SIMULTANEOUS MEASUREMENT OF RADON AND THORON EXHALATION RATE FROM SOIL AND BUILDING MATERIALS

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ABSTRACT

The mainly sources for indoor radon concentrations in all types of houses are the soil radon flux under the building (about 80%) and the radon exhalation from building materials (about20%). Our paper presents two methods for simultaneous measurement of this exhalation from soil and building materials: (1) -charcoal adsorption, respectively (2)-Lucas cell method.

The both methods are applicable especially in the case of a soil flux enhanced in thoron gas. In the Lucas cell case a short accumulation time was used (10-15 minutes). The thoron concentration under accumulation volume was measured immediately after the sample gas extraction and a regression equation is used for determining equilibrium thoron concentration.

In the case of charcoal method the thoron was measured 4 hours after a special degassing of the sample. Using the LUK-3A device for the building materials, the thoron flux was measured only on a special enhanced thorium+radium sample and the results are not presented here.

INDEX TERMS

Radon and Thoron, Exhalation, Charcoal adsorption, Soil radon

INTRODUCTION

The radon isotopes ^{220}Rn (thoron) and ^{222}Rn (radon) were intensely studied in the last time due to their involvement in the lung cancer risk (Nazaroff and Nero, 1988) and also for geological purposes (Akerblom and Melander, 1996). The main sources of the indoor radon are the soil radon and radon exhalation from building materials (Stranden, 1988). The active charcoal was often utilized for radon measurement especially for indoor radon (George, 1984; Prichard and Marien, 1985). The method of the soil radon and exhalation measurement from the soil and building materials using adsorption in active charcoal was also used (Megumi and Mamuro, 1979; Li, Schery and Turk, 1992; Oberstedt and Vanmarke, 1996) with good results but in this case corrections regarding charcoal humidity related of the break point of charcoal are needed (Scarpitta, 1996).

In all these cases the radon adsorbed in charcoal is measured by gamma spectrometry, commonly with NaI(Tl) detectors. For economical and practical reasons, the charcoal canisters must be degassed and re-used of many times. Commonly, the charcoal cleaning is made by degassing the charcoal at 120-140°C during 12-14 hours, but in the case of high radon content adsorbed in charcoal, for a complete desorption of radon, 2-3 steps are necessary (Cosma, Van Deynse and Poffijn, 1997). The method for charcoal degassing proposed in this work has the advantage to be more rapid (10-15 minutes) and it produces a much better degassing. On the other hand, the manner of degassing allows the thoron determination from charcoal even in the case when it is measured by low gamma

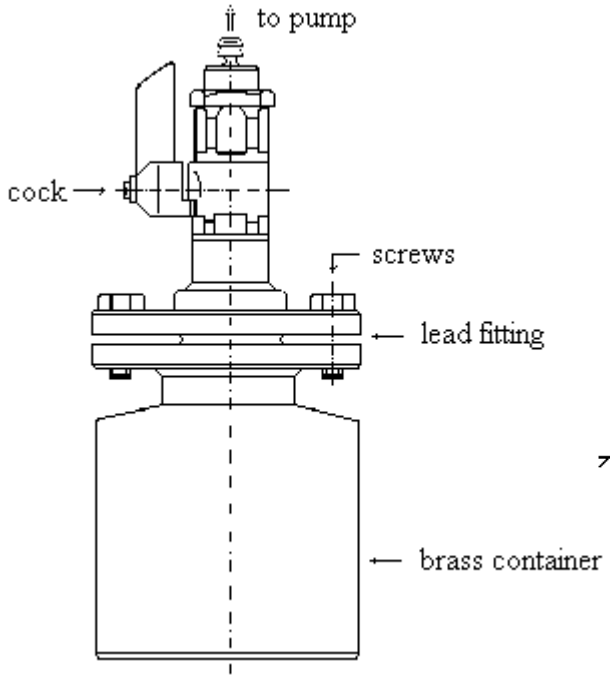
spectrometry using NaI (Tl) detectors. Generally, the measurement methods for thoron determination require either alpha or high gamma resolution spectroscopy.

This work presents the method for simultaneous measurement of radon and thoron from soil using charcoal adsorption. Exhalation of radon from some Romanian buildings materials obtained by charcoal adsorption method was verified in several cases by Lucas cell method. In the case of one sample the result was compared with those obtained in the frame of ERRICCA Intercomparison Exercise from Athens in April 1999 (Petropoulos, Anagnostakis and Simopoulos, 1999). Finally, the both radon and thoron exhalation from a special concrete sample made in our laboratory, artificially enhanced in ^{226}Ra and also in $^{224}Ra(Th[NO_3]_2)$ was made.

