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A METHOD FOR THE MEASUREMENT OF THE EMANATION FACTOR FOR ^{222}Rn IN SMALL SAMPLES OF POROUS MATERIALS

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Abstract — A method to determine the emanation factor for ^{222}Rn in small samples of soils and building materials is proposed. The method is based on the principle of enclosing the sample to be tested in a hermetically sealed modified Lucas cell, designed and built in our laboratory, and subsequently measuring the radon concentration growth in the cell. The method has been tested for a total of 27 samples with different properties, including soils, building materials and phosphate fertilisers, which were previously evaluated for their physical properties and measured by gamma spectrometry to determine their radium activity concentrations.

INTRODUCTION

The measurement of the emanation factor for ^{222}Rn in porous materials is of great interest because it characterises the emanation of radon from the mineral grains to the pore system. It is a highly significant parameter which determines the amount of radon available for transport through the pores or cracks to the surface of the material. Many exhalation measurements reported in the literature show that porous materials with similar radium concentrations, porosities and diffusion coefficients have, however, very different total exhalation rates of ^{222}Rn because of the significant differences existing in their emanation factors(1).

The emanation factor for ^{222}Rn in a given porous material is usually determined from exhalation measurements made on a sample of the material under specific controlled conditions. Normally, the method used in these measurements involves enclosing the material sample to be tested in a container and, subsequently, deriving the total exhalation rate of the material from the activity concentration growth of radon inside the container, which, in turn, is measured by taking several air samples from the container up to 10 days(2). Nevertheless, this method has certain disadvantages. First, only a small number of measurements can be made because of the volume of the container, and so as not to disturb or dilute the radon concentration inside.

In fact, normally only one sample is collected under steady-state conditions which causes a delay of several weeks in the evaluation of the emanation factor. Secondly, removal of air from the container can induce changes in pressure. This can produce a convective transport of radon into the material, which modifies the gradient concentration in the sample and, hence, alters the total exhalation rate from the material(3). Finally, the third drawback is the relatively low sensitivity of the method with minimum measurable exhalation rates typically $10^{-3} - 10^{-4}$ Bq.s^{-1}.

DESCRIPTION OF THE METHOD

The method proposed here avoids these disadvantages and evaluates the emanation factor for ^{222}Rn in small samples of porous materials from the measurement of their total free exhalation rates. Essentially, the idea behind the method developed in our laboratory to measure the emanation factor is the same as that of the commonly named accumulation methods. The main innovation is that the container used for enclosing the material sample to be tested is a hermetically sealed modified Lucas cell. This cell, designed and built in our laboratory, has a capacity of 1 litre and its inside walls are lined with ZnS(Ag)-coated Mylar sheets. It is coupled to a photomultiplier and a standard counting system. It allows measurement of the activity concentration growth of the radon exhaled from the sample into the outer volume of the cell by directly counting the impulses due to the alphas from radon and its short lived daughters. The material sample to be studied is placed in a cylindrical PVC can of 7×10^{-2} litre capacity which is hung from the top of the cell as indicated in Figure 1. Assuming that the wall of the PVC can is non-absorbant, a one-dimensional theory is applicable to the analysis of the diffusion process of radon in the material sample. Taking into account that the outer volume of the cell is very large as compared to the sample volume, the total radon exhalation rate from the sample may be considered to remain constant in time and equal to the total free exhalation rate(4).

In these experimental conditions, a material sample with a total free exhalation rate E (Bq.s^{-1}) will produce an increase per unit time of the radon concentration, C (Bq.m^{-3}) in the cell

$$dC/dt = E/V - \lambda C \qquad (1)$$

where λ is the radioactive decay constant for ^{222}Rn and V is the outer volume of the cell in which radon is being exhaled. Obviously, V is the volume of the cell V_0 minus the sample volume V_s, but in our case $V = V_0$.

