NUKLEONIKA 2003;48(3):133–136

ORIGINAL PAPER

Determination of exhalation rates through measurements of alpha and beta radiation with the aid of liquid scintillation counter

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Abstract In this paper, a method for the determination of the relative and mass exhalation rates is presented. For the measurement of radon content in an emanation chamber, an α/β liquid scintillation counter was applied. The method was preliminarily tested on the following samples: (1) samples of uranium tailing piles in the Kowary region, (2) samples obtained by mixing of the mentioned waste with various materials such as cement, gypsum and anhydrite and (3) samples of some building materials.

Key words radon • exhalation rates • liquid scintillation counter

Introduction

Radon is a natural radioactive element in the gaseous form. It is a decay product of radium, which is present in all rocks and building materials. Radon escapes from them via diffusion and convection. The effect of radon releasing from solid materials to the pore air is called emanation. The parameters that characterise quantitatively this effect are the emanation coefficient and/or exhalation rate. The emanation coefficient η, expressed as percentage, is defined as a ratio of a number of radon atoms released from grains to the air in the pores to the entire number of radon atoms, produced by the decay of radium in an investigated sample. For many years the geologists have been exploiting the emanation effect for the purposes of the radioactive deposit prospecting and of general geology survey [10]. In the radiological protection, the exhalation rate can be used for the evaluation of the radon risk [9]. The exhalation rate is defined as an exhaled radon activity per mass or surface unit of the material sample and per unit of time.

Determination of the radon concentration in air can be done with the help of various methods and devices: the Lucas cells, track detectors, electroscope detectors, activated charcoal detectors etc. Theoretical bases of enumerated methods can be found in papers describing radon measurements [3, 5, 11]. All of these methods are relative, which means that they use a reference method in order to be calibrated. Every calibration is valid for a certain range of values. In nature, radon concentrations vary in very broad range from several Bq/m^3 in air upon the sea to the several hundreds kBq/m^3 in soil gas in the vicinity of uranium deposits. In this paper, a method for measuring the radon content in an emanation chamber with the help of a liquid scintillation counter is presented. This method may be applied for a very wide range of radon concentrations and requires neither calibration nor reference methods.

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Received: 18 January 2002, Accepted: 24 March 2003

Physical basis of the method

The three natural radon isotopes ^{222}Rn (radon), ^{220}Rn (thoron) and ^{219}Rn (actinon) with the half-lives $T_{1/2}$ = 3.82 days, 54.5 s and 3.92 s, respectively occur in all environments. However, as ^{220}Rn and ^{219}Rn have very short half-lives, their role in practical situations can be neglected.

The isotope ^{222}Rn decays according to the following scheme:

$$^{222}\mathrm{Rn}\ \xrightarrow[3.825\ \mathrm{d}]{\alpha}\ ^{218}\mathrm{Po}\ \xrightarrow[3.06\ \mathrm{min}]{\alpha}\ ^{214}\mathrm{Pb}\ \xrightarrow[26.6\ \mathrm{min}]{\beta}$$
$$^{214}\mathrm{Bi}\ \xrightarrow[19.7\ \mathrm{min}]{\beta}\ ^{214}\mathrm{Po}\ \xrightarrow[16.4\times10^{-4}\ \mathrm{s}]{\alpha}\ ^{210}\mathrm{Pb}$$

From the scheme some, quantitative conclusions can be deduced: (1) the radioactive equilibrium is reached after 3 h since the moment of radon gas sampling; (2) in this time the total alpha activity is 3 times greater than the radon activity; and (3) the ratio of beta to alpha activity is 2/3.

To determine the mass exhalation rate, a sample of investigated material must be hermetically closed in an emanation chamber through a certain interval of time. Then the radon activity in the air space of the emanation chamber is to be measured. To perform the measurement by a liquid scintillation counter, a radon air sample is collected from the chamber and mixed with a liquid scintillator, next the mixture is loaded to the vials. The efficiency of alpha particles detection for a liquid scintillation counter is equal to 100% [4], so the radon activity N_V (Bq) in the measured vial at the air sampling moment can be calculated by the following formula:

$$N_V = \frac{I_\alpha}{60\ F_\alpha(t)} \tag{1}$$

where: I_α – alpha count rate (cpm) from the vial measured by a liquid scintillation counter; t – period between the gas sampling and measurement; $F_\alpha(t)$ – the ratio $A_\alpha(t)/A_0$, where $A_\alpha(t)$ is the total alpha activity (radon + daughters) at the time t, and A_0 – radon activity at the sampling moment. Function $F_\alpha(t)$ may be expressed by the following equation (after Bateman [7]):

$$F_\alpha(t) = 3.01\exp(-\lambda_1 t) - 1.024\exp(-\lambda_2 t) - 4.28\exp(-\lambda_3 t) + 3.29\exp(-\lambda_4 t) \tag{2}$$

where: λ_1, λ_2, λ_3 and λ_4 are the decay constants of ^{222}Rn, ^{218}Po, ^{214}Pb and ^{214}Bi, respectively.