EXPERIMENTAL METHODS

A brass cylindrical vessel of 0,7 L, Fig. 1, was used for charcoal degassing. In the upper part it is connected to a vacuum pump. After filling (250 g of charcoal) and closing using six screws and a lead fitting, the vessel is put into another larger vessel with water. The later is placed on an electric boiling. When reaching the water boiling temperature, the pump is started for 15 minutes. The charcoal is then pulled out and can be used for radon-thoron adsorption in the next exposure.

Measuring the charcoal radioactivity after this operation, the control of degassing efficiency is made. After about 3,5 hours from the end of the degassing process, the gamma radioactivity is



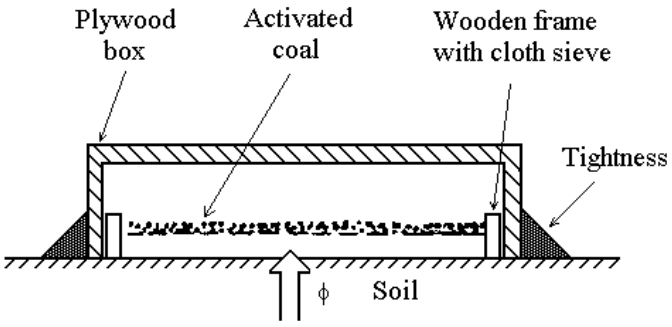


Fig.1.The brass container for radon degassing **Fig.2.**The device for radon flux gathering

reduced to the background radioactivity of the charcoal.

For radon exhalation gathered from soil was used the device from Fig. 2, which consists from a frame box of 0.23 m^2 (37x62 cm), and about 200g degassed charcoal. After exposure (as a rule, 4-12 hours) the charcoal is introduced into the Marinelli vessel and gamma radioactivity is

measured over the whole spectrum with an NP424 gamma spectrometer equipped with a large NaI(Tl) detector (76x45 mm) enclosed in a lead castle of 30 mm wall thickness to reduce the

background. In the case of thoron absence from the charcoal, the radon exhalation rate Φ_{Rn} can be calculated following equation:

$$\Phi_{Rn} = C \frac{A_0}{t_e(h)S(m^2)} \cdot e^{\frac{\ln 2}{T_{Rn}} \cdot t_c} \quad (\mathrm{Bq/m^2 s}) \qquad (1)$$

where C=1.412*10^{-3} is the calibration factor, A_0-counts per second under integral spectrum (E_0>60keV); t_e- exposure time (usually 4-12 hours); S=0.23m^2 the collecting area; T =3.825 days is the half-life time of radon and t_c the time interval between the middle of the exposure time and the beginning of the measurement. A time interval longer as 3.5 hours must be considered between the end of sampling and the start of measurement.

In the case of an important thoron exhalation that accompanies the radon flux, this equation can be used only if t_c is 5-6 times greater than the half time of thoron descendants (10.65h) that is about two days. In this time the charcoal from Marinelli vessel must be very well sealed so that the radon must to rest in this container. The thoron exhalation can be determined if the exposed charcoal is firstly degassed as above and the sample is gamma measured after 4 hours when the radon progeny decreased of about 500 times. Fig.3 represents such experiment. Therefore the initial sample must be divided in two: one for radon and other for thoron measurement.

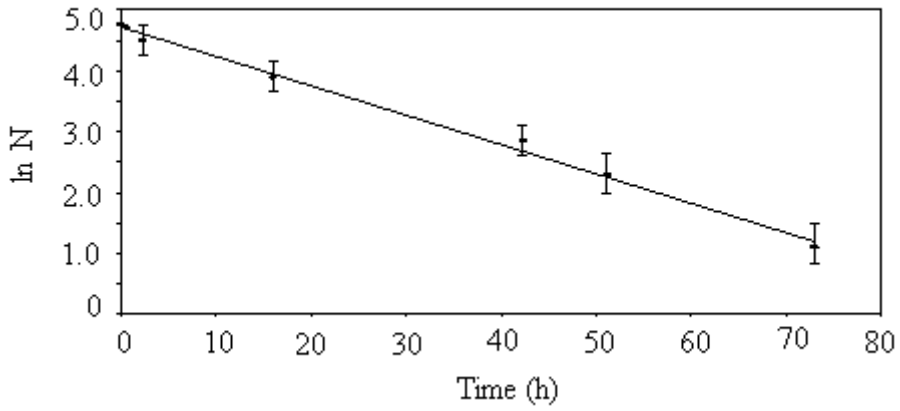


Fig.3. Thoron progeny decreasing four hours after the exposed sample was degassed.
($T_{1/2}$ = tgα = 11.3 h)

