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Thoron exhalation rate in stony materials: A simplified approach

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HIGHLIGHTS

- Theoretical method for the evaluation of the thoron exhalation rate from radon exhalation rate data.
- Test of the method by measuring the radon and thoron exhalation rate in a sienites sample by means of alpha spectrometry.
- Proposed formula as a screening tools for the estimation of the thoron exhalation rate in stony materials.

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ABSTRACT

A significant percentage of the thoron present in indoor environments is often attributable to building materials. It is therefore interesting to estimate the thoron exhalation rate from these materials. However, the detection of thoron is not a simple task: the very short half life of thoron and the lack of strong gamma emissions, prevents the use of the simplest and powerful radiometric techniques normally utilized in radon detection. Specific instruments have thus been developed in order to measure this radionuclide: alpha delayed coincidence counters (measuring the alphas emitted in a short sequence by ^{220}Rn and ^{216}Po) and alpha spectrometry are the most widely used instruments. However, a more simple approach, allowing the evaluation of the thoron (^{220}Rn) exhalation rate by measuring only the radon one (^{222}Rn), would be very welcomed. This goal can be achieved taking into account that a mathematical relationship between the thoron and radon exhalation rates J_{Th} and J_{Rn} (Bq/(m^2·s)) can be theoretically established. The thoron exhalation rate of any building material can thus be calculated by measuring only the corresponding radon exhalation rate, provided that the activity concentrations of the parents radionuclides (^{224}Ra and ^{226}Ra) are known. Operating in this way, it would be possible to obtain the thoron exhalation rates for the large majority of the building materials normally employed: in fact, the activity concentration data of many building materials are, in general, easily available. Following this approach a formula is proposed as a screening tools for stony materials.

1. Introduction

A significant percentage of the thoron (^{220}Rn) present in indoor environments is often attributable to building materials. It is therefore interesting to estimate the thoron exhalation rate from those materials. However, the detection of thoron is not a simple task: the very short half life of thoron and the lack of strong γ emissions prevents the use of the simplest and powerful radiometric techniques normally utilized for radon (^{222}Rn) detection. Specific instruments have thus been developed in order to measure this radionuclide: alpha delayed coincidence counters (measuring the alphas emitted in a short sequence by ^{220}Rn and ^{216}Po) and alpha spectrometry are the most widely used instruments [1–3].

Unfortunately, devices of this kind are not always available in all laboratories. More simple approaches would be therefore welcomed. In this paper is proposed a method allowing the evaluation of the thoron exhalation rate from the radon exhalation rate data, provided that the activity concentration of both parents (^{224}Ra and ^{226}Ra) inside the material (Bq/kg), are known.

Operating in this way, it would be possible to obtain the thoron exhalation rates for the large majority of the building materials normally employed: in fact, the activity concentration data of many building materials are, in general, more easily available.

2. Material and methods

2.1. Theoretical model

The production rate of radon and thoron inside a generic stony material and in any other kind of building material depends of course on the activity concentration of their parents, namely ^{226}Ra and ^{224}Ra. In particular, in a completely closed and

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isolated system, as can be considered the inner part of any rock and building material, if the activity concentrations c_{Ra226} and c_{Ra224} of ^{226}Ra and ^{224}Ra are expressed in Bq/kg, the specific production rate of radon and thoron (atoms/(s·kg)), r_{Rn} and r_{Th}, equals precisely the corresponding activity concentration values.

Once produced inside a rock, the radon and thoron atoms, being noble gases, are free to wander through the lattice of the material without any significant interaction with the matter. Therefore the movements of such atoms inside the lattice are governed by diffusion laws and can thus by modeled by the well known II order differential equation that, considering only one dimension, can be written as follows:

$$-D \cdot \frac{d^2C}{dx^2} + \lambda \cdot C = r \cdot \rho$$

where C in the radon (thoron) activity concentration per unit of volume (Bq/cm^3) in the material, D is the diffusion coefficient of the radon (or thoron) atoms inside the material, ρ is its density (g/cm^3), while λ and r are, respectively, the decay constants (s^{-1}) and the specific production rates (atoms/(s·kg)) of the radon radioisotopes.