The radon activity N_{Sc} in the mixture of liquid scintillator and air sample is the sum of activities of the all measured vials, and N_{Sc} can be calculated by the formula:

$$N_{Sc} = \sum_{i=1}^{n} N_{Vi} \tag{3}$$

where: N_{Vi} – the radon activity in the i-th vial; n – number of measured vials.

A certain part of radon can be lost during mixing procedure of the radon air sample with the liquid scintillator. So, the radon activity in the air sample N_{pg} should be calculated as follow:

$$N_{pg} = k \cdot N_{Sc} \tag{4}$$

where: k is the correction factor for a loss of radon during mixing procedure of the air sample together with liquid scintillator.

Therefore, the radon activity N (Bq) in the air space of an emanation chamber at the moment of radon air sampling may be expressed by the formula:

$$N = \frac{N_{pg}\left(V_K - V_{pm}\right)}{V_{pg}} \tag{5}$$

where: V_K, V_{pm}, V_{pg} – volumes of the emanation chamber, the sample of the investigated material and air sample, respectively.

The radon activity N_1 (Bq) produced by the radium activity in the investigated material sample is calculated by the following equation:

$$N_1 = Ra \cdot m(1 - \mathrm{e}^{-\lambda_1 t_1}) \tag{6}$$

where: Ra is the activity concentration of ^{226}Ra in the investigated material in Bq/kg (it can be determined by the gamma spectrometry method [6]); m is the mass of the material sample (kg), and t_1 is the time of the ^{222}Rn activity growth inside the emanation chamber from a material sample.

A relative exhalation rate R in per cent is defined as the ratio of the measured radon activity exhaled from a sample to the radon activity produced by this sample

$$R = \frac{N}{N_1}100\%. \tag{7}$$

The mass exhalation rate E_m (Bq·kg^{-1}·h^{-1}) may be calculated by the formula [1]:

$$E_m = \frac{N \cdot \lambda_1}{m \cdot \left(1 - \exp\left(-\lambda_1 \cdot t_1\right)\right)} \tag{8}$$

where: m is the mass (kg) of the sample and the rest of symbols are the same meaning as above.

Measurement procedure and results

In our laboratory, for the determination of the exhalation rates a chamber of dimensions of 20 × 20 × 25 cm (10 dm^3) was used. An investigated material sample of volume of about 0.5 dm^3 is hermetically closed in the chamber. Depending on the kind of the investigated material, the time of radon activity growth in the chamber amounted from several days to 30 days. The air sample of the volume of 100 cm^3 was extracted from the emanation chamber through a filter with the help of a syringe. The gas sample was mixed with 40 cm^3 of the liquid scintillator in a laboratory glass vessel of 1.8 cm inner diameter and 25 cm high (ca 64 cm^3). A view of the mixing system is presented in Fig. 1. Then, the liquid mixture was divided into two vials (the volume of each one amounts to 22 cm^3) and after 3 h since the extraction of the gas sample, the count rates of the vials were measured with the help of the α/β liquid