For building material exhalation measurement two types of square (33×33 cm) metallic and plastic containers of 8 cm height and 20 cm height respectively were used. These were placed on a shiny surface and air sealed with a special material. At the upper part, there is a small orifice (Ø=0,3 mm), which must be also sealed by a small quantity of same material. These orifices allow the extraction, with a special medical syringe, of a determined gas volume from container, which is then analyzed using Lucas cell method with LUK 3A device. LUK 3A device produced in Czech Republic is a portable instrument, programmable for three kinds of measurements: soil radon, soil radon and thoron and radon in water.
The samples (concrete, bricks, tills) were enclosed into these containers together with 50 g of activated charcoal recently degassed. The charcoal thickness under this accumulation volume was 1,5-2 mm assuring an efficient adsorbing process both for radon and thoron atoms.
After adsorption time (2-96h) the charcoal was put into well sealed standard metallic cans and measured by means of the same NaI(Tl) spectrometer for 500 s. The calibration for measuring

under whole spectrum (E_0>60 keV) was made using ^{226}Ra and ^{224}Ra (^{232}Th) sources and, the obtained values were c_1=3,02 Bq/count/s for radon and c_2= 4.64 Bq/count/s for thoron respectively. For energy values higher then 2400 keV this factor for thoron is 320 Bq/count/s. The emanated radon quantity A_0 (Bq) was adjusted considering the quantity of radon adsorbed in charcoal A_c and the quantity A_a present into the real emanation volume V_0-V_s following equations:

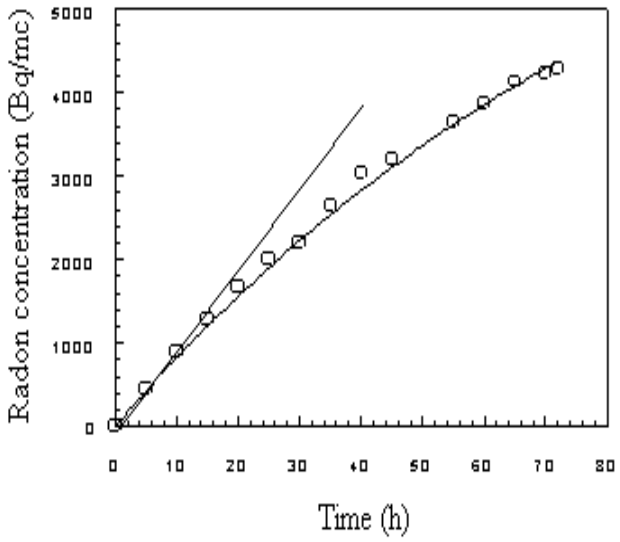
$$A_0 = A_c + A_a \quad (2)$$

$$A_c = km\frac{A_a}{V_0 - V_s} \quad (3)$$

where k=1.41 m^3/kg is the adsorption constant for our charcoal (type CAS-Buzau), m =50g being the mass of adsorbing charcoal, V_0 and V_s represent the container volume and the sample volume respectively. The radon exhalation flux can be calculated as following:

$$\Phi = \frac{\ln 2}{T_{Rn}} \cdot \frac{A_0}{1 - e^{-\lambda T}} \quad \text{(Bq/s)} \quad (4)$$

In the case of a long collection time a correction of about 10-15% is needed due to leakage and back-diffusion processes. In the case of the sample "Specimen 3" measured in the frame of ERRICCA Intercomparison Exercise another method were also used to determine the radon exhalation from this sample. In this case under accumulation volume a device Radim (Czech Republic) that measures continuously the radon concentration was introduced. The instrument Radim registers the α radiation from radon and its descendants using a Si detector. In Fig.4 are shown the results of radon activity in the emanation volume as a time function measured with Radim instrument.



From the curve in Fig.4 it is possible to determine the radon exhalation rate by interpolation of this curve with the equation (Jonassen, 1983):

$$C = C_0[1 - \exp(-\lambda_1 t)] \quad (5)$$

Fig.4. Radon variation in accumulation container measured with Radim cell method.

