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Measurement of radon (^{222}Rn) and thoron (^{220}Rn) concentration with a single scintillation cell

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Abstract. A single scintillation cell (ϕ 54 × 74 mm) is used for the measurement of radon and thoron. The radon and thoron laden air is filtered and forced to flow at 1 dm^3/min through the scintillation cell in the period of 1–10 min. The count number from alpha radiation is registered in the periods of 3–10 min and 20–30 min. Two values of detection and deposition efficiency of alpha radiation are used for radon (at air flow and air at rest in the cell) and for thoron. Measurements of radon laden air and thoron laden air showed good agreement between the reference concentration and the measured concentration, not worse than 1% for radon and not worse than 2% for thoron. Combination of radon + thoron concentration showed also a small interference ("cross talk") not worse than 1%.

Key words: radon • thoron • measurement • scintillation cell

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Introduction

Radon ^{222}Rn concentration in air is usually much higher than thoron (^{220}Rn) concentration, but in some cases thoron concentration is also high and radon with thoron have to be measured simultaneously at the same time. A wide review of passive and active instruments for measurement of radon and thoron is given in [2]. Thoron and radon is measured by counting ^{218}Po radiation from radon and ^{216}Po radiation from thoron by a semiconductor detector as described in [8, 9]. A single scintillation cell was used for the measurement radon and thoron concentration by Tokonami *et al.* in [10]. Here, the air containing radon and thoron was injected into a scintillation cell and the count number was measured in two time intervals of 20–120 s and 10–15 min after injection of the air. Radon concentration is proportional to the count rate in the second time interval, thoron concentration is proportional to the count number in the first time interval corrected by the count number from radon. Two scintillation cells were used for continuous measurement of radon and thoron concentration by Coleman [1]. Radon and thoron laden air was forced to flow through two scintillation cells connected in series with a delay line between the cells. Count rate in the first cell is proportional to the radon and thoron concentration, count rate in the second cell is proportional to the radon concentration. The difference of the count rate in both cells is proportional to the thoron concentration. Another method for radon and thoron measurement employs two ionization chambers [3].

The aim of the presented investigation was the development of a model gauge for the measurement of radon and thoron concentration with forced flow of radon and thoron laden air through a single scintillation cell ensuring thus higher sensitivity of measurement of thoron.

Principle of operation

Thoron and radon laden air is forced to flow at 1 dm^3/min through a Lucas cell of size 0.17 dm^3 (ϕ 54 × 74 mm), during the period of 10 min, and simultaneously count rate from the Lucas cell is measured. After 10 min, the air flow is switched off. Count rate is measured in two time intervals of 3 to 10 min and 20 to 30 min after the start of air flow. The chosen time intervals are a compromise between the length of measuring cycle and the size of random errors that decrease with increasing counting time. Simulated alpha activity, when only radon or only thoron is forced to flow through the cell is shown in Fig. 1, at hundred percent assumed detection and deposition efficiency. The curve rs in the upper diagram shows the variation of alpha activity from radon, when at time $t = 0$ the activity of 1 dis/min (disintegration per minute) is introduced into the cell and the radon gas is at rest inside the cell. The curve rsr shows variation of alpha activity when the same radon activity flows through the cell during the period of 1–10 min. Both curves in the period of 20–30 min practically coincide. Measurements with the Lucas cell in a radon chamber showed that the deposition and detection efficiency of alpha radiation at a flow rate of 1 dm^3/min of radon laden air was 20% lower than the efficiency when radon laden air was injected into the cell, without flow. The explanation may be that part of the ^{218}Po nuclei produced inside the cell are taken away and do not deposit on the walls of the cell. In the remaining period (after the first 10 min), when radon is at rest inside the scintillation cell all ^{218}Po decays contribute to the alpha activity registered. Series decay of $^{222}\mathrm{Rn} \to {}^{218}\mathrm{Po} \to {}^{214}\mathrm{Pb} \to {}^{214}\mathrm{Bi}$ (^{214}Po) was simulated. For the lower diagram, the series decay of $^{220}\mathrm{Rn} \to {}^{216}\mathrm{Po} \to {}^{212}\mathrm{Pb} \to {}^{212}\mathrm{Bi}$ was simulated.

