

In the end, we would like to note another important, in our opinion, circumstance. The determination of the radon emanation coefficient in building materials is carried out simultaneously with the measurement of the specific activity of radium using a gamma-ray spectrometer, by Method 1 or Method 2. Both these parameters (k and C_{Ra}) characterize radon release rate of the material and allow to determine the radon exhalation rate from the surface of finished building product, according to ([10], Chapter 3), if its minimum overall dimension does not exceed 0.1–0.2 m (no more than 0.1 m, if radon is released only from one side). In the case of massive building materials, also the calculation can be used, but then it is necessary to take into account the radon diffusion coefficient, which can be determined in the rapid mode no more than 18 h), according to [12,5]). For calculations, it is recommended to use the value of the diffusion coefficient of radon. For example, the value in a concrete slab at a level of $1 \cdot 10^{-7}$ m^2 s^{-1} [12], but not less than $1 \cdot 10^{-8}$ m^2 s^{-1}.

6. Conclusions

1. The activity concentration index of any **building material** is recommended to determine through test screening by approximate formula (14), without a need to keep the sample sealed. For estimation that is more accurate formula (1) is recommended; it is based on the measurements of the sample sealed for at least three weeks ($\mu < 0.5$ is required for the container).
2. During testing **natural soil materials of a mineral origin** (sands, clays, loams, rocks etc.) extremely tight sealing of the test sample is not needed. Quite satisfactory sealing of the measuring container ($\mu < 0.2$) can be easily provided by the wide insulating tape of the scotch type that tightly glued using three layers (without wrinkles) along the perimeter of joint between the container and the lid, as shown in Fig. 3b ("BS" container).
3. In case of testing **artificial materials** ($\mu < 0.1$), such as artificial absorbents, filter materials, radioactive waste etc., the standard "F" flask containers made of polyterephthalate (PET) are recommended for reliable sealing of the test samples. By means of these flasks determination of radon emanation coefficient in the test material with high accuracy, and then calculation of ^{226}Ra specific activity by formula (2) is possible. In this case the calibration of a gamma-ray spectrometer in the flask geometry is not required.
4. The "F" flask made of PET with a lid, which can be closed hermetically, is recommended as a standard reference tool in assessing radon leakage from other measuring containers by formula (12), using a highly emanating homogeneous material. In addition, a simple and clear procedure for determining the radon leakage is given in Annex 2.
5. The radon emanation coefficient and its uncertainty is recommended to determine using rapid Method 1 or more precise Method 2 (by the accumulation of radon in the container until the radioactive equilibrium with radium is reached). The principles of both methods and calculation formulas are given in section "Theoretical background" and Annex 1.
6. Method 1 allows determining radon emanation coefficient and radium specific activity in the test sample with a sufficient accuracy during the first week after sealing the sample. It is especially relevant for testing samples with a high content of radium (100 Bq kg^{-1} or more).

7. Method 2 requires keeping the sample sealed for at least three weeks before starting the test and is more suitable for testing materials with low content of radium.
8. The results of the measurements using both methods allow calculating radon exhalation rate from the surface of finished building materials.

Acknowledgement

Annex 1

Formulas for calculating radon emanation coefficient and specific activity of Ra-226 in the material by Method 1 (at $\mu = 0$)

Radon emanation coefficient (rel) is

$$k = \frac{\alpha_1}{1 + \alpha_1}, \text{ where } \alpha_1 = \frac{\beta_2 \cdot \beta_4 - \beta_1 \cdot \beta_3}{\beta_1 \cdot \beta_5 - \beta_3 \cdot \beta_4},$$

^{226}Ra specific activity in material (Bq kg^{-1}) is

$$C_{Ra} = \frac{C_{Ra}^*}{1 - k}, \text{ where } C_{Ra}^* = \frac{\beta_1 \cdot \beta_5 - \beta_3 \cdot \beta_4}{\beta_2 \cdot \beta_5 - (\beta_3)^2},$$

where

$$\beta_1 = \sum_j C_j, \ \beta_2 = j, \ \beta_3 = \beta_2 - \sum_j e^{-\lambda \cdot t_j},$$

$$\beta_4 = \beta_1 - \sum_j C_j \cdot e^{-\lambda \cdot t_j}, \ \beta_5 = \beta_2 - 2 \cdot \sum_j e^{-\lambda \cdot t_j} + \sum_j e^{-2 \cdot \lambda \cdot t_j},$$

The designations correspond to Section 2.

Annex 2

Determination of radon leakage (μ) from the measurement container

The leakage test is based on quantitative comparison of the results of periodic (daily) measurements of the decreasing activity of radon in a sealed container using a gamma-ray spectrometer. The tests should be performed in the mode of measuring the specific activity of radium for a duration of at least 1 h. At the beginning of the test, a highly active portion of radon is injected into the container just prior to sealing. The first measurement ($j = 1$) should begin no earlier than one day after the injection of radon. For the purpose of more stable distribution of the activity of radon and its progeny inside the container, it is recommended to fill the container with a neutral material, such as quartz sand. During the testing and storage period, the testing container must be protected from direct sunlight and other sources of heat or cold.

Radon leakage μ (dimensionless) from a test specimen container expressed in part of the radon decay constant (λ) is determined by the formula:

$$\mu = \frac{1}{n - 1} \sum_{j=2}^{j=n} \left[\frac{Ln(C_{j-1}/C_j)}{\lambda \cdot (t_j - t_{j-1})} - 1 \right],$$

where

C_j – the result of the j-th measurement on a gamma-ray spectrometer (the activity of the injected radon has to meet the condition: $C_j > 100$ Bq per sample);

t_j – start time of the j-th measurement with an accuracy of one minute;

n – number of measurements ($n > 5$).

It is recommended to test at least 3–5 containers. As a decisive value of radon leakage, the maximum value of the sum $\mu + 2 \cdot \Delta\mu$ (relative to all test containers) should be taken, where $\Delta\mu$ is the standard deviation.

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