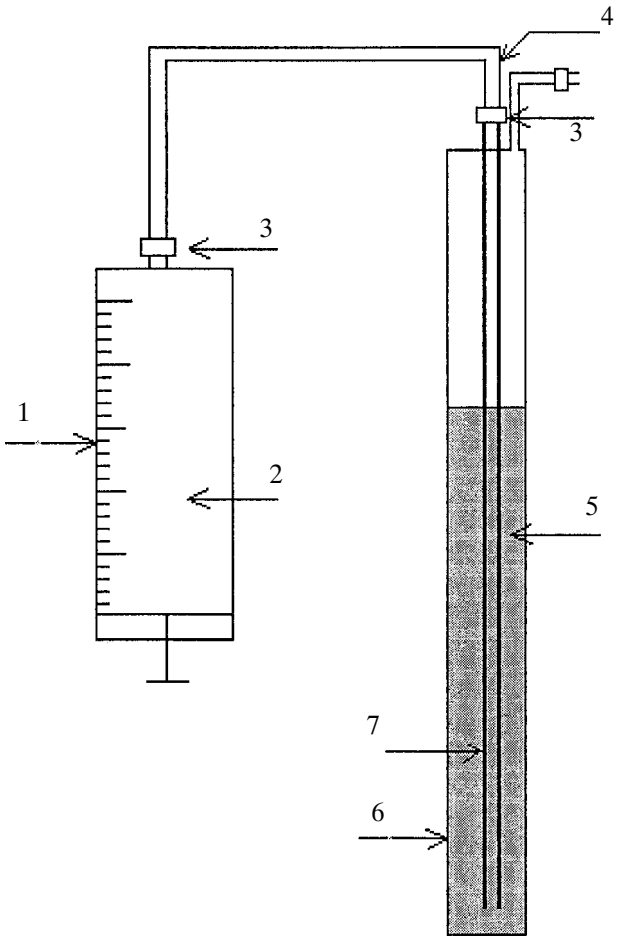

Fig. 1. A view of mixing system: 1 – calibrated syringe; 2 – radon gas; 3 – valve; 4 – plastic pipeline; 5 – liquid scintillator; 6 – glass vessel; 7 – glass tube.

scintillation counter. The value of the PSA (pulse shape analysis) parameter was adopted this way that ensured that

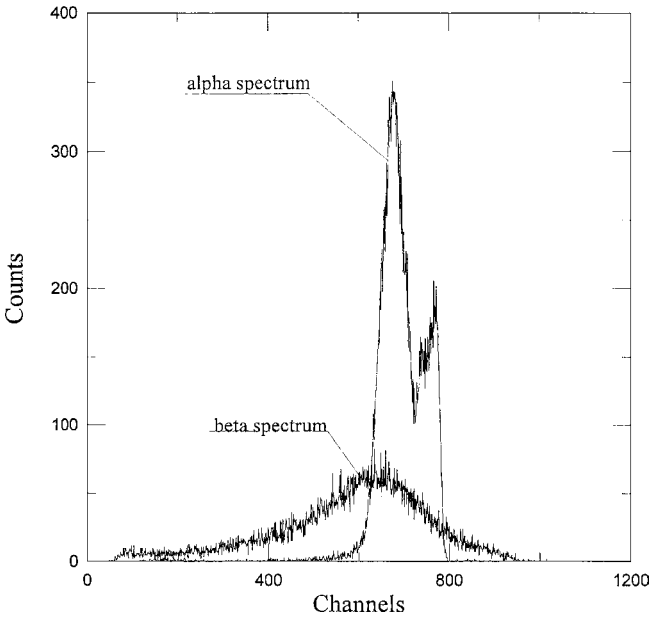

Fig. 2. The measured spectra of a mix of radon gas and liquid scintillator.

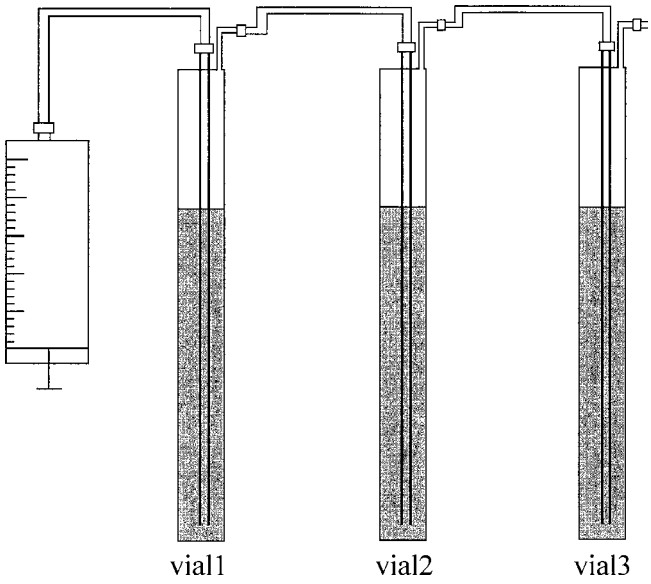

Fig. 3. A scheme of connecting system of vessels.

the ratio of the measured β and α net count rates was equal to 2/3. In Fig. 2, the spectra of α and β radiation of the radon sample measured with the help of liquid scintillation counter are presented.

To determine the correction factor for a loss of radon during mixing procedure of the radon air sample with the liquid scintillator, we connected three glass vessels in a series (Fig. 3) (each vessel contains 40 cm^3 of liquid scintillator). Then, 100 cm^3 of the radon gas of an activity concentration of 54 ± 3 kBq/m^3 was pumped into the system. Next, the liquid scintillator, with the mixed-in radon gas, was transferred from the three vessels to 6 vials. Then, the vials were measured by the α/β liquid scintillation counter. The radon activity in each vessel was calculated by formulas (1) and (3), (in this case the number of the vials for every vessel was equal to (2)) and it was equal to 3.67 ± 0.24, 1.157 ± 0.055 and 0.364 ± 0.024 Bq in the first, second and third vessel, respectively. From these data, one can deduce that the ratio of radon activity in vessel 2 to the radon in vessel 1 is statistically equal to the ratio of radon activity in vessel 3 to the radon in the vessel 2, both ratios being equal to 0.315 ± 0.025. One can thus conclude, that the radon activities in the consecutive vessels create a decreasing geometrical series with a common ratio (q) of value 0.315. Therefore, the radon activity in the air sample can be calculated as follows:

$$N_{pg} = \frac{N_{Sc1}}{1-q} = \frac{N_{Sc1}}{1-0.315} = 1.460 \cdot N_{Sc1} \tag{9}$$

where: N_{Sc1} is the radon activity in the first vessel and N_{pg} is the radon activity of the air sample of a volume of 100 cm^3.