Assuming that E is constant in time and that initially $C(t = 0) = 0$, the solution of Equation 1 describing the radon concentration in the cell as a function of time is

$$C(t) = [E/(\lambda\ V)]\ (1 - \exp(-\ \lambda t)) \tag{2}$$

According to Equation 2, the total free exhalation rate E from the sample can be evaluated in two ways: either from the radon concentration measured at any time t, or, better, from the radon concentration growth in the cell during a given time interval by adjusting experimental data to a function specified as in the above equation.

In our experiment, if we solve the well-known differential equations that theoretically govern the production and decay of radon and its short-lived daughters and as we know the alpha efficiency of the cell for the geometry used, the radon concentration in the cell at any time t can easily be calculated from the net count rate obtained over a given counting time T after instant t. From these data, it is possible to construct the curve describing the radon activity concentration growth from which the total free exhalation rate can be derived. Nevertheless, this total free exhalation rate, E, can also be evaluated by integrating the one dimensional differential equation for radon diffusion into the sample. Thus, if radon is only exhaled from one surface of the sample, the total free exhalation rate, in $Bq.s^{-1}$, is given by the expression

$$E = \lambda\ \rho\ f\ S\ C_{Ra}\ L\ tgh(d/L) \tag{3}$$

where ρ is the bulk density of sample, in $kg.m^{-3}$; f the emanation factor; S the exhalation surface, in m^2; C_{Ra} the activity mass concentration of ^{226}Ra in the material, in $Bq.kg^{-1}$; L the diffusion length and d the thickness of the tested sample, both in m.

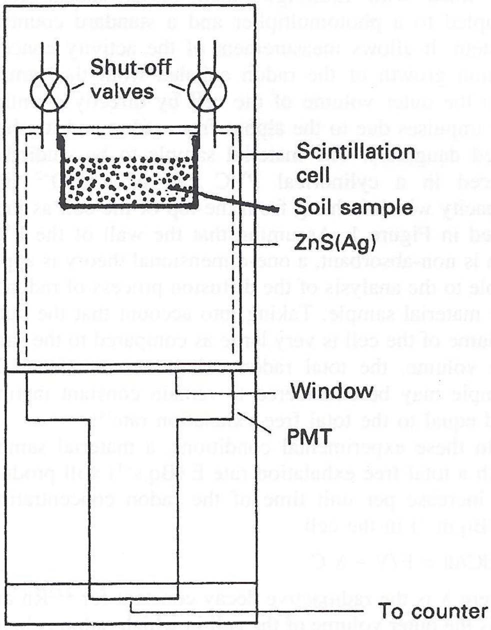

Figure 1. Schematic diagram of the experimental apparatus used to determine the emanation factor of the samples.

Equation 3 can be simplified by the approximation $tgh\ (d/L) = d/L$, if the diffusion length L is large compared to the sample thickness d. When this condition is fulfilled, as occurred in our experiment, the emanation factor can be determined through the expression

$$f = E/(\lambda\ C_{Ra}\ m) \tag{4}$$

where m is the mass of the sample. Once the activity mass concentration of ^{226}Ra in the sample is measured by gamma spectrometry and the total free exhalation rate is derived from the radon activity concentration growth in the outer volume of the cell, then, the emanation factor for ^{222}Rn in the material sample can be easily determined from Equation 4. Taking into account that the lower limit of detection for the radon concentration using the scintillation cell described above for counting times of 15 min, is typically $1.85 \times 10^{-2}\ Bq.l^{-1}$, the minimum total exhalation rate which can be measured by our method, from the steady-state radon concentration is of about $4 \times 10^{-8}\ Bq.s^{-1}$. For our experimental conditions this value corresponds to a free mass exhalation rate of $5 \times 10^{-7}\ Bq.kg^{-1}.s^{-1}$. Nevertheless, one of the advantages of the method proposed is, as described above, the comparatively short period for the measurements, in general less than 12 h. For this accumulation period, the minimum free mass exhalation rate which can be derived from the radon concentration growth in the cell is then $6 \times 10^{-6}\ Bq.kg^{-1}.s^{-1}$. Therefore, considering that on average the mass of the material sample enclosed in the cell is approximately 75 g and taking a