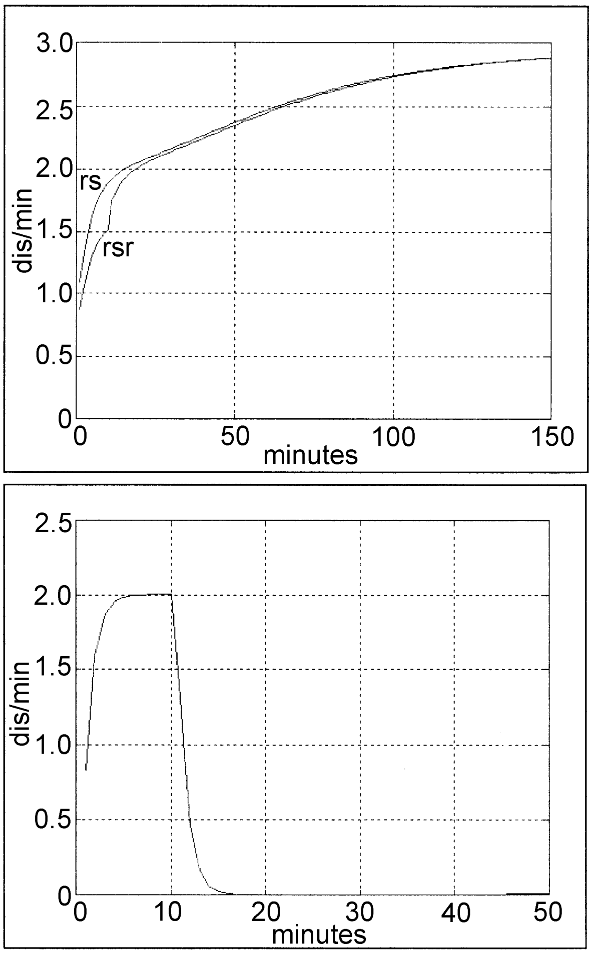


Fig. 1. Upper diagram – rsr simulated alpha activity deposited inside a scintillation cell when 1 dis/min of radon ^{222}Rn is flowing through the cell during 10 min, rs simulated alpha activity when radon 1 dis/min is at rest in the cell. Lower diagram – simulated alpha activity deposited inside the scintillation cell when 1 dis/min of ^{220}Rn is flowing through the cell during 10 min.

The relation between the count numbers n_1 and n_2 in the two time intervals and the alpha activity deposited inside the cell can be written as:

$$\begin{aligned} n_1 &= m_{11}{*}\varepsilon_{11}{*}A + m_{12}{*}\varepsilon_{12}{*}B \\ n_2 &= m_{21}{*}\varepsilon_{21}{*}A + m_{22}{*}\varepsilon_{22}{*}B \end{aligned} \tag{1}$$

where: A – radon alpha activity; B – thoron alpha activity; ε_{11}–ε_{22} – deposition and detection efficiencies. The coefficients m_{11},.., m_{22} are determined from the simulated activity of alpha radiation as presented in [4–6], see Fig. 1. The coefficients m_{11}, m_{12} are equal to the total (sum) alpha activity in the 3–10 min interval from radon and thoron respectively, and m_{21}, m_{22} to the total alpha activity in the interval of 20–30 min from radon and thoron. After computation the coefficients matrix **m** is equal to:

$$\mathbf{m} = \begin{bmatrix} 13.6299 & 15.8317 \\ 23.2505 & 0.0218 \end{bmatrix} \text{– coefficients determined from simulation of alpha activity}$$

Alpha detection efficiency of ^{222}Rn, ^{218}Po, ^{220}Rn and ^{216}Po were estimated with Monte Carlo calculations [10] for the scintillation cell (ϕ 53 × 70 mm) similar to the cell used in present investigations. The average detection efficiency for ^{222}Rn, ^{218}Po, was 0.722 p/dis (pulse per alpha disintegration), and for ^{220}Rn and ^{216}Po 0.804 p/dis. Detection efficiency for thoron was thus 1.114 times higher than that for radon. Alpha deposition and detection efficiency ε for radon of the scintillation cell used in the investigations was $\varepsilon_{21} = 0.66$ p/dis at rest inside the cell. Laboratory measurements showed that at an air flow of 1 dm^3/min containing radon, the deposition and detection efficiency were 0.8 times lower. Similar investigations showed that thoron detection and deposition efficiency at an air flow of 1 dm^3/min was 0.7 times lower. The matrix of deposition and detection efficiencies for the Lucas cell used can thus be written as:

$$\varepsilon = \begin{bmatrix} 0.528 & 0.515 \\ 0.660 & 0.735 \end{bmatrix} \text{– deposition and detection coefficients}$$

and Eq. (1) can be rewritten as:

$$\text{(2)} \qquad \begin{aligned} n_1 &= 7.1966\,A + 8.1533\,B \\ n_2 &= 15.3453\,A + 0.0160\,B \end{aligned}$$