In relation (5) $\lambda_1 = \lambda_{Rn} + \lambda^*$, λ^* being the leakage and the back-diffusion constant. The exhalation rate can be found from relation:

$$\Phi = C_O\,(V_o - V_s)\cdot\lambda_1 \quad (6)$$

By fitting the experimental points with equation (5) we obtained the following values C_0=8950 Bq/m^3, λ_1=0.0105 h^{-1}and and $\lambda^* = \lambda_1 - \lambda_{Rn}$=0.0035 h^{-1}. From this later value one can obtain information about leakage and back-diffusion processes.

RESULTS AND DISCUTION

Table 1 presents the results for soil exhalation using the both charcoal adsorption and Lucas cell methods measured in an area from Cluj-Napoca, where preliminary high thoron concentration was found in the soil. As one can see from this table there is a good concordance between the results obtained using the two methods for different places of the sampling. The thoron flux is higher as radon flux in good agreement with other works (Crozier, 1969; Schery, 1986)

Table 2 shows the results of radon exhalation from seven different materials. The first sample in this table is Specimen 3, which was used in ERRICCA Intercomparisson Exercise from Athens (April 1999) where many laboratories from Europe have been participated. Our results obtained using charcoal method in two variants and the results obtained with Lucas cell and Radim methods are in very good concordance with the average value found for this sample by ERRICCA participants (Petropoulos, Anagnostakis and Simopoulos, 1999).

The sample No.2 and No.3 from Table 2 were prepared in our laboratory for verifying some models of radon migration in building materials. They are the concrete samples poured following standard technology but enhanced in ^{226}Ra on two manners. For preparing the sample No.2. a $RaCl_2$ solution was used and for the sample No.3 a fine powder of pitchblende (uranium ore) was melted with the cement used for obtaining the sample.

The results for two of these samples are quite different because for sample No.2 the radium atoms are fixed to surface of mineral grain (emanation coefficient being high) whereas for sample No.3 the radium atoms are uniformly distributed in all small mineral grains of pitchblende ore (Cosma, Dancea, Jurcut et al., 2000).

Table 1. Radon and thoron exhalation from soil in Cluj-Napoca area

Place	*Charcoal method (mBq/m^{2}s)*		*Lucas cell method (mBq/m^{2}s)*	
	Radon flux	*Thoron flux*	*Radon flux*	*Thoron flux*
A	25.5±2.4	2,180±150	27.1±1.8	1,980±130
B	36.1±3.5	3,350±210	37.4±2.5	3,060±170
C	30.7±3.5	2,430±180	32.2±2.1	2,230±140

Table 2. Exhalation radon rates from building material

No	Sample	Mass (kg)	^{226}Ra (Bq/kg)	Radon exhalation (Bq/kgh)	
				Charcoal	Lucas cell/Radim
1	Spec 3 ERRICCA	19	107 ± 5	0.262±0.022	0.252±0.015
2	Enhanced Ra	5.8	275 ± 10	0.845±0.035	0.810±0.024
3	Enhanced U	4.2	272 ± 5	0.067±0.004	0.059±0.004
4	Concrete BC-20	8.5	-	0.025±0.002	-
5	Bricks	3.2	-	0.014±0.002	-
6	Tills	5.9	-	0.016±0.002	-
7	BCA	4.5	-	0.059±0.003	0.051±0.003

The samples No. 4-7 are building materials used in Romania at this time. The emanation rates found by us range in the interval mentioned in the classical literature (Stranden,1988).

To test the possibility of simultaneously measurement of radon and thoron exhalation using charcoal adsorption method and gamma counting with NaI(Tl) or other detectors without high

resolution, a special sample of concrete containing 1520 Bq of ^{226}Ra and 4230 Bq of ^{224}Ra was prepared. ^{226}Ra precedes from the same $RaCl_2$ solution and for ^{224}Ra an amount of 2g of old $Th(NO_3)_24H_2O$ in which ^{224}Ra is in equilibrium with ^{232}Th was dissolved in the water used to prepare the concrete sample.

The thoron exhalation rate was determined by charcoal adsorption method obtaining 49 Bq/kgh and also by Lucas cell method obtaining a similar value of 44.5 Bq/kgh. The radon exhalation rate was similar with sample No.2 from Table 2.

Fig.5. The results of ERRICCA Intercomparison Exercise…

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