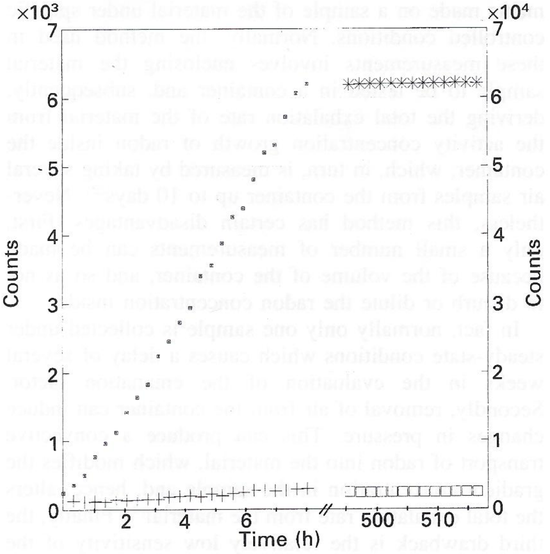

Figure 2. ^{222}Rn concentration growth in the cell from the samples analysed.

typical mean for the radium concentration in the porous material of 50 Bq.kg^{-1}, the minimum emanation factor for ^{222}Rn, which can be measured by the method proposed here, is of about 5×10^{-3} which practically cover the wide spectrum of values for the different existing porous materials.

RESULTS AND DISCUSSION

The technique described has been applied to measure the exhalation rate and the emanation factor for ^{222}Rn in a series of samples of soils, building materials and fertilisers. Table 1 summarises the results obtained, including the main parameters measured for each sample. Figure 2 shows a typical growth of the radon concentration inside the cell for two typical samples studied showing high and normal values of the radon exhalation rate. The curves fit very well with the theoretical Equation 2, and from them it is possible to derive the values of the parameters studied. With regard to the results for the emanation factor, a good agreement was found between the values obtained from the growth of the radon concentration over the first six hours and those obtained with the radon concentration value for the steady state conditions. However, when the total exhalation rate or the emanation factor was very low, the growth of radon concentration in the first hours does not correspond with the theoretical prediction. This was found to be related to the time required to place the sample into the cell in the laboratory, where the radon concentration was evaluated as 50 Bq.m^{-3}. The problem of this influence was solved by preparing the experiment in a radon free room.

In summary, the technique proposed not only solves the problems related to the existing methods but also makes it possible to evaluate easily and quickly the total exhalation rate and emanation factor for ^{222}Rn from porous materials.

Table 1. Emanation factor and exhalation rates of ^{222}Rn from the samples.

Material (No of samples)	Bulk density range (kg.m^{-3}) × 10^3	Porosity range	Radium content range (Bq.kg^{-1})	Exhalation range (Bq.kg^{-1}.s^{-1}) × 10^{-4}	Emanation factor range
Uraniferous soils (10)	1.1–1.3	0.45–0.53	10,000–12,000	930–2300	0.20–0.27
Cement (10)	1.1–1.4	0.50–0.55	40–70	0.03–0.05	0.02–0.03
Phosphate fertiliser (7)	0.7–0.9	0.40–0.50	400–500	1.1–1.20	0.15–0.30

REFERENCES

1. Wilkening, M. *Radon in the Environment.* (Amsterdam: Elsevier) (1990).
2. Damkjaer, A. and Korsbech, U. *Measurement of the Emanation of Rn-222 from Danish Soils.* Sci. Total Environ. **45**, 343–350 (1985).
3. Samuelson, C. and Petterson, H. *Exhalation of Tn-222 from Porous Materials.* Radiat. Prot. Dosim, **7**(1–4), 95–100 (1984).
4. Berkvens, P, Kerkhove, E. and Vanmarcke, H. *Three Dimensional Treatment of Steady State Rn-222 Diffusion in Building Materials: Introducing a Practical Modified One Dimensional Approach.* Health. Phys. **55**, 793–799 (1988).