The general solution of this non homogeneous II order differential equation is given by the following expression:

$$C(x) = A \cdot e^{-\sqrt{\frac{\lambda}{D}}x} + B \cdot e^{\sqrt{\frac{\lambda}{D}}x} + r \cdot \rho \cdot \left(1 - e^{-\sqrt{\frac{\lambda}{D}}x}\right) \quad (1)$$

where A and B are arbitrary constants, to be determined by the boundary conditions of the physical system.

A very simplified model of the system is proposed, the schematic representation of which is reported in Fig. 1: the radon (thoron) emitting material is described as a slab in which the only relevant dimension is its thickness d. Moreover, is made the apparently rough assumption that the exhalation process occurs only on the two largest surfaces of the slab, being the areas of the others negligible. The positions of the radon (or thoron) atoms wandering inside the slab are therefore completely individuated by the x coordinate.

The emission of radon and thoron from the two surfaces of the slab can be treated separately and in the same way by means of a simple change of the reference system (x coordinate): the exhalation is thus assumed to occur in a symmetric way on both sides of the slab.If is made the further assumption that the thickness d of the slab largely exceeds the typical diffusion length $\ell = \sqrt{D/\lambda}$ of the radon and thoron atoms in the material, ($\ell \ll d$), the following boundary conditions can be established:

For $x = 0$, i.e., at the surface emission of the slab, the boundary condition can be expressed as: $C(x = 0) = C_0$, where C_0 is the volumetric activity concentration of radon (thoron) at the interface slab-atmosphere.The boundary condition for $x = d$, being $\ell = \sqrt{D/\lambda} \ll d$, is equivalent to consider the behavior of $C(x)$ as $x \to +\infty$: As a consequence of that, the constant B in Eq. (1) must be set equal to zero, $B = 0$, in order to prevent the divergence of the expression. Therefore, the following solution is obtained:

$$C(x) = C_0 \cdot e^{-\sqrt{\frac{\lambda}{D}}x} + r \cdot \rho \cdot (1 - e^{-\sqrt{\frac{\lambda}{D}}x}) \quad (2)$$

that allows the calculation of the profile of the radon and thoron activity concentration inside the material. The applicability of Eq. (2) in real cases is briefly discussed in the following. It must be beared in mind that this expression was derived considering the condition $\ell \ll d$, that is certainly always true for thoron, due to the very low values of its diffusion length in all materials, but not necessarily for radon: in fact, the longer half-life of radon can lead to a value for the diffusion length ℓ that is sometimes of the same order of magnitude or even greater than the typical thickness d of most building materials (d = 2–10 cm).

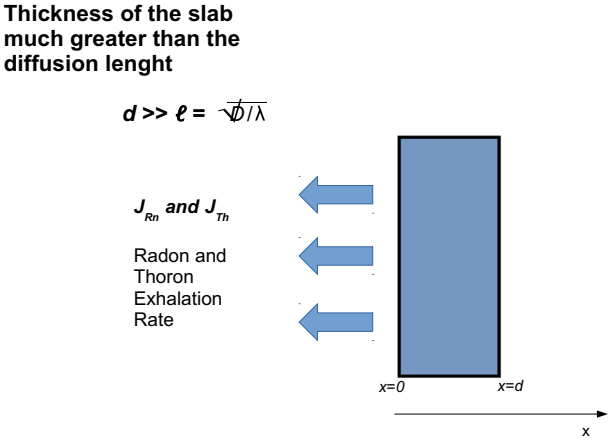

Fig.1. Radon and thoron exhalation from a slab with the condition d ≫ ℓ: the exhalation process can be modeled considering only one coordinate, x.