Concentration of radon A and thoron B from the set of two Eqs. (2) can be computed employing matrix computations

$$\text{(3)} \qquad \mathbf{y} = (\mathbf{M}^T\mathbf{M})^{-1}\mathbf{M}^T\mathbf{n}$$

where:

$$\mathbf{M} = \begin{bmatrix} 7.1966 & 8.1533 \\ 15.3453 & 0.0160 \end{bmatrix}, \ \boldsymbol{n} = \begin{bmatrix} n_1 \\ n_2 \end{bmatrix}, \ y = \begin{bmatrix} A \\ B \end{bmatrix} - \text{dis/min}$$

Employing Cramer rules, the concentration of radon and thoron can be given in a more convenient form for microprocessor processing:

$$\text{(4)} \qquad \begin{aligned} A &= -0.0001n_1 + 0.0652n_2 \\ B &= 0.1228n_1 - 0.0576n_2 \end{aligned} \quad \text{(dis/min)}$$

Conversion factor from dis/min into Bq/m^3 is equal to:

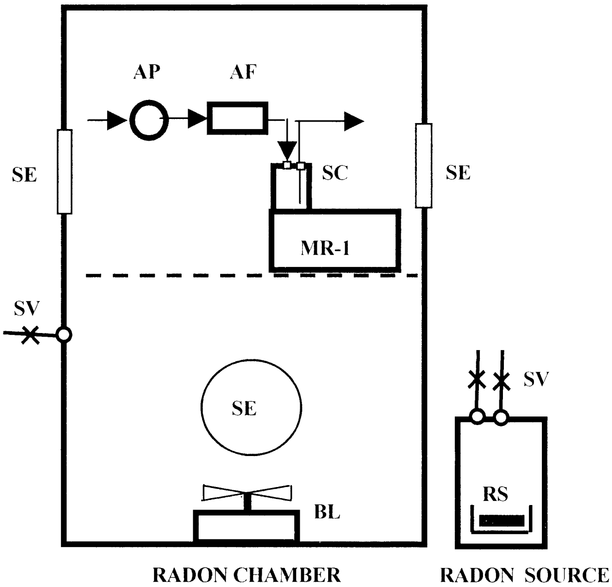
$$\text{(5)} \qquad k = \frac{1000}{60^* \, v} = 98$$

where: $v = 0.17$ dm^3 – volume of scintillation cell.

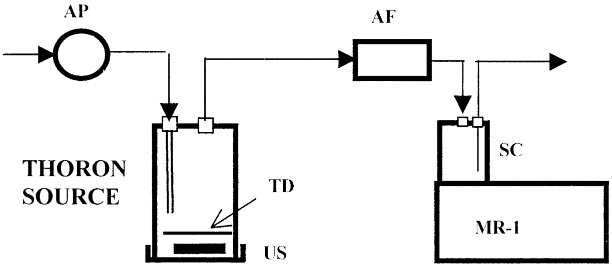
Multiplying thus the concentration computed from Eq. (3) or (4) by coefficient concentration of radon and thoron is expressed in Bq/m^3.

The method presented in [10] for simultaneous measurement of radon and thoron employs also a single scintillation cell into which a single injection of thoron and radon air is made. Count number C_1 is measured in the time interval of 20 to 120 s and count number C_2 in 10 to 15 min interval after sampling. Radon concentration is proportional to the count number C_2. Thoron concentration is proportional to $C_1 - kC_2$, where $k = 0.2$. Random error of thoron concentration is not given in Ref. [10], but as the count rate C_1 is measured within 100 s on the falling slope of thoron activity, the count number is low, and consequently a high random error of thoron concentration is inevitable.

In the method presented radon and thoron laden air is forced to flow through the scintillation cell for quite a long period of time enabling thus a longer counting time, higher count number from thoron, and lower random error. For the same reason, the counting time for radon was increased. As the air flow through the scintillation cell influences deposition and detection efficiency of radon and thoron decay products, the deposition and detection efficiency had to be taken into account at rest and at the flow of radon and thoron laden air. Finally, the approach to the signal processing is quite different from that in [10].



