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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.

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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]:

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

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, radon isotopes' release from the sample results in uncontrolled underestimation of the measured activity concentration index.

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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]:

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

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

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

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:

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

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 a definition is wrong, since air exchange with the external medium does not occur in a sealed container (including measuring devices

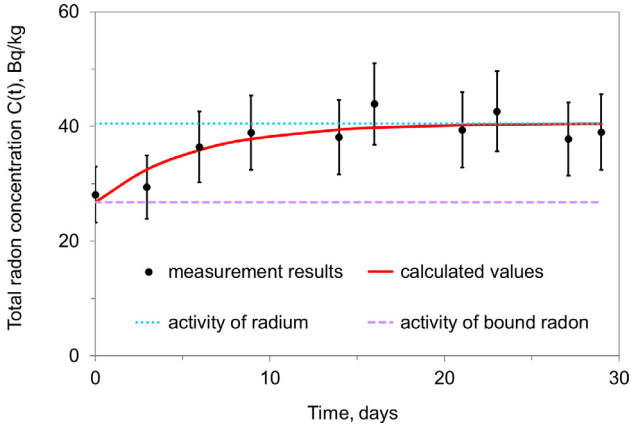

Fig. 1. The example of radon accumulation in the initially deemanated and then sealed sample with the following characteristics: C^*_{Ra} = 26.8 ± 0.7 Bq/kg, C_{Ra} = 40.9 ± 1.3 Bq/kg, k = 0.34 ± 0.09.

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