It can be easily seen from Eq. (2), that the asymptotic value of $C(x)$, i.e., the activity concentration $C(x \to +\infty)$ deeply inside the slab, tends to a value corresponding to $r \cdot \rho$, the product of the production rate r and the density of the material. As far as the value of C_0 is concerned, it can be guessed that, being at the interface material-atmosphere, it should be substantially lower than the asymptotic concentration in the inner part of the material, because of the losses due to the direct radon and thoron exhalation from the surface: one can imagine that about one half of the ^{222}Rn and ^{220}Rn atoms produced near the surface enters immediately into the free atmosphere due to the alpha decay recoil. It's not important, however, to know the exact value of C_0: it's enough to assume that a simple dependence on the corresponding production rate r holds:

$$C_0 = \alpha \cdot r \cdot \rho$$

in which α is an a-dimensional parameter, mainly depending on the geometry of the problem and whose value must be less than 1. Putting this expression in Eq. (2), gives for $C(x)$ the following:

$$C(x) = r \cdot \rho - r \cdot \rho \cdot e^{-\sqrt{\frac{\lambda}{D}}x} \cdot (1 - \alpha) \quad (3)$$

in which the activity concentrations of radon and thoron are expressed as a function of the corresponding activity concentrations ($r \cdot \rho$) of the parents radionuclides.

Thus, the radon (thoron) flux J (Bq/(m^2·s) at the interface slab-atmosphere (x = 0) can now be evaluated simply applying the Fick's law:

$$J = -D \cdot \frac{dC}{dx}$$

and calculating this expression for $x = 0$. The activity concentration function $C(x)$ in the above equation is given by Eq. (3). Therefore, the final expression for J is obtained:

$$J = -\sqrt{\lambda D} \cdot r \cdot \rho \cdot (1 - \alpha) \quad (4)$$

Considering Eq. (4) for the radon and thoron flux J_{Rn} and J_{Th} and with the reasonable assumption that $D_{Rn} \approx D_{Th}$, the following expression can be written:

$$J_{Th} = J_{Rn} \frac{C_{Ra224}}{C_{Ra226}} \cdot \sqrt{\frac{\lambda_{Rn220}}{\lambda_{Rn222}}} \quad (5)$$

where c_{Ra224} and c_{Ra226} are, respectively, the mass activity concentrations (Bq/kg) of ^{224}Ra and ^{226}Ra in the material, while λ_{Rn220} and λ_{Rn222} are the thoron and radon decay constants.

2.2. Experimental procedures

The experimental set up for the radon exhalation measurements is reported in Fig. 2.

The sample to be tested is put inside a radon-tight chamber, i.e., a barrel, the volume of which V is approximately 19L, carefully sealed in order to reduce any significant radon leakage during the experiment. An electric fan assures an accurate mixing of the air, improving the homogenization of the activity concentrations inside the barrel. A radon-thoron detector, registers simultaneously the activity concentration levels of both radioisotopes every 60 min.

Following the theoretical description given by [4], the ingrowth of the radon and thoron activity concentration (Bq/m^3) inside the chamber can be described by the equation:

$$C_{exp}(t) = \frac{e \cdot \lambda}{V \cdot (\lambda + \lambda^*)} \left(1 - e^{-(\lambda+\lambda^*)\cdot t}\right) \quad (6)$$

where e is atomic concentration production rate (atoms/s) inside the chamber and λ* is a parameter that accounts for a possible radon leakage and for the so called "back-diffusion effect".[1] The radon and thoron flux J (Bq/m^2·s) emerging from the material can thus be calculated from the experimental data by means of the following expression:

$$J = C_{exp}(t \to +\infty) \frac{V \cdot (\lambda + \lambda*)}{S} \quad (7)$$

in which $C_{exp}(t \to +\infty)$ is the asymptotic value of Eq. (6) and S is the emitting surface of the material. The results obtained with this expression should be compared with the theoretical predictions given by Eqs. (4) and (5).

In the experiment the Thoron-Scout monitor, developed by the SARAD® company [5], was used as radon/thoron detector. In this instrument, both radon and thoron are simultaneously measured via its short lived daughters, namely ^{218}Po and ^{214}Po for ^{222}Rn and ^{216}Po for ^{220}Rn. The short lived daughters, once produced,

[1] The term backdiffusion should be avoided as backdiffusion doesn't exist as a physical phenomenon, being just a slowing down of the diffusion process due to the increasing level of the radon in the container: however is still used in this paper for practical reason.