AP – air pump, 1 dm^3/min flow; AF – air filter; SC – scintillation cell; MR-1 – portable radon monitor [7]; SE – sealed entrance opening; SV – valve with rubber sealing; RS – open Ra-226 source; BL – blower for mixing air in the chamber



AP – air pump, 1 dm^3/min flow; AF – air filter; SC – scintillation cell; US – open U-232 source; TD – exchangeable thoron diffusion membrane; MR-1 – portable radon monitor [7]

Fig. 2. Measuring arrangements for investigation of radon – upper drawing, and of thoron concentration – lower drawing. A medical syringe was used to introduce radon from the radon source to the radon chamber. The different thoron concentration of thoron source was achieved thanks to the different material and thickness of exchangeable diffusion membrane TD.

Experimental measurements

Radon laden air was forced to flow at 1 dm^3/min, during 10 min, through an air filter and through the Lucas cell that was installed in an MR-1 radon monitor [7] and was placed inside the radon chamber filled with radon, see Fig. 2 (upper diagram). To protect the scintillation cell against daylight, the inlet and outlet of air from the cell was pumped through U shaped black rubber pipes. Simultaneously, the pulse count rate of Lucas cell was measured during the time period of up to 220 min by the MR-1 monitor operating as pulse counter. The measured count rate (broken curve) is shown in Fig. 3 (left diagram). The simulated count rate (smooth curve) is also given for comparison. Summing up count rates in the 3–10 min and 20–30 min intervals the following count numbers were obtained: $n_1 = 581$, $n_2 = 1287$. The corresponding computed radon and thoron concentrations, Eq. (3), expressed in Bq/m^3 are $A = 8212$ Bq/m^3 and $B = 0$ Bq/m^3 (from computation B was –265 Bq/m^3 but as no negative concentration exists $B = 0$ was taken). The real (reference) radon concentration was determined from the mean count rate at radiation equilibrium

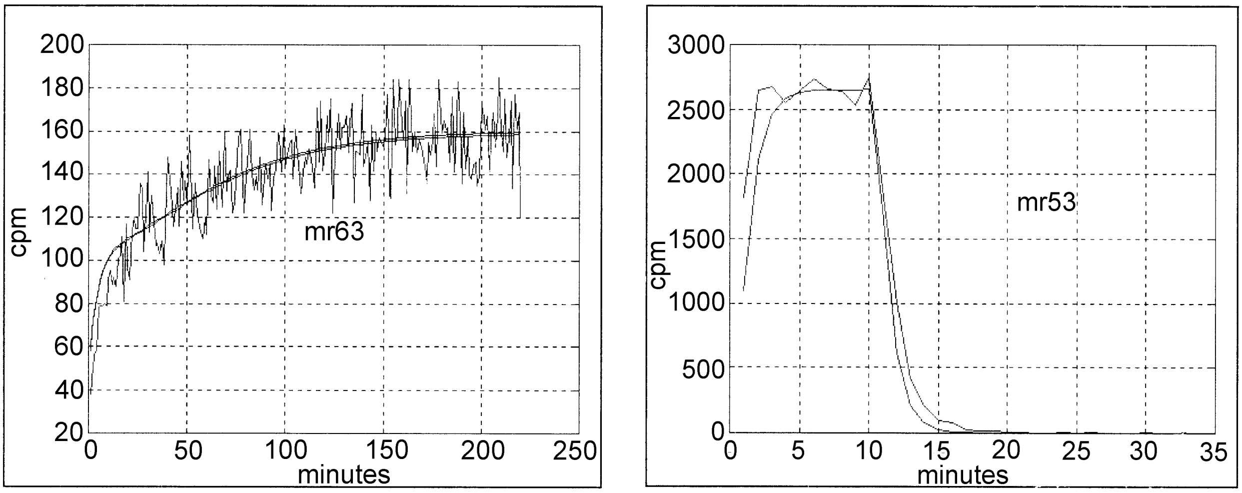


Fig. 3. Measured and simulated count rate, cpm (count per min), against time for radon (left diagram) and thoron (right diagram). Broken curves – measured cpm, smooth curves – simulated cpm.

(191–210 min) at $\varepsilon_{21} = 0.66$ which was equal to 7879 Bq/m^3.