Thus, in our experimental procedure the radon activity N_{Sc} in the vessel should be multiplied by the loss coefficient of 1.46 to obtain the radon activity N_{pg} in the radon air sample extracted from the emanation chamber. Knowing the radon activity in the air sample, we can calculate the radon activity in the air space of the emanation chamber by the formula (5).

Table 1. Relative and mass exhalation rates measured for some of the investigated samples.

Sample	Mass of sample (g)	^{226}Ra ± 2σ (Bq/kg)	R ± 2σ (%)	E_m ± 2σ (Bq·kg^{-1}·h^{-1})
SP6*	904.22 ± 0.05	4210 ± 300	10.95 ± 1.12	3.48 ± 0.36
90% SP6 + 10% cement	998.39 ± 0.05	3800 ± 300	15.3 ± 1.6	4.39 ± 0.44
70% SP6 + 30% cement	1046.05 ± 0.05	2950 ± 200	17.1 ± 1.8	3.81 ± 0.35
90% SP6 + 10% anhydrite	959.45 ± 0.05	3790 ± 280	11.76 ± 1.16	3.36 ± 0.33
70% SP6 + 30% anhydrite	975.90 ± 0.05	2580 ± 160	4.29 ± 0.54	0.83 ± 0.09
90% SP6 + 10% gypsum	885.80 ± 0.05	2966 ± 220	9.00 ± 1.05	2.01 ± 0.23
70% SP6 + 30% gypsum	861.41 ± 0.05	2840 ± 180	1.04 ± 0.08	0.223 ± 0.021
Ytong	887.57 ± 0.05	20.8 ± 2.0	2.29 ± 0.73	0.0036 ± 0.0011
Copper slag	1051.0 ± 0.05	304 ± 16	0.054 ± 0.023	0.0012 ± 0.0005
Full red brick	1250.63 ± 0.05	95.2 ± 2.0	0.248 ± 0.088	0.0018 ± 0.0006

* The uranium tailing piles at Kowary in Poland.

Experiments for the determination of the total radon activity in the liquid scintillator as a function of variation of the air volume in the glass vessel were carried out. The measurements have shown that about 3% of the total radon activity remained in the air, filling 30% of the vessel volume above the scintillator. The results are in good agreement with the data of Murase *et al.* [8].

Calculated values of the relative and mass exhalation rates of the samples taken from the uranium tailing piles and building materials are presented in Table 1.

The results from this Table reveal that the samples obtained by mixing of the waste of the uranium tailing with cement have the highest relative exhalation rate, while the lowest rate belongs to the copper slag.

The minimum detectable activity (MDA) was calculated according to the criterion defined by Curie [2] as follows:

$$\text{MDA (Bq·m}^{-3}) = \frac{L_D}{VT\varepsilon \cdot 60 \cdot 30} \tag{10}$$

$$L_D = 2.7 + 4.65\sqrt{C_B \cdot T} \tag{10a}$$

where: V – volume of gas sample in (m^3); T – background measurement time (min), C_B – background count rate (cpm) and ε is the detection efficiency. The number 3 in the nominative of formula (10) results from the fact that during measurement of the vial, the radon in the liquid scintillator is in equilibrium with two alpha radioactive daughters. Thus, just 3 counted alpha particles comes from radon itself.

In our case $V = 1 \times 10^{-4}$ m^3, $T = 30$ min, $\varepsilon = 1$ and $C_B = 1.1$ cpm in the alpha channel, therefore the MDA for radon concentration in the air of this method is equal to 0.054 kBq·m^{-3}.

Conclusions

The presented method for the determination of radon activity requires neither calibration nor reference standards and is easy to perform.

The efficiency of the method is high enough and we can apply it for the determination of the exhalation rates of building materials.

Acknowledgments The present authors thank S. Kalita Ph.D., P. Jodłowski Ph.D., A. Bolewski Ph.D. and A. Lenda D.Sc. for preparing, measuring of the ^{226}Ra activities of researched samples and language corrections. We are also greatly indebted to the Reviewers for their valuable remarks.

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