Similarly, thoron was forced to flow through the air filter and through the Lucas cell from thoron source cell (0.23 dm^3) containing ^{232}U, see Fig. 2 (lower diagram). Thoron source was ventilated at an air flow of 1 dm^3/min until radiation equilibrium was reached, and then the outlet of thoron source was connected through the air filter to the Lucas cell at the moment of starting count rate measurement. The ^{232}U source was an old source in radiation equilibrium with ^{228}Th and ^{224}Ra, ($^{232}\mathrm{U} \rightarrow {}^{228}\mathrm{Th} \rightarrow {}^{224}\mathrm{Ra} \rightarrow {}^{220}\mathrm{Rn}$). The measured and simulated count rate of thoron is given in Fig. 3 (right diagram). The registered count numbers were $n_1 = 21{,}185$ and $n_2 = 48.6$. The reference concentration was computed from the mean count rate at radiation equilibrium with $\varepsilon_{12} = 0.515$ and was equal to 254,506 Bq/m^3. The computed thoron concentration was equal to 251,782 Bq/m^3. The results of series of measurements of radon and thoron concentration are given in Table 1.

Apart from the individual measurements of radon and thoron, count numbers of both elements in both time intervals were added, and radon and thoron concentrations were computed according to the developed relations. The last line in Table 1 is an example of such a radon and thoron combination. To check repeatability of the measurements, sixteen such combinations for different radon and thoron concentration were investigated. They are not shown in Table 1.

The following conclusions can be drawn from the data given in Table 1, from the combinations of radon and thoron, and from general relations of the presented method.

- Mean ratio of the columns 2/4 in Table 1 for radon is 0.992 ± 0.034 and the ratio of columns 3/5 for thoron is equal to 1.018 ± 0.018, indicating quite a good agreement of the measured concentration with reference concentration. The measured thoron concentration is approximately 2% too high and can be, if necessary, corrected (divided by a factor of 1.018).

Table 1. Results of radon and thoron measurements t_1 = 3–10 min, t_2 = 20–30 min

Measurement	^{222}Rn (Bq/m^3)	^{220}Rn (Bq/m^3)	^{222}Rn (Bq/m^3)	^{220}Rn (Bq/m^3)	Note
	measured		reference		
1	2	3	4	5	6
mr62	10,260	862	10,701	0	Radon
mr63	8212	0	7879	0	„
mr64	8241	0	8310	0	„
mr65	6419	0	6575	0	„
mr53	49	254,506	0	251,762	Thoron
mr147	0	120,540	0	119,070	„
mr148	49	171,892	0	170,030	„
mr150	49	7330	0	7271	„
mr151	14	8085	0	7702	„
mr151+mr65	6409	7967	6575	7702	R+T

- Mean ratio of the columns 2/4 for all the radon + thoron combinations is equal to 0.995 ± 0.028 for radon, and the columns 3/5 for thoron is equal 1.024 ± 0.024. This means that the interference of radon signal in thoron reading and *vice versa* ("cross talk") between radon and thoron is negligible. The interference is mostly visible when the thoron concentration is zero and radon concentration is measured. In three cases the computed concentration was negative and this is indicated in Table 1 as "0".
- The random error due to the count rate fluctuations depends on the radon and thoron concentration and number of counts registered. The random error of radon concentration can be calculated from the relation for propagation of errors.

$$s^2(A) = \left(\frac{\partial A}{\partial n_1}\right)^2 s^2(n_1) + \left(\frac{\partial A}{\partial n_2}\right)^2 s^2(n_2) \tag{6}$$

The random error for thoron can be computed from a similar relation, see Eqs. (4) and conversion coefficient (5). The random errors for data given in the last line in Table 1 are equal to 202 Bq/m^3 (3.1%) for radon and 443 Bq/m^3 (5.8%) for thoron at 1 σ level.

For a radon and thoron concentration of 98 Bq/m^3 (1 dis/min), the corresponding count numbers in both time intervals are $n_1 = 15.4$ and $n_2 = 15.3$ counts, see Eq. (2). Standard deviation for such a concentration is 25.5 Bq/m^3 (26%) for radon, and 52 Bq/m^3 (53%) for thoron.

Conclusion

The radon and thoron concentration in air can be measured with a single Lucas cell, through which the radon and thoron laden air is forced to flow in the period of 1–10 min, and count rate is measured in two time intervals: 3–10 min and 20–30 min. Proper processing of the count numbers registered ensures that the measurements are repetitive and only a negligible interference exists between the radon and thoron channels. The error due to the way of processing is not worse than 2%. Random error due to fluctuations of the registered count number is 26% for radon and 53% for thoron concentration at 1 σ level, and at a radon and thoron concentration of 98 Bq/m^3.

The presented investigations indicate that a gauge with a single scintillation cell can be used not only for concentration measurement of radon, but also of thoron, with acceptable accuracy. The measuring possibilities of the gauge developed earlier in the Institute of Nuclear Chemistry and Technology [7] can thus be extended.

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